



Incineration of Pelletized Fly Ash in a Bench-scale Fluidized Bed Combustor

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ABSTRACT

The purpose of this study was to reduce the dioxin content of fly ash from an incinerator by thermal treatment. To increase the residence time of fly ash in the incinerator, the fly ash was mixed with different amounts of coal and clay and then pelletized. All experiments were conducted in a bench-scale fluidized bed combustion system. The polychlorinated dibenzo-*p*-dioxin and dibenzofurans (PCDD/Fs) content of the fly ash were investigated following combustion. The results showed that the I-TEQ (Toxic Equivalents Quantity of 2,3,7,8-tetrachlorinated dibenzo-*p*-dioxin) values of the ash collected from the cyclone met Taiwan's regulatory standards ($< 1 \text{ ng g}^{-1}$). The total toxicity of dioxin, including those in the fly and bottom ash, can be reduced by 88–95% after combustion. The strength of the pellets is enhanced by the addition of clay, resulting in their aggregation in the bed and discharge as bottom ash. The distribution profiles of the PCDD/Fs homologs in the fly ash collected from the baghouse in each experiment were similar. The most prevalent congeners in the ash were 1,2,3,4,6,7,8-HpCDF, OCDD, and OCDF. The predominant contributor to the total I-TEQ values was 2,3,4,7,8-PeCDF. The amount of residual carbon in the fly ash is strongly correlated with the dioxin content of the fly ash collected from the baghouse.

Keywords: Dioxin; Fly ash; Fluidized bed combustor; Thermal treatment

INTRODUCTION

The incineration of municipal solid or medical waste is prone to high dioxins emissions. The operating conditions and pollution control devices are key factors for controlling dioxin emissions. The emissions of dioxins to the atmosphere from incineration systems must be decreased, and, typically, the dioxins produced from incinerators are captured by the air pollution control division (APCD) and remain in the fly ash.

Lustenhouwer *et al.* (1980) suggested that polychlorinated dibenzodioxins (PCDDs) and related compounds may form *de novo* from carbon sources and natural chlorine. *De novo* synthesis is catalyzed by a heterogeneous gas-phase reaction between the fly ash particles and flue gas (Vogg and Stieglitz, 1986; Stieglitz *et al.*, 1989). A theory proposed by Griffin (1986) and tested by Hagenmaier *et al.* (1987) demonstrated that formation occurs through the *de-novo* synthesis of compounds within the flue gas and fly ash. This

involves the Deacon process, in which the Cl_2 produces chlorinated aromatic ring structures through substitution reactions. The chlorine formed by the Deacon reaction is essential for the *de-novo* synthesis of PCDD/Fs (Bruce *et al.*, 1991). Yan *et al.* (2012) stated that fly ash functions as the key catalyst of the heterogeneous formation of PCDD/Fs. Buekens and Huang (1998) suggested that residual carbon in fly ash is the main carbon source for the *de novo* synthesis of PCDD/Fs. Stieglitz *et al.* (1989) found that PCDD/Fs form in the fly ash as the furnace effluent cools to moderate temperatures near 300°C. Addink and Olie (1995) stated that the optimum temperature for *de novo* synthesis is between 300 and 400°C, while Gullett *et al.* (1992) found that the temperature for the formation of PCDD/Fs in municipal waste combustors ranged from 200 to 400°C. Aurell and Marklund (2009) stated that the dioxin formation pathways are not fully understood, but they are believed to be formed by heterogeneous reactions occurring in the post-combustion zone of the incinerator.

Zhou *et al.* (2015) noted that fly ash is important in the formation of PCDD/Fs and dioxins also concentrate in the fly ash collected from the baghouse. Abad *et al.* (2002) found that fly ash constitutes 70 to 90% of the total PCDD/Fs emitted from incinerators. Hung *et al.* (2018) found that the proportion of total PCDD/Fs in the solid phase ranged from 45.5% to 73.8%. The disposal of fly ash

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with high concentrations of PCDD/Fs must be sufficiently. Stanmore and Clunies-Ross (2000) stated that all PCDD/Fs from medical waste incinerators are related to fly ash, which has been supported by experimental data (Hell *et al.*, 2000; Stanmore, 2002; Tame *et al.*, 2003). Most PCDD/Fs are formed through heterogeneous reactions that occur in the post-combustion zone of the incinerator at temperatures of 250–400°C and then remain in the fly ash (Gullett *et al.*, 1992; Faengmark *et al.*, 1994; Stanmore, 2004).

Some studies have shown that dioxins can be destroyed at high temperatures. Vogg and Stieglitz (1986) found that, under oxidative conditions, a temperature of 400–600°C could decompose PCDD/Fs in the air stream. Similar results have been obtained under different conditions in subsequent studies by other researchers (Glatzmaier *et al.*, 1990; Kahr *et al.*, 1990; Kobylecki *et al.*, 2001; Lin *et al.*, 2015). Besides thermal treatment methods, many studies on the treatment of dioxin in fly ash have considered other methods, such as the non-thermal plasmas (Zhou *et al.*, 2003), supercritical water oxidation (Sako *et al.*, 1997), inhibitor-addition (Yan *et al.*, 2012; Chen *et al.*, 2014; Fu *et al.*, 2015; Zhan *et al.*, 2016), re-burning of the froths obtained after flotation (Wei *et al.*, 2017a), microwave energy (Wei *et al.*, 2017b), and microwave irradiation (Liu *et al.*, 2019) method.

