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Simultaneous control of acid gases and PAHs using a spray dryer combined with a fabric filter using different additives

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Abstract

The purpose of this research was to simultaneously evaluate the removal efficiency of acid gases and PAHs from the flue gas emitted by a laboratory incinerator. This flue gas contained dust, acid gases, organics and heavy metals. A spray dryer combined with a fabric filter was used as the air pollution control device (APCD) in this study. The operating conditions investigated included different feedstock additives (polyvinyl chloride (PVC) and NaCl) and spray dryer additives (SiO_2, $CaCl_2$ and $NaHCO_3$).

The removal efficiency for SO_2 could be enhanced by adding inorganic additives, such as SiO_2, $CaCl_2$ and $NaHCO_3$. The presence of PVC in the incinerator feedstock also increased the removal efficiency of SO_2in the spray dryer. The improved removal of PAHs could be attributed to the addition of feedstock additives (PVC and NaCl) and spray dryer additives (SiO_2, $CaCl_2$ and $NaHCO_3$).

Keywords: Acid gases; PAHs; Spray dryer; Fabric filter; Additives

1. Introduction

A spray dryer combined with a fabric filter are the common air pollution control devices (APCDs) for incinerators. The effect of inlet concentration of acid gases, relative humidity, species of acid gases, reaction temperature and various additives on the removal efficiency of SO_2 using the spray dryer has been carried out in earlier studies by others [1–3]. Ruiz-Alsop and Rochelle [4] investigated three groups of additives including buffer acids (adipic and glycolic), organic deliquescents (EG, TEG and MEA) and inorganic deliquescents ($NaSO_4$,

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$CaCl_2$, $NaSO_3$, NaCl, $Na_2S_2O_3$ and $NaNO_3$). Their results showed that the inorganic deliquescents were the only additives that could increase the reactivity of $Ca(OH)_2$ for SO_2 since they could retain a greater amount of water on the absorbent surface. Cunill et al. [5] and Izquierdo et al. [6] indicated that inorganic deliquescents played two major roles during desulfurization process. One effect was to enhance the deliquescence of $Ca(OH)_2$. The other was to act as an alkalinity sorbent like NaOH, $NaHCO_3$ and $NaCO_3$. Moreover, previous studies indicated that relative humidity, as well, played an important role in SO_2 capture [5–8]. The removal efficiency of SO_2 increased with increasing relative humidity.

Organic compounds, such as PAHs, BTEX and PCDDs/PCDFs have received considerable attention due to their mutagenic and carcinogenic properties. The physical and chemical properties of the organic compounds in the flue gas are related to the combustion temperature, waste composition, excess air and type of APCD employed [9,10]. Since PAHs have high boiling points and low vapor pressure, they tend to migrate to the condensed phase via nucleation, condensation and adsorption. Previous studies showed that the amount of PAHs adsorbed on fly ash were affected by the specific surface area, particle size, heavy metals and the amount of carbon in the fly ash [11–15]. Lee et al. [11] pointed out that PAHs adsorbed by fly ash were proportional to the amount of carbon. Wey et al. [13–15] indicated that heavy metals (Cr and Cd) in the fly ash would provide activity sites required for the reactions between the lower-ring PAHs and PAHs precursors and promote them to form higher-ring PAHs.

A spray dryer combined with a fabric filter has been widely used to remove acid gases emitted by municipal wastes incinerators. However, the effects of additives on the removal efficiency of acid gases and PAHs are rarely discussed. Because organic volatiles adsorb on the fly ash and the fabric filter is very efficient in removing fly ash, it is possible to simultaneously remove acid gases and PAHs from an incinerator flue gas containing fly ash, acid gases, organic compounds and heavy metals. Here, a spray dryer combined with a fabric filter is used as the APCD in this study.

2. Experimental

2.1. Preparation of simulated feedstocks

The simulated incinerator feedstocks used in the experiments were composed of sawdust, plastic, sulfur and a heavy metal solution. Five metals (Cd, Pb, Cu, Cr and Zn) were dissolved in distilled water containing 0.5 wt.% nitrate. These feed materials were enclosed in a polyethylene (PE) bag. The total weight of each bag was 3.5 g. The compositions of the simulated feedstocks and operating parameters are shown in Table 1.

