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The influence of heavy metals on the formation of organics and HCl during incinerating of PVC-containing waste

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Abstract

The heavy metal, organic pollutants and HCl emissions from incineration pose a grave threat to human health. However, submicro metal particles and organics in gas phase are removed with difficulty by conventional air pollution control apparatus, and could be released into the atmosphere along with the flue gas. This research studied the formation of organics and HCl (hydrogen chloride) during incineration when the feedstock contained different amounts of PVC and heavy metals. The experiments were performed in a bubbling fluidized bed incinerator with two cyclones and scrubbers. Experimental results indicated that the greatest amount of organic compounds produced were aromatic compounds. The amount of organic compounds and HCl increased when the feedstock contained PVC (polyvinyl chloride), and decreased when the feedstock contained heavy metals. © 1998 Elsevier Science B.V. All rights reserved.

Keywords: Incineration; Heavy metals; HCl; Aromatic compounds

1. Introduction

The composition of waste is very complex. Many types of pollutants are produced during the incineration of solid waste, such as polycyclic aromatic hydrocarbons [1] and heavy metals [2,3]. Controlling the emission of pollutants is a critical parameter in incinerator design.

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Many factors influence the generation of organic pollutants during incineration.

(1) Temperature: low-cyclic aromatic hydrocarbons are formed easily at low temperature (400–500°C). High-cyclic aromatic compounds are formed easily at high temperatures, such as 5 to 6 cyclic PAHs produced at 700-900°C [4].

(2) The composition of wastes: organic wastes contaminated by benzene can promote the formation of PAHs during incineration [5]. PVC or chlorine can promote long-chain alkenes cyclization through dechlorine reactions, and generate aromatic organics, chloro-aromatic hydrocarbons or char [6]. Under oxygen-deficit conditions, metal chlorides promote the thermal decomposition of polystyrene plastic, and subsequently react with benzene, which ultimately favors the formation and growth of PAHs [4]. Metals in waste catalyze or inhibit the formation of PAHs at high temperatures [7,8].

(3) The quantity of air and retention time affect the combustibility of waste. Under oxygen deficit conditions, wastes are incompletely combusted. When oxygenated during combustion, the incineration process temperature is reduced and retention time is decreased [9].

(4) The air pollution control apparatus and properties of pollutants.

All pollutants from incineration process are not released into the air. Some pollutants will condense onto fly ash in the flue gas pipeline and can be removed by ESP or baghouse. The distribution characteristics of pollutants in fly ash relates to the specific surface area of the fly ash, the amounts of carbon in the fly ash, and the specific properties of the pollutants [10]. Griest (1986) pointed out that the affinity of aromatic compounds for fly ash is higher than that for aliphatic compounds. The higher-cyclic compounds have a greater π bond affinity [11].

Heavy metals volatilize at higher temperatures during incineration and form sub-micron particulates by condense in the flue gas pipeline [12]. The high incineration efficiency increased the proportions of a smaller size of fly ash [13]. A high HCl concentration in the gas stream can increase the volatilization of heavy metals and induce the formation of metal chlorides [14]. The concentration of HCl in urban waste combustion gases is of the order of 10^2-10^3 mg/N m³. An increase in the feed chlorine content markedly increases the volatility of metals. The presence of chlorine appears to shift the particles to a finer distribution [15].

The present work was performed to determine the relationship of the heavy metals to the formation of organics and HCl during incinerating the PVC-containing waste.

2. Experimental

2.1. Preparation of samples

The composition of feedstock used in the experiment contained sawdust, high-density polyethylene (HDPE), polyvinyl chloride (PVC), water, and three classic heavy metals. Each sample of mixed feedstock was enclosed in a small PE plastic bag. The total weight of each sample was 3.4 g. Table 1 lists the composition of the feedstock and the operating conditions.

No	Sawdust	Feedstoc	Feedstock (g/bag)			Temperature (°C)		
		HDPE	PVC	Metal		Sand bed	Secondary chamber	
Run 1	1.1	1.0	0.5	_	0.3	700	800	
Run 2	1.1	1.5	_	_	0.3	700	800	
Run 3	1.1	0.5	1.0	_	0.3	700	800	
Run 4	1.1	1.0	0.5	0.018 Pb	0.3	700	800	
Run 5	1.1	1.0	0.5	0.018 Cr	0.3	700	800	
Run 6	1.1	1.0	0.5	0.018 Cd	0.3	700	800	
Run 7	1.1	1.0	0.5	0.054 mix	0.3	700	800	