From a practical viewpoint, well-developed technologies are always preferred over advanced technologies. Cost is a key factor affecting their successful application in commercial processes. Vehlow *et al.* (1990) proposed the 3R process, which involves the purification of flue gas and residue treatment, to resolve the issue of fly ash from municipal waste incinerators, and their results showed that the products of the process easily met the limits of the Swiss eluate test. Kobylecki *et al.* (2001) found that the dioxin in fly ash from municipal waste incinerators could be reduced by pelletizing a mixture of fly ash, cement, and sodium phosphate, and then re-burning these materials in a laboratory-scale bubbling fluidized bed furnace. At 700°C and with a sufficient incineration time for complete burnout, the efficiency of reducing the dioxin content of the pellets was 99.7–99.9%. Huber *et al.* (2018a) demonstrated that the PCDD/Fs contents of pelletized fly ash can be reduced significantly through thermal treatment in an electrically heated pilot-scale rotary kiln at temperatures above 650°C.

As mentioned above, the pelletizing and re-burning method for decomposing the dioxins contained in fly ash is relatively simple and inexpensive. Although much research has been conducted on this topic, the formation of dioxin in the fly ash at the post-combustion zone. The environmental impact of the co-treatment of municipal solid waste incinerator (MSWI) fly ash with a combustible hazardous waste is lower than that of the thermal treatment of MSWI fly ash in a separate furnace (Huber *et al.*, 2018b). Re-burning using the original incineration system is proposed in this study owing to its economic benefits. The purpose of this study is to investigate the formation of dioxin in the fly ash in the baghouse during the re-burning of pelletized fly ash in a bench-scale fluidized bed combustor. The reduction of the total dioxin toxicity of the fly and bottom ash are also studied.

METHODS

Materials

The raw feeding material used in this study was fly ash from an industrial waste incinerator. After thoroughly mixing various proportions of clay and coal into the ash, the material was pelletized into particles with a diameter of 6 mm and length of approximately 10 mm (as shown in Fig. 1). The compositions of the feeding materials denoted as A, B, C, and FA are listed in Table 1. Feed material of A was raw material mixed with 5 wt% clay, feeding material of B was raw material mixed with 4.8 wt% of clay and 9.5 wt% of coal, feeding material of C was raw material mixed with 4.5 wt% of clay and 13.5wt% of coal, feeding FA was raw material without any other substances. Clay was added to enhance the cohesive force within the pellets. Coal serves as a source of sulfur and may inhibit the formation of dioxins. The ultimate and approximate analyses of the feeding materials are also shown in Table 2. Silica sand (99.5% SiO₂ and 0.5% Al₂O₃) with a mean diameter of 0.54 mm and apparent density of 2500 kg m⁻³ was used as the bed material in this study.

Apparatus

All experiments were conducted in a bench-scale fluidized bed combustor (FBC). The FBC test facility was



Fig. 1. Image of the pelletized fly ash.

Table 1. Composition of the feeding materials.

Additives	Feeding materials			
	FA	A	B	C
Fly ash (wt%)	100	94.7	85.7	81.
Clay (wt%)	0	5.3	4.8	4.5
Coal (wt%)	0	0	9.5	13.5
HHV (WB)	4421.5	4236	4586.5	4833
LHV (WB)	3347	3258	3449	3705

Table 2. Proximate and ultimate analyses of the feeding materials.

feeding material	Proximately analysis (wt% as-received)			Ultimate analysis (wt% Dry and ash free)					
	Moisture	Combustible	Ash	C	H	O	N	S	Cl
Fa	19.39	62.07	18.54	77.07	2.36	17.19	0.96	0.63	1.79
A	18.76	55.79	25.45	75.70	2.47	18.10	0.81	0.69	2.22
B	19.69	56.84	23.47	75.47	2.93	18.18	0.85	0.69	1.87
C	20.41	52.32	27.27	75.57	2.95	18.17	0.90	0.70	1.71

a staged-combustion facility, and a schematic diagram of this system is presented in Fig. 2. The system consists of a FBC, a screw feeder, a gas supply system, an electric heating system, and air pollution control devices (APCDs).

The FBC included a windbox, distributor, combustion chamber, and freeboard. The cross-section of the combustion chamber was a rectangle of $0.22 \times 0.11 \text{ m}^2$, and its height was 680 mm. The distributor, with a cross-section of $0.22 \times 0.11 \text{ m}$, consisted of 13 tuyères with an opening ratio of 0.64%. The freeboard of the FBC was a SUS 310 stainless-steel column reactor with an internal diameter of 154 mm and height of 4 m. The combustion chamber and freeboard were enclosed in a thermal insulation material with a thickness of 25 mm.

The test rig was equipped with a feedstock hopper. The inert bed material and pelletized fly ash were conveyed to the bottom of the combustor by a screw feeder. The variable rotating motor of the screw feeder, based on the calibration curve, controlled the feeding rate, and the feeding point was located 0.5 m above the distributor.

The primary gas and secondary air were supplied by a 20-horsepower air compressor. Four equally spaced secondary air injection nozzles with a diameter of 13 mm were installed tangentially 800 mm above the distributor.

The outsides of both the combustion chamber and freeboard were equipped with electric heating systems to minimize heat loss. During the tests, the temperatures of the bed and freeboard were maintained by the electric heating system.

The flue gas generated by the combustor traveled through a cyclone to remove most of the unburned char and ash particles, and was then cooled by air to approximately 150°C . The fly ash in the flue gas was collected in the baghouse. Then flue gas was then released through a stack into the atmosphere.