2.2. Apparatus

The reactor for this experiment was a laboratory-scale fluidized bed incinerator equipped with a spray dryer and a fabric filter as the air pollution control system (Fig. 1). The height and inner diameter of the spray dryer were 885 and 165 mm, respectively. Nomex was the filter (Nomex is a registered trademark of E.I. Du Pont De Nemous & Co., USA). Its area was 3.39 m^2. Silica sand (200 g) was used as the fluidized bed material; a thermocouple

Table 1
Feedstock compositions and operating parameters for each test of the air pollution control system for incinerator gases

Test (run)	Operating parameter	Feedstock composition (g per bag)						
	Absorbent in spray dryer[a]	Sawdust	PE	PVC	NaCl	Sulfur	Water[b]	PE bag
1	$Ca(OH)_2$	1.5	0.6	–	–	0.1	1.0	0.3
2	$Ca(OH)_2 + SiO_2$	1.5	0.6	–	–	0.1	1.0	0.3
3	$Ca(OH)_2 + CaCl_2$	1.5	0.6	–	–	0.1	1.0	0.3
4	$Ca(OH)_2 + NaHCO_3$	1.5	0.6	–	–	0.1	1.0	0.3
5	$Ca(OH)_2$	1.5	0.4	0.2	–	0.1	1.0	0.3
6	$Ca(OH)_2$	1.5	0.4	–	0.2	0.1	1.0	0.3
7	$Ca(OH)_2$	1.5	0.4	0.2	–	–	1.0	0.3

[a] 73 g $Ca(OH)_2$/1.2 l H_2O was used in runs 1, 5, 6 and 7; 73 g $Ca(OH)_2$ + 11 g (SiO_2, $CaCl_2$ or $NaHCO_3$)/1.2 l H_2O were used in runs 2, 3 and 4. The surface area of $Ca(OH)_2$, SiO_2, $CaCl_2$ and $NaHCO_3$ were 12.561, 0.8157, 1.4976 and 2.0176 m^2/g. The spray flow rate of the slurry was 10 ml/min. The mole ratio of $Ca(OH)_2$ to acid gas was 1.5.

[b] 0.036 g of $Pb(NO_3)_2$, 0.092 g of $Cr(NO_3)_2 \cdot 9H_2O$, 0.107 g of $Cd(NO_3)_2 \cdot 4H_2O$, 0.085 g of $Cu(NO_3)_2 \cdot 3H_2O$ and 0.105 g of $Zn(NO_3)_2 \cdot 6H_2O$ were contained in the solution.

was used to determine the temperature in the sand bed. The combustion gases were treated in the spray dryer/fabric filter system and then released into atmosphere by an induced fan.

2.3. Experimental procedure

After calculating the required amount of air for an excess air factor of 30%, 40 l/min of air at room temperature was employed. The combustion chamber was heated to a desired