Table 1 The composition of feedstock and operating conditions

2.2. Experimental procedure

The apparatus used in this study is shown in Fig. 1. The reactor was a fluidized bed incinerator consisting of a stock feeder, a preheated chamber 50 cm long, a major combustion chamber with 110 cm height and 10 cm inner diameter, and a secondary combustion chamber with 100 cm height and 25 cm inner diameter. The incinerator was equipped with a perforated stainless steel gas distributor. Six thermocouples were employed to determine the temperature profile in the preheated chamber, sand bed and freeboard chamber. The combustion gas was treated with cyclones and wet scrubbers. PLC (program logical control) was used to record the data and display it on a monitor. Therefore, the stability of incineration system can be adequately controlled. By theoretically calculating the required air, the proper air ratio required for combustion was



Fig. 1. The fluidized bed incinerator. (1) blower, (2) flow meter, (3) air preheated chamber, (4) air distributor, (5) first combustion chamber (sand bed zone), (6) 2nd combustion chamber (freeboard) 7. thermo-couples, (8) cooling system for feeding, (9) cyclones, (10) scrubbers, (11) I.D. fan, (12) burner.

determined, and the excess air ratio was around 30%. The main combustion chamber had to be initially heated electrically so that the sand bed zone could be preheated to the desired temperature. A gas burner was then used to aid the secondary combustion chamber in reaching the desired temperature. The temperature of the sand bed was 700°C. The temperature of the secondary chamber was 800°C.

2.3. Sampling and analysis of organics

Samples were taken from the outlet duct after being processed in the cyclones. Isokinetic sampling was performed. The sampling apparatus is shown in Fig. 2, which refers to USEPA modified method 5 (MM5). Flue gas was sampled during the 60-min incineration process using a stainless sampling probe. The flue gas was passed through a heated filter packed with glass fiber, which removed fine particles escaping from the cyclones. The flue gas was then passed through a cooling tube to allow capture of the remaining organics using XAD-2 resin.

The organic-containing samples were initially extracted for 20 h using a Soxhlet extraction process and condensed down to 9 ml using a KD evaporative concentrator. A total of 3 ml of the condensed solution was concentrated down to 1 ml by purging it with pure N_2 , and analyzed for C_6-C_9 organic compounds. The remaining solution was



Fig. 2. The sampling train for organics. (1) sampling probe, (2) heated filter, (3) cooling tube and XAD-2 adsorption tube, (4) Impingers, (5) flow meter, (6) connect of vacuum pump.

passed through a purifying tube packed with silica gel and aluminum oxide. $C_{10}-C_{22}$ organic compounds were extracted from this solution using a purifying suspension. Furthermore, the purified solution was condensed to 2 ml using a KD evaporative concentrator and purged with the pure N₂ to yield a quantity of 1 ml. Finally, the samples were stored at 4°C. These samples were analyzed with GC/FID. The column was CP-SIL 8 CB Cat. No. 8751 (30 m × 0.25 mm i.d., 0.25 μ m film).

2.4. Sampling and analysis of metals

The same sampling location used for organics was used to sample metals as well. The sampling apparatus is shown in Fig. 3, which refers to USEPA method 5 (M5). The gas phase metallic compounds in the flue gas were sampled during a 30-min incineration process. The flue gas was sampled using a stainless sampling probe and passed through a heated filter packed with glass fiber, which removed the fine particles. The flue gas was then passed through impingers with a mixed adsorption solution of 200 ml 10% H_2O_2 and 5% HNO₃, which captured the metals through absorption. In order to enhance condensations, the impingers were submerged in an ice bath.

The metal samples included fly ash, fiber filter and absorption solution. They were pretreated with microwave digestion and the recovery efficiency was measured. The metal concentration was analyzed using atomic adsorption spectroscopy (AAS) according to the USEPA method 3050.



Fig. 3. The sampling train for heavy metals and HCl. (1) sampling probe, (2) heated filter, (3) Impingers, (4) flow meter, (5) connect to vacuum pump.

2.5. Sampling and analysis of HCl

The HCl in the flue gas was sampled during a 20-min incineration process. The same sampling method for metals was used to sample HCl as well. Impingers with 100 ml adsorption solution of 0.1 N NaOH were used to capture the HCl through absorption. In order to enhance condensations, the impingers were submerged in an ice bath. The HCl analysis was performed by titration of the solution with a mercuric nitrate solution.

3. Results and discussion

Compounds of more than six carbons are of particular interest, because these organics are more poisonous than lower molecular weight compounds, particularly in the case of aromatic compounds. Experimental results indicate that the particular species of organics formed during incineration were aromatic compounds and their isomers, less aliphatic compounds. The organic chlorides produced were chloro-aromatic compounds when the feedstock contained PVC.