Data Acquisition

After each test, the dioxin concentrations of the fly ash collected from the cyclone and the baghouse, and the bottom ash were analyzed. The samples were sent to the

Industrial Technology Research Institute of Taiwan (ITRI) for analysis following the high-resolution gas chromatography/high-resolution mass spectrometry/data system (HRGC/HRMS) method (EPA Method 8290A). An Agilent 6890N HRGC device and a Thermo Finnigan MAT 95XL HRMS device were used. The components of the flue gas, such as CO , O_2 , and NO_x , were detected using HORIBA-PG250 gas analyzers. In particular, NO_x was analyzed following the chemiluminescence detection method; the repeatability and linearity are $\pm 0.5\%$ and $\pm 2.0\%$ of the full scale, respectively. To reduce the influence of the memory effect, the sand was replaced and the remaining fly ash in the system was removed with a large amount of air for each test.

Experimental Conditions

Before start-up, silica sand was added to the combustion chamber, and the static bed height was fixed at 25 cm throughout the experiments. The electric heater of the chamber was then switched on to heat the bed material. The bed material was stirred every 30 min by the primary gas for homogenous heating. The freeboard temperature was controlled to be close to the bed temperature. When the bed temperature reached 400°C , the feeding material was fed into the combustor, and the gas flow rates were adjusted to the set values. The freeboard temperature was set at 850°C , and maintaining this temperature requires an average of 20 kW of energy. When the bed temperature's deviation was $\pm 5^\circ\text{C}$, it was considered to have reached a steady state and sampling begun.

In this study, the feeding rate was fixed at 2 kg h^{-1} , the total input gas was constant at 330 NL min^{-1} , the primary gas flow rate was 250 NL min^{-1} , and the fixed secondary gas flow rate was 80 NL min^{-1} . The experimental conditions for all tests are shown in Table 3.

RESULTS AND DISCUSSION

Table 2, shows that the proportions of moisture, volatile matter, and fixed carbon in the raw material of ash were

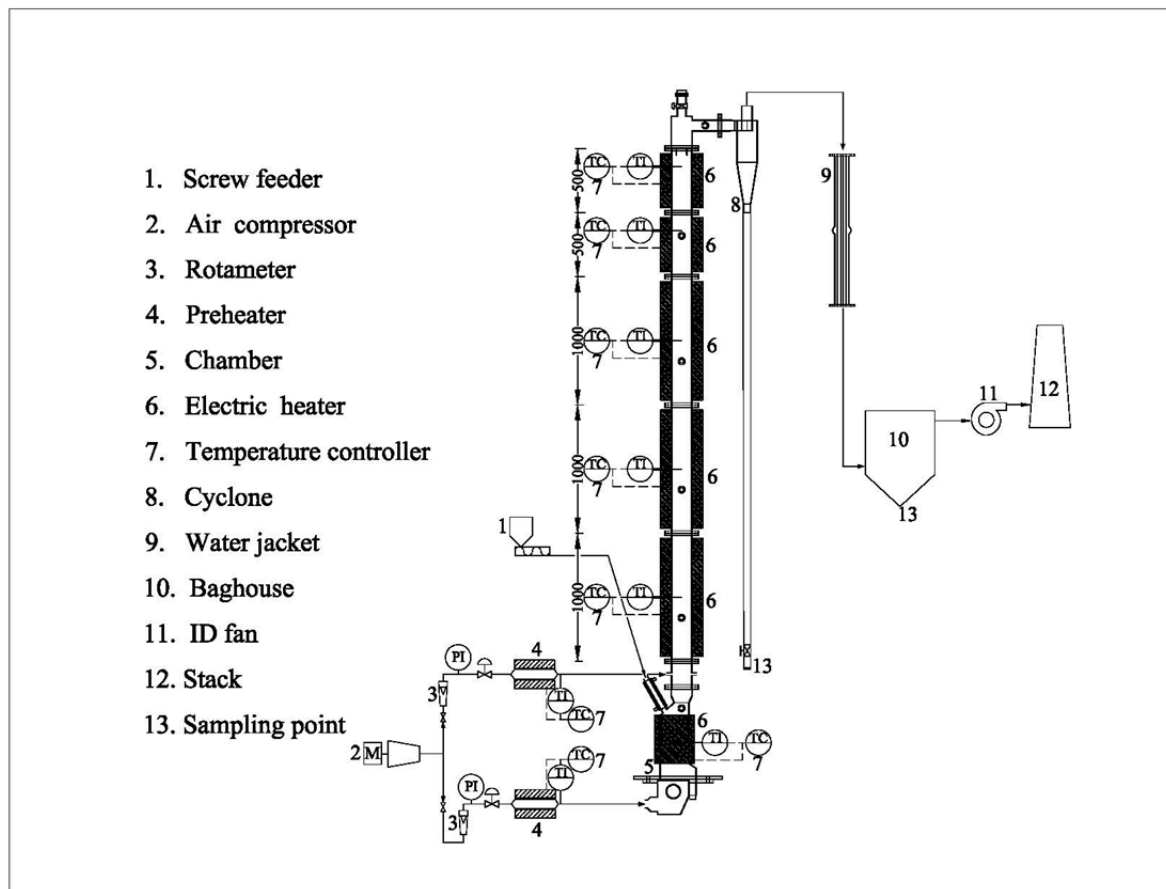


Fig. 2. Schematic diagram of the bench-scale fluidized bed combustor.

Table 3. Experimental conditions.