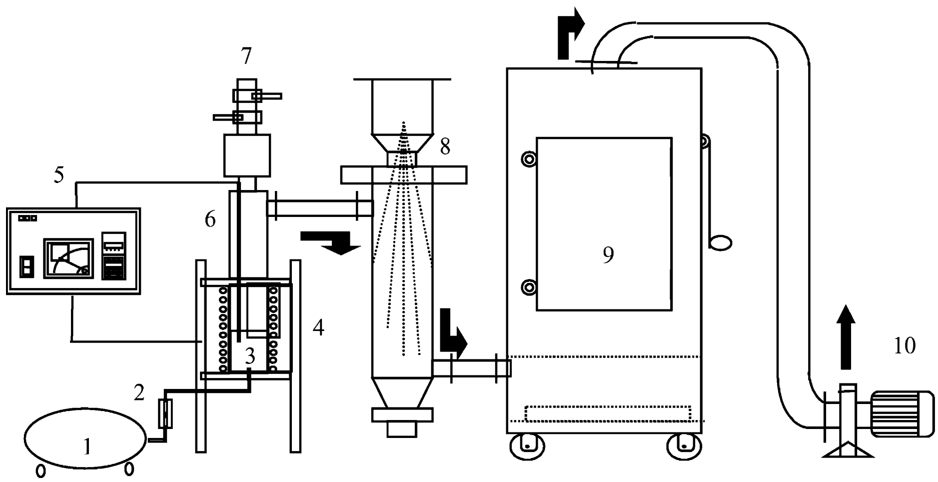


Fig. 1. Schematic of fluidized bed incinerator and APCDs used in the research: (1) air compressor; (2) flow meter; (3) combustion chamber; (4) electrical heater; (5) thermal feedback controller; (6) thermocouple; (7) feeder; (8) spray dryer; (9) fabric filter; (10) induced fan.

temperature (800 °C) by electrical heaters. When the temperature reached steady state, the simulated feedstock was fed into the incinerator at the rate of one bag per 30 s and the APCDs actuated. The inlet and outlet temperature of the spray dryer were maintained at 400–450 and 110–100 °C, respectively. Table 1 lists the other operating parameters of the spray dryer, such as the concentration and volumetric of the absorbent solution. The temperature at the exit of the fabric filter was approximately 45 °C. The pressure drop and filtration velocity of the fabric filter were less than 10 mm H_2O and 0.54 cm/s, respectively. Sampling was performed after 20 min of operation. The total operating time for each test was 100 min.

2.4. Sampling and analysis methods

To estimate the removal efficiency of the spray dryer/fabric filter, the flue gas was simultaneously sampled prior to and after the spray dryer and after the fabric filter. The samples contained acid gases and PAHs. The concentration of SO_2 was analyzed using a flue gas analyzer (Bacharach model 300). The analyzer was calibrated with standard gases before each experiment. The range of concentration for SO_2 monitoring was 0–1999 ppm and the accuracy was ±10 ppm.

The HCl sampling train including a probe, a fiberglass filter and impinger solutions (100 ml of 0.1N NaOH) is shown in Fig. 2. The flue gas containing HCl was sampled using a stainless probe. The gas passed through a fiberglass filter and impingers that removed particles and absorbed HCl, respectively. The sampling flow rate was 12 l/min; the sampling

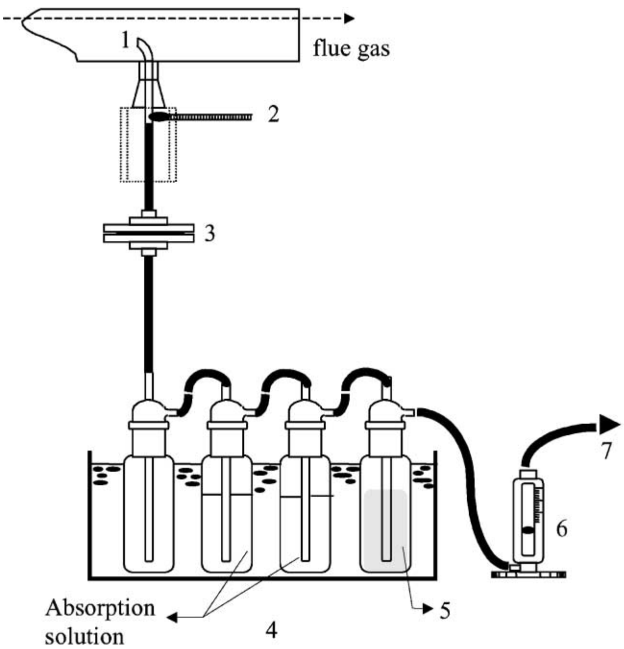


Fig. 2. Sampling train for HCl: (1) sampling probe; (2) thermometer; (3) filter holder; (4) impingers; (5) silica gel; (6) flow meter; (7) connect to vacuum pump.