Tables 2–5 list the concentrations of organics in the flue gas when the feedstock contained different amount of PVC. The cyclohexenes had the highest concentration in pollutants, 3200 μ g/N m³. The concentration of benzene was about 1000 μ g/N m³, toluene was 550 μ g/N m³, naphthalene was 400 μ g/N m³, dibutyl phthalate was 300 μ g/N m³, and others were less than 150 μ g/N m³.

When the feedstock contained PVC, the concentration of cyclohexene was reduced. However, other compounds increased. The concentration of single-cyclic and two-cyclic aromatic compounds increased when the amount of PVC was increased. Polycyclic and chloro-aromatic compounds, however, were not altered. Some aliphatic compounds present in the flue gas when the feedstock had no PVC, such as eicosane, heneicosane,

Table 2

Organic species	No PVC	0.5 g PVC	1.0 g PVC	
Benzene	332.4	1098.2	1248.7	
Cyclohexene	3604.8	3259.6	3228.1	
Toluene	364.6	575.3	697.2	
<i>m</i> -Xylene	40.4	57.7	81.5	
Phenylethyne	ND	95.5	72.6	
Styrene	18.7	69.4	146.5	
Benzaldehyde	24.3	69.7	143.2	
Benzofuran	ND	60.5	80.5	
Indene	ND	42.3	19.3	
1-methyl-2-(2-propenyl)-benzene	49.7	50.1	54.8	
Azulene	18.5	40.2	24.5	
1,3,5-triethyl-benzene	ND	40.2	ND	

The concentration of single-cyclic compounds in gas phase when feedstock contained different amount of PVC (μ g/N m³)

Table 3

The concentration of two-cyclic compounds in gas phase when feedstock contained different amount of PVC ($\mu g/N m^3$)

Organic species	No PVC	0.5 g PVC	1.0 g PVC	
Naphthalene	70.5	356.6	516.3	
2-Ethenyl-naphthalene	ND	33.1	44.4	
Biphenyl	ND	33.3	71.8	

Table 4

The concentration of polycyclic compounds in gas phase when feedstock contained different amount of PVC ($\mu g/N m^3$)

Organic species	No PVC	0.5 g PVC	1.0 g PVC	
Acenaphthylene	28.9	31.0	ND	
Dibenzofuran	ND	28.6	38.0	
Fluorene	14.7	22.5	28.9	
9-H-Fluoren-9-one	ND	51.7	26.8	
Phenanthrene	ND	82.8	62.4	
Anthracene	ND	29.5	ND	
Dibutyl phthalate	192.7	310.3	377.2	
Fluoranthene	ND	61.8	24.6	
Pyrene	ND	29.9	34.5	
Benzo(a)anthracene	ND	28.2	48.7	
Benzo(b)fluoranthene	36.6	129.8	84.8	
Benzo(a)pyrene	23.4	48.8	ND	
Indeno(1,2,3-cd)pyrene	20.0	24.6	22.8	

docosane, tricosane, and tetracosane. These results indicated that chlorine promoted the cyclization of organics and the formation of aromatic compounds during incineration. The reasons: (1) The electronegativity of chlorine was higher than carbon and hydrogen bonded to chlorine easily. Thus, the unstable alkynes and alkene organics were formed from straight chain and branch chain aliphatic compounds. Most organics were destroyed and formed CO_2 and H_2O ; however, some of them formed more stable

Table 5

The concentration of chloro-aromatic compounds in gas phase when feedstock contained different amount of PVC ($\mu g/N m^3$)

Organic species	No PVC	0.5 g PVC	1.0 g PVC	
Chloro-benzene	ND	84.44	115.58	
2-chloro-phenol	ND	63.36	54.74	
Benzyl chloride	ND	31.03	29.52	
1-Chloro-2-methyl-benzene	ND	31.64	55.30	
1,2-Dichloro-benzene	ND	53.12	59.42	
1,2,3-Trichloro-benzene	ND	95.33	ND	

Table 6

Organic species	No metal	Pb	Cr	Cd	Mixing
Benzene	1098.2	1108.5	994.5	870.4	906.1
Cyclohexene	3259.6	3520.5	3486.6	3002.7	3151.9
Toluene	575.3	732.1	758.6	388.6	449.7
<i>m</i> -Xylene	57.7	65.2	73.1	75.3	69.0
Phenylethyne	95.5	92.2	80.3	70.4	73.3
Styrene	69.4	60.4	44.2	37.6	48.5
Benzaldehyde	69.7	53.1	41.0	33.7	37.0
Benzofuran	60.5	44.0	36.3	24.8	35.3
Indene	42.3	ND	ND	ND	ND
1-Methyl-2-(2-propenyl)-benzene	50.1	59.5	53.6	69.9	57.6
Azulene	40.2	ND	ND	38.6	26.6
1,3,5-triethyl-benzene	40.2	37.0	36.5	43.1	39.6