Operating parameter	Conditions
Feeding rate (kg h ⁻¹)	2
Bed temperature (°C)	750, 850
Freeboard temperature (°C)	850
Superficial gas velocity (m s ⁻¹)	0.68
Primary air rate (NL min ⁻¹)	250
Secondary air rate (NL min ⁻¹)	80
Bed material	Silica sand
Average particle size of bed material (μm)	566
Bed weight (kg)	10

19.4%, 18.5%, and 62.1%, respectively, indicating that the raw material contains a high amount of unburned carbon. Table 4 shows that the concentration of PCDD/Fs in the feeding materials ranged from 187.7 to 273.5 ng g⁻¹, and the I-TEQ ranged from 17.2 to 23.3 ng I-TEQ g⁻¹, which exceeds the regulatory standard of the Taiwan environmental protection administration (EPA) (limit of 1 ng I-TEQ g⁻¹). The PCDF/PCDD ratios of the feeding materials ranged from 1.97 to 2.08, indicating that PCDFs were the dominant congeners. The results of the elemental analysis of the fly ash after burning are shown in Table 5, and the major metals were Ca, Fe, Sn, and Mg.

The temperature in the freeboard zone was maintained at 850°C by an electrical heating system, and the temperature

at the furnace outlet was 650°C owing to the loss of heat to the surroundings. Owing to the natural heat loss through air, the temperature at the outlet of the cyclone was 420°C. The flue gas in the cooling pipe (between the cyclone outlet and the inlet of baghouse) was cooled by air. The air flow rate in this experiment was 330 L min⁻¹, the length of the pipe was 5.2 m, and the inner diameter of the pipe was 73 mm. The flue gas flow rate (Q_f) can be calculated by the mass balance using the ultimate analysis data shown in Table 2 and the air flow rate. The residence time of the flue gas in this region was approximately 7.12 seconds, which was calculated by Eq. (1).

$$4\pi \times \left(\frac{D}{2}\right) \times L^2 \left\{ Q_f \times \left[\frac{(273 + T)}{273} \right] \right\} \quad (1)$$

D: inner diameter of the cooling pipe, cm.
 L: distance from the outlet of the cyclone, cm.
 Q_f : flue gas flow rate, cm³ sec⁻¹.
 T: temperature at L, °C.

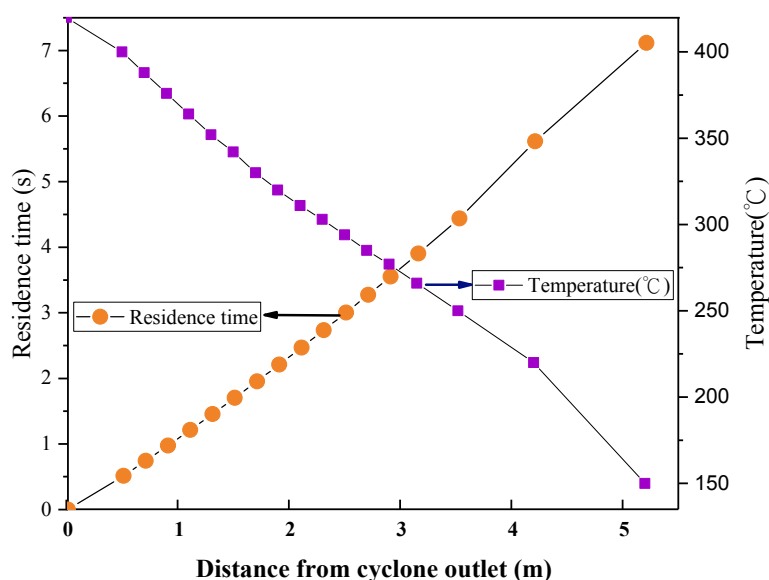
Fig. 3 shows that the temperature of the flue gas decreased to 400°C 0.5 m after exiting the cyclone's outlet, and to 250°C at a distance of 3.52 m. The residence span from 400 to 250°C is approximately 3.93 s. The flue gas appears to remain longer in a favorable environment for *de novo* synthesis, which will be discussed later.

Table 4. PCDD/Fs concentrations and I-TEQ values of the fly ash.

Concentration of PCDD/Fs		Feeding materials			
		FA	A	B	C
PCDD/Fs (ng g ⁻¹)	Raw material	273.42	244.83	214.59	187.58
	Fly ash collected from cyclone	9.99	12.01	10.96	26.67
	Fly ash collected from baghouse	1121.07	1474.82	1794.38	2163.79
I-TEQ (ng g ⁻¹)	Raw material	23.3	20.5	17.7	17.2
	Fly ash collected from cyclone	0.42	0.33	0.36	0.93
	Fly ash collected from baghouse	50.3	60.4	67.1	76.3
PCDF/PCDD ration	Raw material	2.04	2.08	2.00	1.97
	Fly ash collected from cyclone	4.53	5.00	3.07	3.45
	Fly ash collected from baghouse	3.82	1.35	2.76	2.25

Table 5. Metal analysis of the ash after burning.

Feeding materials		element								
		Mg (wt%)	Ca (wt%)	Fe (wt%)	Ni (ppm)	Cu (ppm)	Zn (wt%)	Pd (ppm)	Sn (ppm)	Pb (wt%)
F A	Baghouse fly ash	1.564	6.507	4.549	2956	9360	1.355	302.3	6890	1.564
	Cyclone fly ash	1.397	6.146	8.325	3923	8433	1.767	289.6	7803	1.397
	Bottom ash	0.257	1.079	1.235	641.9	1136	2265	44.62	1273	0.257
A	Baghouse fly ash	1.536	6.899	4.248	2771	9983	1.337	322.4	6256	1.536
	Cyclone fly ash	1.212	6.455	7.121	4836	7460	1.552	280.8	6408	1.212
	Bottom ash	0.214	0.866	2.399	891.7	972.6	2140	46.60	1030	0.214
B	Baghouse fly ash	0.794	5.545	4.393	2886	10180	1.392	340.8	6363	0.794
	Cyclone fly ash	1.272	5.834	6.597	3277	6995	1.453	272.5	6407	1.272
	Bottom ash	0.137	0.499	2.173	343.0	481.7	1205	22.60	513.0	0.137
C	Baghouse fly ash	1.352	7.887	4.515	2768	9358	1.347	343.5	6508	1.352
	Cyclone fly ash	1.236	5.547	6.645	4148	6475	1.376	289.5	5713	1.236
	Bottom ash	0.112	0.333	3.804	1632	453.9	1387	27.10	595.9	0.112