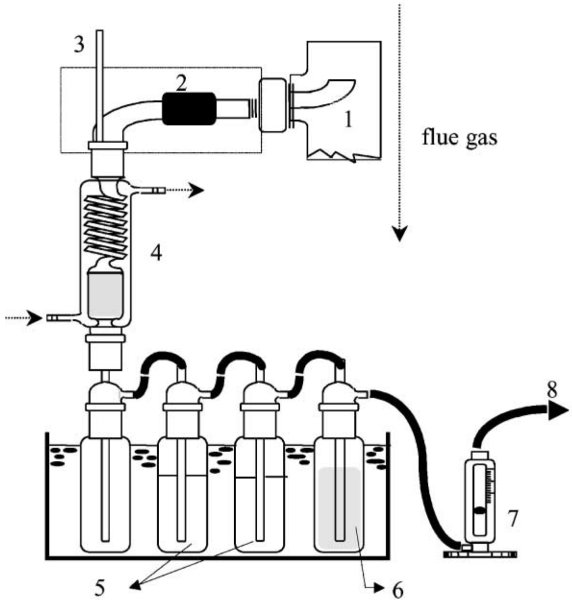


Fig. 3. Sampling train for PAHs: (1) sampling probe; (2) heated filter and heating hose; (3) thermometer; (4) cooling tube and XAD-4 adsorption tube; (5) 200 ml distilled water; (6) silica gel; (7) flow meter; (8) connect to vacuum pump.

period was 10 min. The concentration of HCl was analyzed by the method of colorimetry using mercuric thiocyanate solution [16].

USEPA modified method 5 (MM5) was used for sampling PAHs pollutants. The sampling train for PAHs is shown in Fig. 3. The flue gas containing organics was sampled using a stainless probe. The gas passed through a heated filter packed with fiberglass to collect particles. Then it passed through a cooling tube to capture the remaining organics on XAD-4 adsorbent. Next, the organic samples were extracted for 20 h using the Soxhlet extraction process. The extraction solution was concentrated to 1 ml using a KD evaporator concentrator. The concentrated solutions were put into 1.8 ml brown vials and stored at 4 °C. These samples were later analyzed using a gas chromatograph with a flame ionization detector (Perkin-Elmer Autosystem GC). The recovery efficiency of PAHs was approximately 60.6–99.7%, and the standard deviation of the PAHs analysis was approximately 1.1–15.3. The amount of carbon in fly ash was analyzed using an elemental analyzer (EA).

3. Results and discussion

3.1. Removal efficiency of acid gases

Fig. 4 shows the removal efficiency of SO_2 with various spray dryer additives. The results show that the fabric filter can remove extra acid gases since unreacted sorbents deposited

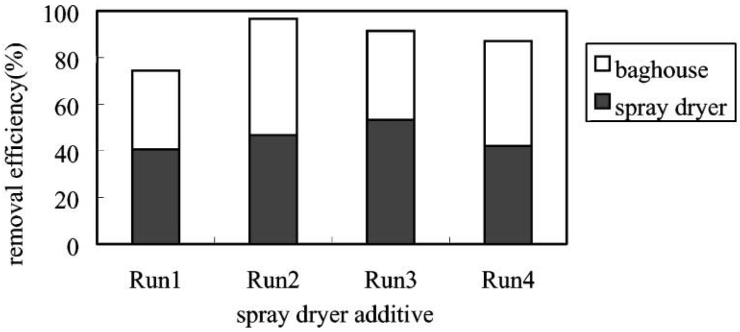


Fig. 4. Effect of spray dryer additives on the removal efficiency of SO_2: $Ca(OH)_2$ (run 1); $Ca(OH)_2 + SiO_2$ (run 2); $Ca(OH)_2 + CaCl_2$ (run 3); $Ca(OH)_2 + NaHCO_3$ (run 4).

upon the fabric filter could further react with the air pollutants. The results also indicate that the better removal efficiency can be attributed to the addition of spray dryer additives. It may be that inorganic deliquescents can retain a greater amount of water on the absorbent surface and thus, increase the reactivity of $Ca(OH)_2$ with SO_2 [4–6]. Izquierdo et al. [6] also indicated that the presence of $CaCl_2$ could increase the kinetic rate constant and the reaction rate increased with increasing concentration of $CaCl_2$. Since the influence of spray dryer additives on the formation of particles and the filtration properties of the fabric filter are not alike [17–20], the removal efficiency of SO_2 in the fabric filter with various additives is different.