The concentration of single-cyclic compounds in gas phase when feedstock contained different heavy metals ($\mu g/N m^3$)

compounds at high temperatures, such as aromatic compounds. (2) PVC or chlorine can promote long-chain alkenes cyclization through a dechlorine reaction, and generate aromatic compounds, chloro-aromatic hydrocarbons or char.

Tables 6-9 list the concentrations of organics in flue gas when the feedstock contained different heavy metals. The analysis results indicated that heavy metals influenced the concentration of organics in the gas phase. Interalios, Pb and Cr increased the concentrations of cyclohexene, toluene, *m*-xylene, benzo(*a*)anthracene, and benzo(*b*)fluoranthene in flue gas. The concentration of most organics was reduced when the feedstock contained heavy metals, especially Cd. This illustrated that heavy metals reduced the concentration of organics during incineration. The concentration of chlorine was reduced when heavy metals reacted with it to form metal chloride, thus reduced the opportunity for chlorine to react with organics. Heavy metals can inhibit the formation of chloro-organics at a high temperature.

Tables 10 and 11 list the amount of organics adsorbed on fly ash in the cyclones. The concentration of two-cyclic and chloro-aromatic compounds in fly ash is too low to detect. The experimental results indicated that the polycyclic compounds, with a higher molecular weight and boiling point, were adsorbed on fly ash favorably. The major single-cyclic organics absorbed on fly ash were benzene and cyclohexene. Because the

Table 7

The concentration of two-cyclic compounds in gas phase when feedstock contained different heavy metals ($\mu g/N \ m^3)$

Organic species	No metal	Pb	Cr	Cd	Mixing
Naphthalene	356.6	312.8	297.5	239.0	275.3
2-Ethenyl-naphthalene	33.1	31.9	15.7	20.0	29.7
Biphenyl	33.3	31.8	27.1	ND	25.8

Table 8

Organic species	No metal	Pb	Cr	Cd	Mixing	
Acenaphthylene	31.0	35.8	31.5	34.7	27.2	
Dibenzofuran	28.6	ND	ND	ND	32.5	
Fluorene	22.5	22.3	19.8	ND	ND	
9-H-Fluoren-9-one	51.7	23.9	20.2	ND	51.3	
Phenanthrene	82.8	57.8	64.1	47.9	79.2	
Anthracene	29.5	27.9	29.8	23.8	29.9	
Dibutyl phthalate	310.3	164.4	94.4	124.1	106.5	
Fluoranthene	61.8	23.4	26.9	28.3	48.4	
Pyrene	29.9	22.9	21.5	18.0	30.3	
Benzo(a)anthracene	28.2	80.1	52.2	ND	63.7	
Benzo(b)fluoranthene	129.8	263.1	189.2	121.7	106.7	
Benzo(a)pyrene	48.9	44.8	31.1	28.8	42.5	
Indeno(1,2,3-cd)pyrene	24.6	23.7	31.8	ND	22.8	

The concentration	of	polycyclic	compounds	in	gas	phase	when	feedstock	contained	different	heavy	metals
$(\mu g/N m^3)$												

Table 9

The concentration of chloro-aromatic compounds in gas phase when feedstock contained different heavy metals ($\mu g/Nm^3$)

Organic species	No metal	Pb	Cr	Cd	Mixing	
Chloro-benzene	84.4	70.1	59.1	35.6	43.0	
2-Chloro-phenol	63.4	50.7	26.7	24.6	35.4	
Benzyl chloride	31.0	29.2	16.7	17.2	20.5	
1-Chloro-2-methyl-benzene	31.6	ND	ND	ND	ND	
1,2-Dichloro-benzene	53.1	50.4	42.2	25.5	33.8	
1,2,3-Trichloro-benzene	95.3	67.8	50.4	21.5	25.7	

Table 10

The concentration of single-cyclic compounds adsorbed on fly ash when feedstock contained different heavy metals ($\mu g/g$)