**Fig. 3.** Temperature and residence time distribution profile of flue gas from the cyclone outlet to baghouse inlet.**Concentration of PCDD/Fs in the Fly Ash after Burning**

After burning, the concentration of PCDD/Fs in the fly ash collected from the cyclone and the baghouse ranged from 9.99 to 26.67 ng g⁻¹ (0.326 to 0.926 ng I-TEQ g⁻¹), and 1121.07 to 2163.79 ng g⁻¹ (50.3 to 76.3 ng I-TEQ g⁻¹),

respectively, as shown in Table 4 and Fig. 4. The results showed that the I-TEQ values of the ash collected from the cyclone meet Taiwan's regulatory standards (< 1 ng g⁻¹), which agrees with previous work (Shen *et al.* (2019).

The distributions of residue collected from baghouse,

cyclone, and bottom ash after burning are shown in Fig. 4 and Table 6. Approximately 10% and 2% of the ash was collected from the cyclone and baghouse, respectively. Adding clay to the feeding materials increased the amount of bottom ash from 5% to 20%.

The PCDD/Fs concentrations of the fly ash collected from the baghouse were two orders higher than those of the fly ash collected from the cyclone. Chang and Huang (1999) demonstrated that the dioxin concentration of fly ash collected from baghouses is higher than that from cyclones, and they found that the PCDD/PCDF ratio of ash collected from a cyclone was higher than that of the fly ash collected from the boiler and baghouse. This can be attributed to the dioxin formation mechanism. Heterogeneous precursor condensation yields more PCDDs than PCDFs (Huang and Buekens, 1995), while *de novo* synthesis yields more PCDFs than PCDDs. The *de novo* synthesis of PCDD/Fs occurred in the presence of oxygen at temperatures of approximately 250–400°C (Vogg *et al.*, 1987; Milligan and Altwicker, 1993; Cunliffe and Williams, 2009). Before the fly ash was trapped by the baghouse, *de novo* synthesis reaction occurs in a suitable environment, resulting in an increase in the dioxin concentration of the fly ash in the bag filter (Lundin and Marklund, 2008). The temperature inside the cooling pipe in this experiment is suitable for *de novo* synthesis, which is the major cause of the high concentration of

dioxin in the fly ash collected from the baghouse.

In this study, the high-temperature flue gas and particulates were separated by a cyclone. Due to the centrifugal force, the particles of larger size were thrown to the wall of the cyclone and falling down along the wall to the bottom, and the flue gas with fine particles was discharged from the outlet of the cyclone. The ash collected at the bottom of the cyclone was located in a gas flow stagnation zone, with no downward flue gas flow. In this zone, *de novo* synthesis can be neglected as the environment was without flue gas. Owing to the extremely low dioxin and PCDD contents of the ash, no *de novo* reaction occurred in this area, and the combustion performance of the combustor is very good. Chang and Huang (1999) found that the dioxin concentration of cyclone ash was lower than that of baghouse ash, and the PCDD was higher than PCDF, indicating that most of the dioxin originated from the precursor reaction. The data shown in Table 4 indicate that the PCDF concentration was greater than that of PCDD for fly ash collected from the cyclone. Vogg and Stieglitz (1986) found that the thermal stability of PCDF is higher than that of PCDD. Luijk *et al.* (1994) found that PCDF is less susceptible to decomposition than PCDD at the same temperature. As the feeding materials already contain high concentrations of dioxin and PCDF > PCDD, as shown in Table 4, the PCDF/PCDD ratio in the fly ash collected from the cyclone is higher

Table 6. Weight of ash collected from various parts of the system after burning for each test run.

Weight (kg)	Feeding materials				
	FA (750°C)	A (750°C)	B (750°C)	C (750°C)	C (850°C)
Raw material	15	15	15	15	15
Fly ash collected from cyclone	1.5	1.65	1.78	1.85	1.9
Fly ash collected from baghouse	0.3	0.36	0.34	0.4	0.5
Bottom ash	0.78	3.24	3.23	3.43	3.39