The effects of feedstock additives (polyvinyl chloride (PVC) and NaCl) on the removal efficiency of SO_2 are shown in Fig. 5. The results indicate that the presence of PVC can significantly increase the removal efficiency of SO_2 in the spray dryer because PVC can give rise to HCl formation during incineration process [21] and thus, $Ca(OH)_2$ reacts with HCl to form $CaCl_2$ [22]. As described previously, $CaCl_2$ can retain a large amount of water on the absorbent surface and thus, increase the reactivity of $Ca(OH)_2$ towards SO_2 [4–6]. Therefore, the removal efficiency of SO_2 becomes higher when feedstock contains PVC and sulfur than that when feedstock contains only sulfur.

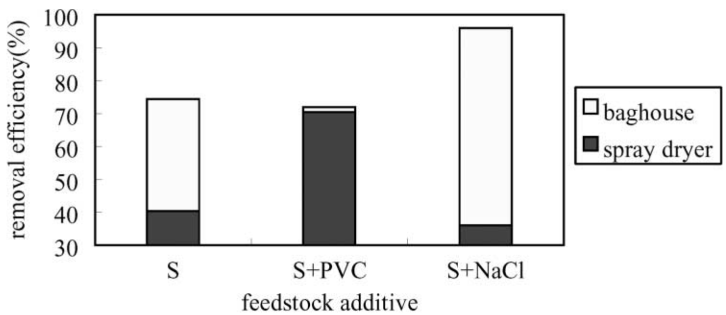


Fig. 5. Effect of feedstock additives on the removal efficiency of SO_2.

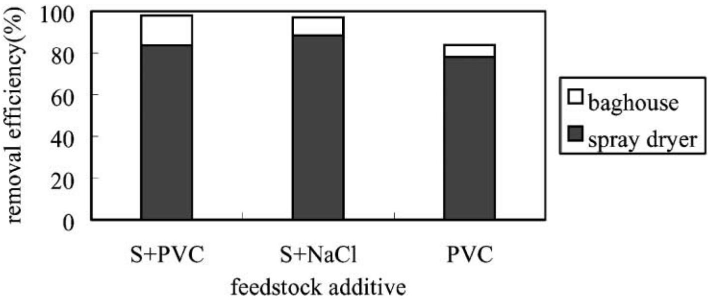


Fig. 6. Effect of feedstock additives on the removal efficiency of HCl.

Fig. 6 shows the removal efficiency of HCl when feedstocks contain sulfur. The results indicate that there is no apparent variation in the removal efficiency of HCl in the spray dryer when the flue gas contains SO_2. This result is due to the reactivity of $Ca(OH)_2$ towards HCl being higher than for SO_2.

3.2. Removal efficiency of fly ash and PAHs

Figs. 7 and 8 show the effects of spray dryer additives (SiO_2, $CaCl_2$ and $NaHCO_3$) and feedstock additives (PVC and NaCl) on the removal efficiency of fly ash and PAHs. The removal efficiency for fly ash is >97%. The better removal efficiency for PAHs can be attributed to the addition of spray dryer additives and feedstock additives. Organic compounds are able to condense or adsorb on fly ash when the temperature in the device decreased. Additionally, the removal efficiency for fly ash is improved by use of a fabric filter. As a result, it is possible to simultaneously remove fly ash and organic compounds using a spray dryer combined with a fabric filter.

The addition of deliquescent additives ($CaCl_2$) and alkali species can increase the cohesivity, roughness and specific surface area of fly ash [17–20]. Furthermore, the particle size distribution of fly ash tends to a finer size when feedstocks contain PVC or NaCl and