Organic species	No metal	Pb	Cr	Cd	Mixing
Benzene	264.7	271.3	267.4	989.2	636.3
Cyclohexene	870.0	849.8	874.1	379.1	195.6
Toluene	7.4	12.1	8.9	13.4	10.9
<i>m</i> -Xylene	31.6	21.1	12.8	23.1	26.5
Phenylethyne	ND	ND	ND	ND	ND
Styrene	15.4	26.4	21.7	25.0	20.5
Benzaldehyde	ND	ND	ND	ND	ND
Benzofuran	ND	ND	ND	ND	ND
Indene	ND	ND	ND	ND	ND
1-Methyl-2-(2-propenyl)-benzene	ND	ND	ND	ND	ND
Azulene	ND	ND	ND	ND	ND
1,3,5-Triethyl-benzene	ND	ND	ND	ND	ND

Organic species	No metal	Pb	Cr	Cd	Mixing	
Acenanhthylene	73	73	7.0	6.0	75	
Dibenzofuran	66	ND	ND	ND	ND	
Fluorene	6.9	9.1	90	14.5	10.2	
9- <i>H</i> -Fluoren-9-one	9.0	15.1	6.8	11.4	6.4	
Phenanthrene	20.6	39.8	27.0	21.6	23.7	
Anthracene	12.6	18.8	18.6	13.2	15.8	
Dibutylphthalate	35.5	39.7	20.0	26.5	20.2	
Fluoranthene	11.8	17.3	10.8	12.7	10.7	
Pyrene	8.3	20.7	48.1	52.7	47.9	
Benzo(a)anthracene	9.7	70.9	53.1	55.8	56.2	
Benzo(b)fluoranthene	11.2	54.0	48.8	54.6	51.7	
Benzo(a)pyrene	10.0	11.8	10.4	9.9	9.8	
Indeno(1,2,3-cd)pyrene	8.0	6.2	ND	ND	ND	

The concentration of polycyclic compounds adsorbed on fly ash when feedstock contained different heavy metals ($\mu g/g$)

Table 12 The concentration of HCl (ppm) in flue gas at different operating conditions

Class of metal in feed stock	Amount of PVC in feed stock (g)	Percentage of water in flue gas	Concentration of HCl flue gas (ppm)
No	0.5	7.21%	685.2
Pb	0.5	7.74%	620.4
Cr	0.5	7.78%	684.3
Cd	0.5	7.58%	598.1
Mixing	0.5	7.30%	520.3
No	1.0	7.68%	1402.0

Table 13 The concentration of heavy metals in gas phase and fly ash at different operating conditions

Temperature of adsorption	Concentration of metal in gas phase (mg/N m ³)				
	Pb	Cd	Cr		
120°C (mix)	0.234	0.068	0.019		
150°C (mix)	0.269	0.056	0.024		
180°C (mix)	0.235	0.042	0.020		
210°C (mix)	0.295	0.063	0.023		
150°C (Pb)	0.239	_	0.024		
150°C (Pb)	_	0.045	_		
Glass fiber filter	5.70	6.06	0.05		
Concentration of metal in fly ash					
(mg / g)					
Fly ash	964.5	1087.9	202.5		

concentrations of these two organics were higher than others, they have a greater opportunity to be adsorbed on fly ash. The heavy metals in fly ash enhance the adsorption of organics.

Table 12 displays the concentration of HCl in flue gas under various operating conditions. The concentration of HCl was 685 ppm when the feedstock had no heavy metals, and it was reduced to 520 ppm with the addition of metals. The metals reacted with chlorine to form metal chlorides at high temperatures and reduced the amount of chlorine available to react with hydrogen to form HCl. The concentration of HCl increased when the amount of PVC was increased.

Table 13 lists the concentration distribution of heavy metals in the gas phase, heated filter, and fly ash. The concentration of Pb exceeded Cd and Cr in the gas phase, and the concentration of Pb and Cd exceeded Cr on the heated filter and in fly ash. Pb and Cd react with chlorine to form volatile metal chlorides at high temperatures. The Cr formed metal oxide with a high boiling point and volatilized with difficulty.

4. Conclusion

(1) PVC or chlorine present in waste promote the cyclization of organics and formed aromatic compounds during incineration, especially in low-cyclic compounds.

(2) The presence of Cd leads to the reduction of the amount of aromatic compounds and HCl in flue gas.

(3) Low-cyclic compounds with a particularly low boiling point are present primarily in the gas phase. Polycyclic compounds are easily adsorbed by fly ash particles. The heavy metals in fly ash enhance the adsorption of organic compounds.

(4) The quantity of heavy metals concentrated in the gas phase follow Pb > Cd > Cr. Pb and Cd have a higher concentration in the heated filter and fly ash than Cr. Because Pb and Cd react with chlorine to form volatile metal chlorides at high temperatures, and Cr formed metal oxide with a high boiling point.

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