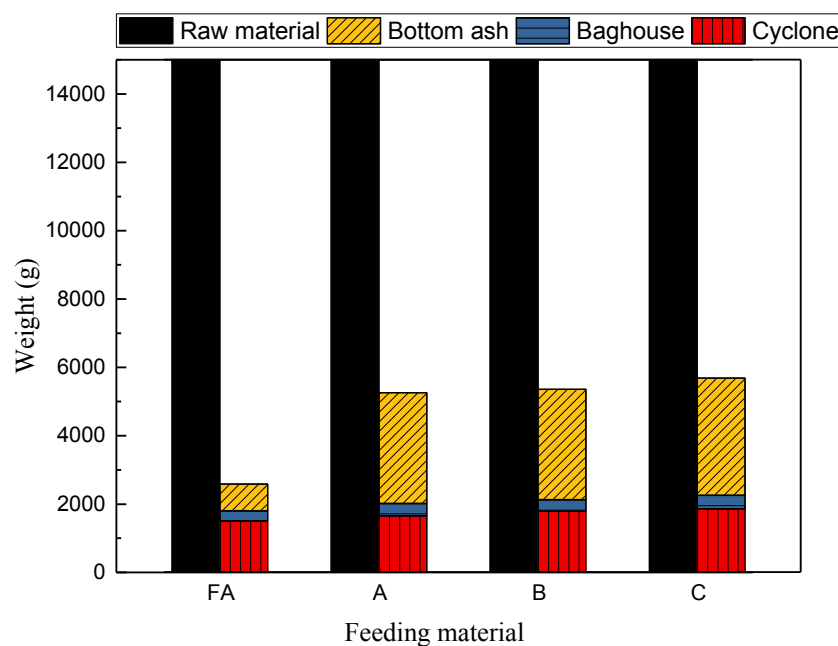


Fig. 4. Weight of ash collected from various parts of the system after burning.

than that of the raw material. To reduce the residence time available for *de novo* synthesis, a quenching tower is suggested for practical application.

Generally, the cyclone trapped a large amount of the fly ash elutriated (31 to 33% of total ash) from the combustor, and the dioxin content of fly ash at the bottom of the cyclone can be neglected. Thus, the quantity of fly ash with a high dioxin content remaining in the baghouse (6 to 7% of total ash) was reduced. In the cyclone, the total residual toxicity generated from the burning process was significantly reduced.

Effect of Coal on Dioxin in Fly Ash

The experimental results obtained in this study indicated that the dioxin concentration of the fly ash collected from baghouse increased with the proportion of coal added to the pelletized fly ash, as shown in Figs. 5 and 6. This disagrees with the results of the previous reports, which indicated that coal inhibits dioxin formation.

Ogawa *et al.* (1996) showed that coal with a relatively high sulfur content can inhibit dioxin formation. Gullett *et al.* (2000) used sulfur-containing coal and waste in a commercial-scale waste incinerator, and the dioxin concentration was significantly reduced. Ogawa *et al.* (1996) found that the

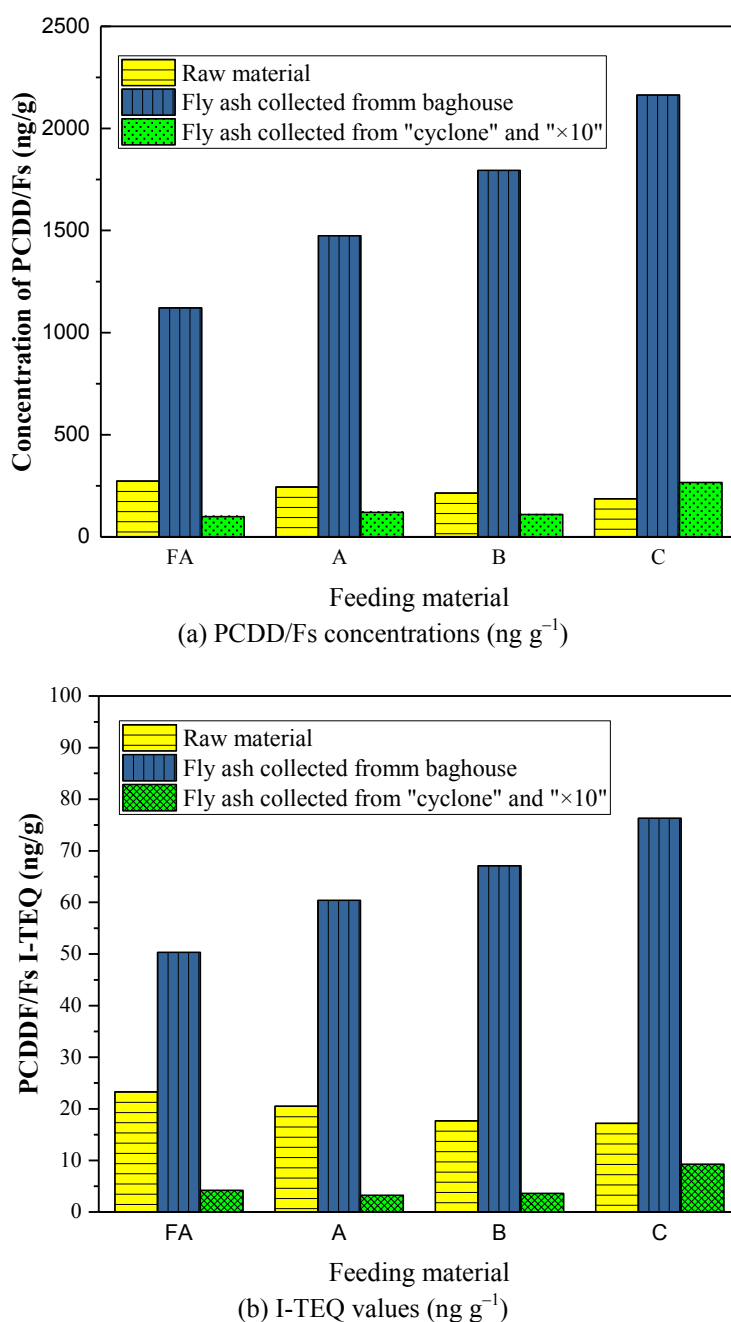


Fig. 5. Dioxin concentration of the feeding materials and fly ash collected from the cyclone and baghouse after burning; (a) PCDD/Fs concentrations and (b) I-TEQ values.