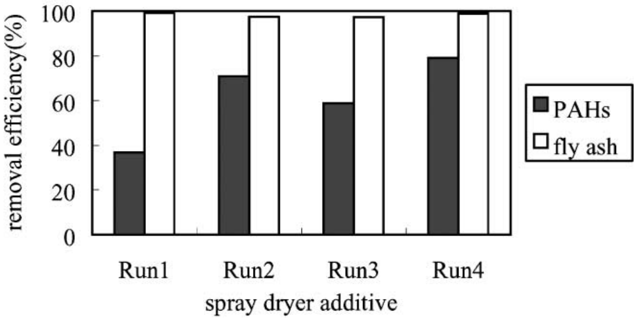


Fig. 7. Effect of spray dryer additives on the removal efficiency of fly ash and PAHs after the fabric filter: $Ca(OH)_2$ (run 1); $Ca(OH)_2 + SiO_2$ (run 2); $Ca(OH)_2 + CaCl_2$ (run 3); $Ca(OH)_2 + NaHCO_3$ (run 4).

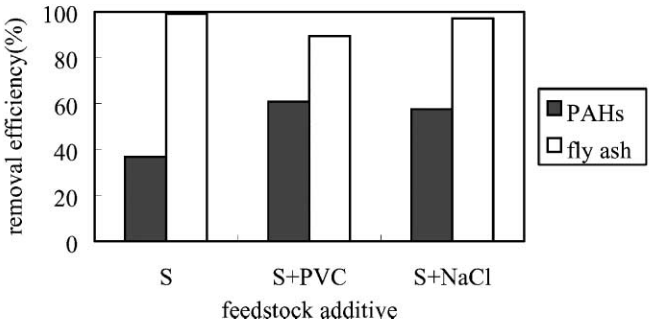


Fig. 8. Effect of feedstock additives on the removal efficiency of fly ash and PAHs after the fabric filter.

thus, increase the specific surface area of fly ash [23]. Therefore, the concentration of PAHs condensed/adsorbed on fly ash can be enhanced either the addition of spray dryer additives (SiO_2, $CaCl_2$ and $NaHCO_3$) or feedstocks additives (PVC and NaCl).

Fig. 9 shows the removal efficiency of signal PAHs using various spray dryer additives (SiO_2, $CaCl_2$ and $NaHCO_3$). The experiments illustrate that the removal efficiency for two-, three- and four-ring PAHs, such as acenaphthylene (AcPy), acenaphthene (AcP), fluorene (Flu), anthracene (AnT), benzo(a)anthracene (BaA) and chrysene (Chr), can be increased using various additives. However, the additives cannot increase the removal efficiency for higher-ring PAHs, such as benzo(b)fluoranthene (BbF), benzo(k)fluoranthene (BkF) and benzo(a)pyrene (BaP). The result can be explained by the fact that lower-ring PAHs (two-, three- and four-ring PAHs) have higher vapor pressures and volatility than higher-ring PAHs (five- and six-ring PAHs) (Table 2). Therefore, the effects of fly ash characteristics (such as specific surface area and pore size) on the removal efficiency of lower-ring PAHs are more significant than that of higher-ring PAHs.

Fig. 10 shows the effects of feedstock additives (PVC and NaCl) on the removal efficiency of signal PAHs. The results indicate that the removal efficiency for two-ring PAHs, such as

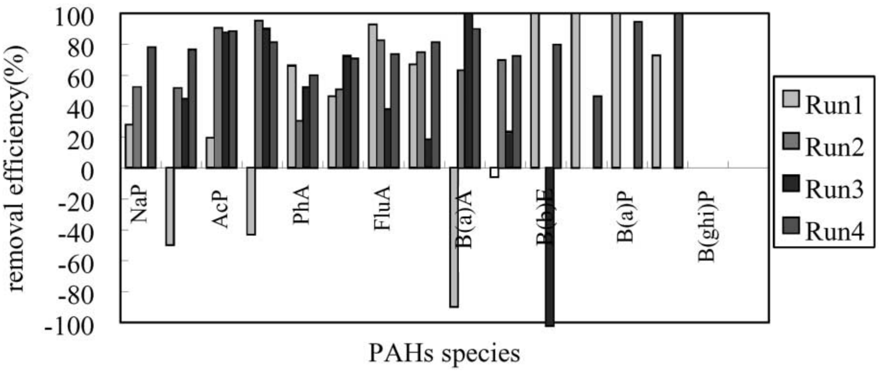


Fig. 9. Effect of spray dryer additives on the removal efficiency of PAHs species after the fabric filter: $Ca(OH)_2$ (run 1); $Ca(OH)_2 + SiO_2$ (run 2); $Ca(OH)_2 + CaCl_2$ (run 3); $Ca(OH)_2 + NaHCO_3$ (run 4).