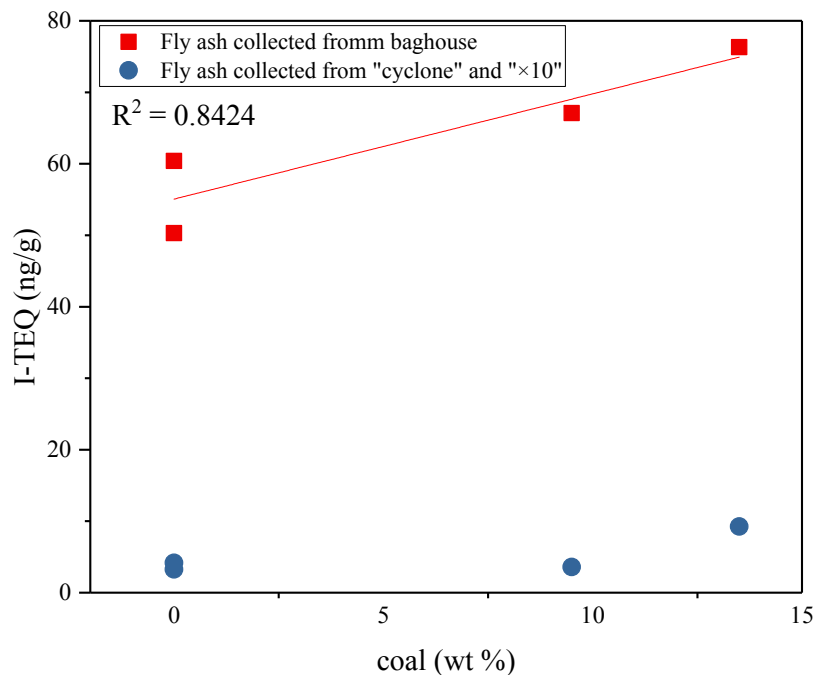


Fig. 6. Relationship between the I-TEQ value of the fly ash after burning and the amount of coal added to the feeding material.

inhibitory effect increased with an increasing S/Cl molar ratio (0.1–0.65). Anthony *et al.* (2001) found that the addition of sulfur at a higher ratio (S/Cl > 1) could reduce the emission of PCDD/Fs; however, no benefit was derived from the addition of higher amounts of sulfur (S/Cl > 2). The ultimate analysis shown in Table 1 indicated that the S/Cl ratios of feeding materials FA, A, B, and C were 0.39, 0.35, 0.41, and 0.45, respectively, and the experimental data obtained from this test show that the S/Cl ratio did not notably affect the reduction of PCDD/Fs, which agreed with the results of Anthony *et al.* (2001).

Xie *et al.* (2000) demonstrated that the inhibitory effect on the formation of dioxin is poor when the sulfur content is below than 0.72 wt% (dry and ash-free basis). The addition of coal may increase the products of incomplete combustion (PICs). The structure and surface area of organic carbon were also key factors influencing dioxin formation (Dickson *et al.*, 1992; Milligan and Altwicker, 1993; Luijk *et al.*, 1994). Buekens and Huang (1998) indicated that the residual carbon in fly ash is the main carbon source for the *de novo* synthesis of PCDD/Fs. The results presented by Wikström *et al.* (2004a, b) indicated that soot and ash are the two main factors contributing to PCDD/Fs. Yan *et al.* (2012) found that fly ash acts as the key catalyst for the heterogeneous formation of PCDD/Fs. Stieglitz *et al.* (1989) added different amounts of carbon to simulate *de novo* synthesis in the same system, and their results showed that the concentration of dioxin increased with increasing addition of carbon. Kakuta *et al.* (2005) found that the amount of dioxin produced by *de novo* synthesis decreases with a decrease in the residual carbon content.

Fig. 7 shows that the amount of residual is positively

correlated with the concentration of PCDD/Fs. The data obtained in this study agreed reasonably well with the results mentioned above. The residual carbon concentration was considered to be the main factor of *de novo* synthesis. The formation of dioxin is also related to the carbon content of the fly ash.

Fig. 8 shows the I-TEQ values of the fly ash collected from the cyclone and baghouse for burning feeding material C at bed temperatures of 750 and 850°C. The results shown in Fig. 8 indicate that an increase in the bed temperature results in more complete combustion, reducing the PICs and thereby reducing dioxin formation.

Owing to the insufficient sulfur content, the inhibitory effect of sulfur on dioxin formation is not clear in this study, while the addition of coal causes an increase in the PICs, resulting in an increase in the formation of dioxins. An increase in the bed temperature can reduce the amount of residual carbon elutriated from the bed, resulting in the reduction of dioxin formation.

Effect of Clay on Dioxin in Fly Ash

Clay was added to the pelletized ash mixture in order to enhance the cohesive forces within the pellet and inhibit cracking, breaking, or crushing, which reduces the entrainment of fly ash into the bag filter, thereby reducing *de novo* synthesis and discharging most of the fly ash as bottom ash. The experimental results indicate that our goal was not achieved. However, the amount of fly ash collected from the baghouse was reduced significantly. Owing to the inclusion of clay, most of the fly ash was collected at the bottom of the cyclone or remained in the bed material as bottom ash, and only a small amount escaped to the baghouse.