Table 2
Physical and chemical properties of PAHs [15,24]

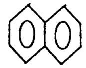
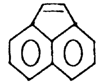
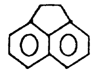
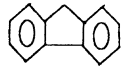
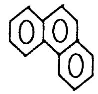
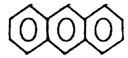
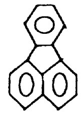
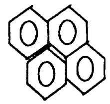
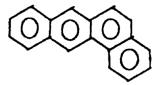
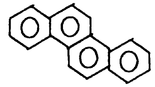
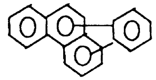
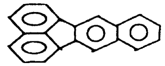
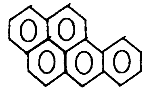
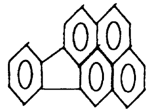
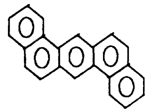
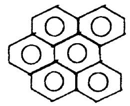
Compound	Chemical formula	Width (Å)	Length (Å)	Thickness (Å)	Molecular weight	Melting point (°C)	Boiling point (°C)	Vapor pressure (mm Hg, 25 °C)	Structure
NaP	$C_{10}H_8$	7.428	9.195	3.884	128.16	80	218	7.1×10^{-2}	
AcPy	$C_{12}H_8$	8.488	9.238	4.221	152.20	93	275	6.7×10^{-3}	
AcP	$C_{12}H_{10}$	8.624	9.242	3.882	154.21	96	279	2.2×10^{-3}	
Flu	$C_{13}H_{10}$	7.521	11.431	4.241	166.22	117	295	6.0×10^{-4}	
Phenanthrene (PhA)	$C_{14}H_{10}$	8.031	11.752	3.888	178.22	100	340	1.2×10^{-4}	
AnT	$C_{14}H_{10}$	7.439	11.651	3.882	178.22	218	342	6.0×10^{-6}	
Fluoranthene (FluA)	$C_{16}H_{10}$	9.240	11.158	3.884	202.26	110	393	9.2×10^{-6}	
Pyrene (Pyr)	$C_{16}H_{10}$	9.279	11.662	3.888	202.26	156	404	4.5×10^{-6}	
BaA	$C_{18}H_{12}$	8.717	13.942	3.887	228.29	159	435	2.1×10^{-7}	

Table 2 (*Continued*)

Compound	Chemical formula	Width (Å)	Length (Å)	Thickness (Å)	Molecular weight	Melting point (°C)	Boiling point (°C)	Vapor pressure (mm Hg, 25 °C)	Structure
Chr	$C_{18}H_{12}$	8.039	13.939	3.922	228.29	256	448	6.4×10^{-9}	
BbF	$C_{20}H_{12}$	9.964	13.817	3.884	252.32	168	393	–	
BkF	$C_{20}H_{12}$	9.242	13.618	3.887	252.32	217	480	9.6×10^{-11}	
BaP	$C_{20}H_{12}$	9.297	13.882	3.891	252.32	177	496	5.6×10^{-9}	
Indeno(1,2,3-c,d)pyrene (InP)	$C_{22}H_{12}$	9.927	13.780	3.884	276.34	162	534	–	
Dibenzo(a,h)anthracene (DbA)	$C_{22}H_{14}$	8.726	15.898	3.890	278.35	262	535	–	
Benzo(g,h,i)perylene (BghiP)	$C_{22}H_{12}$	10.484	11.779	3.887	276.34	273	542	1.01×10^{-10}	

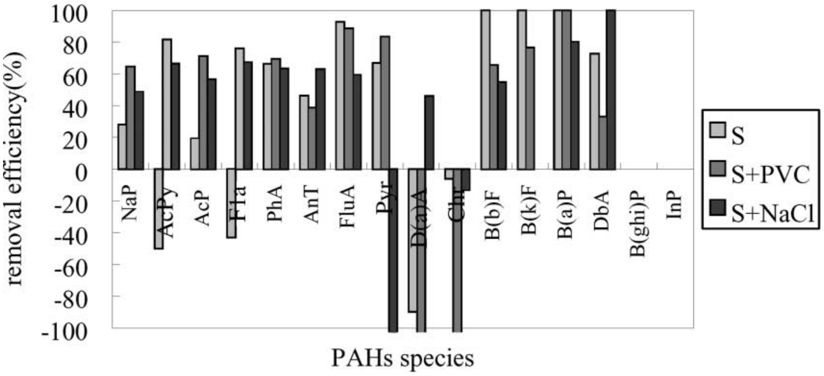


Fig. 10. Effect of feedstock additives on the removal efficiency of PAHs species after the fabric filter.

naphthalene (NaP), AcPy, AcP and Flu, can be enhanced when the feedstocks contains PVC and NaCl. Because metals can react with chlorine to form volatile metal chlorides when feedstocks contained PVC and thus, increase the tendency of metal vapors to condense on foreign nuclei (fly ash and the droplet of $Ca(OH)_{2\,(aq)}$) [25,26]. The presence of metal species in fly ash can provide the activity sites required for the reactions between PAHs precursors and the lower-ring PAHs and thus, increases the formation of organic compounds adsorbed on fly ash [13]. Furthermore, the addition of NaCl can decrease the particle size of fly ash [26,27] and increase the specific surface area of fly ash. As a result, both PVC and NaCl can increase the removal efficiency for lower-ring PAHs. The negative removal efficiency of PAHs (Figs. 9 and 10) implied that lower-ring compounds were catalyzed by heavy metals to form higher-ring compounds and resulted in the higher outlet concentration of higher-ring compounds.

3.3. Relationship of carbon and PAHs

Table 3 reports the amount of carbon in fly ash and the removal efficiency of PAHs. The results show that the amount of carbon in fly ash tends to increase as it passes through the

Table 3
Amount of carbon in fly ash and the removal efficiency of PAHs after the fabric filter

Run	Amount of carbon (wt.%)			Removal efficiency of PAHs (%)
	Fly ash entering a spray dryer	Spray dryer ash	Baghouse ash	
1	0.83	5.12	16.45	36.79
2	2.34	3.14	10.31	70.77
3	3.74	5.63	13.07	58.70
4	0.13	6.21	9.72	78.88
5	0.20	5.71	17.20	60.78
6	0.02	7.05	14.21	57.55
7	1.86	9.22	11.99	42.68

spray dryer and the fabric filter. The results demonstrate that the organic compounds are able to condense/adsorb on fly ash as they pass through the APCDs. However, removal efficiency of PAHs is not proportional to the amount of carbon in fly ash. Because the concentration of organic compounds condensed/adsorbed on fly ash is affected simultaneously by the specific surface area of fly ash, physical and chemical properties of organic compounds and chemical composition of fly ash such as the concentration of heavy metal. Therefore, the amount of carbon in fly ash has inconsistent effect on the removal efficiency of PAHs.

4. Conclusions

This study investigated the influence of operating conditions including feedstock additives (PVC and NaCl) and spray dryer additives (SiO_2, $CaCl_2$ and $NaHCO_3$) on the removal efficiency of acid gases and PAHs. The results indicate that the better removal efficiency for acid gases can be attributed to the reaction of filter cake with the flue gases. The removal efficiency for SO_2 can be enhanced when SiO_2, $CaCl_2$ and $NaHCO_3$ are added to the spray dryer and PVC exists in the incineration feedstock. The addition of spray dryer additives (SiO_2, $CaCl_2$ and $NaHCO_3$) also significantly increase the removal efficiency for two-, three- and four-ring PAHs, such as AcPy, AcP, Flu, AnT, BaA, and Chr. However, these additives cannot increase the removal efficiency for BbF, BkF and BaP. The removal efficiency for two-ring PAHs, such as NaP, AcPy, AcP and Flu is also be enhanced when the feedstock contains PVC and NaCl.

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