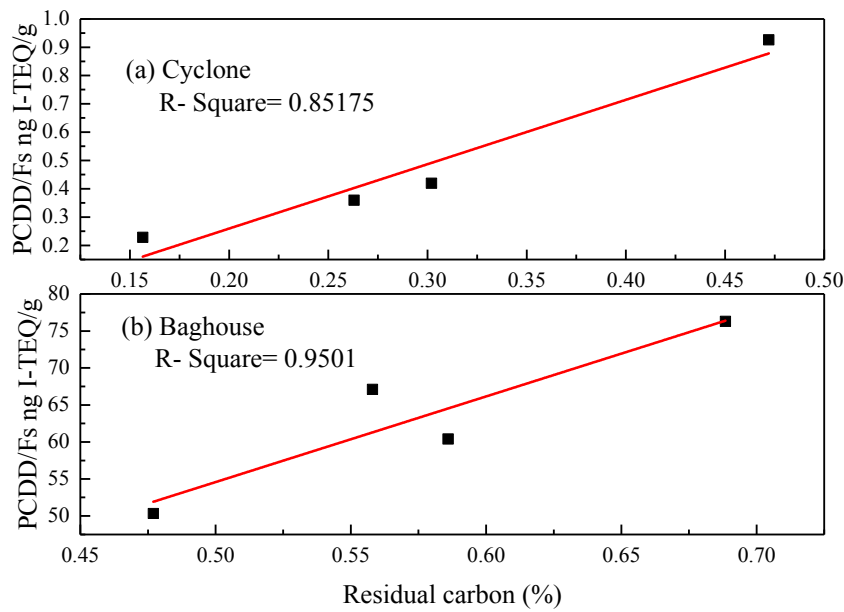


Fig. 7. Relationship between the residual carbon and I-TEQ value of the fly ash collected from the (a) cyclone and (b) baghouse.

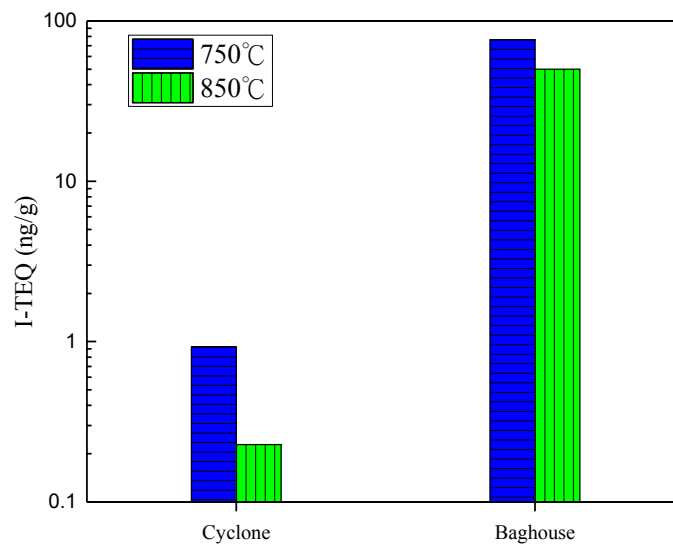


Fig. 8. I-TEQ value of the fly ash from the cyclone and baghouse after burning feeding material C at bed temperatures of 750 and 850°C.

Fig. 9 presents an image of the bottom ash with and without added clay. There are clear differences between these two types of bottom ash. Fig. 9(a) exhibits agglomerated fly ash particles, and Fig. 9(b) exhibits a carbonaceous material produced by incomplete combustion at a high temperature. The strength of the pellet can be enhanced by the addition of clay, which produces aggregated ash in the bed that is discharged as bottom ash. Thus, this indicates the benefit of adding clay to the pelletized fly ash.

The total amount of feeding material in each test run was 15 kg. The fly ash collected from the cyclone and baghouse, and the bottom ash separated from the bed material were weighed and recorded after each run, and the data are listed in Table 6. The total toxicity of the feeding

materials and residue after burning was estimated from the dioxin concentration and quantity of fly ash listed in Table 4 using Eqs. (2) and (3):

$$PCDD/F_{S_{input}} \text{ (ng I-TEQ)} = W_{RM} \times I-TEQ_{RM} \quad (2)$$

$$PCDD/F_{S_{output}} \text{ (ng I-TEQ)} = W_{BFA} \times I-TEQ_{BFA} + W_{CFA} \times I-TEQ_{CFA} + W_{BA} \times I-TEQ_{BA} \quad (3)$$

W_{RM} : Quantity of feeding material in each test run, i.e., 15 kg.

W_{BFA} : Quantity of fly ash collected from the baghouse in each test run, g.

W_{CFA} : Quantity of fly ash collected from the cyclone in each test run, g.

W_{BA} : Quantity of bottom ash in each run, g.

$I-TEQ_{RM}$: I-TEQ value of the feeding material, $ng\ g^{-1}$.

$I-TEQ_{BFA}$: I-TEQ value of the fly ash collected from the baghouse, $ng\ g^{-1}$.

$I-TEQ_{CFA}$: I-TEQ value of the fly ash collected from the cyclone, $ng\ g^{-1}$.

$I-TEQ_{BA}$: I-TEQ value of the bottom ash, $ng\ g^{-1}$.

The results are shown in Fig. 10. The total toxicity of the feeding materials before treatment ranged from 258,000 to 349,500 $ng\ I-TEQ$, while the total toxicity of the residues after burning ranged from 15,700 to 32,200 $ng\ I-TEQ$. The

destructive rate of the total dioxin toxicity ranged from 88 to 95%. It has previously been demonstrated that the dioxin concentration of fly ash is positively correlated with the amount of residual carbon. The experimental results showed that the residual carbon content of the FA feeding material was lower than that of the other feeding materials. Therefore, the dioxin concentration of the fly ash was relatively low, and the removal efficiency was high. Adding coal to the pelletized fly ash increased the amount of residual carbon, which reduced the dioxin removal efficiency.

From the experimental data obtained in this study,



Fig. 9. Images of bottom ash (a) with and (b) without clay.

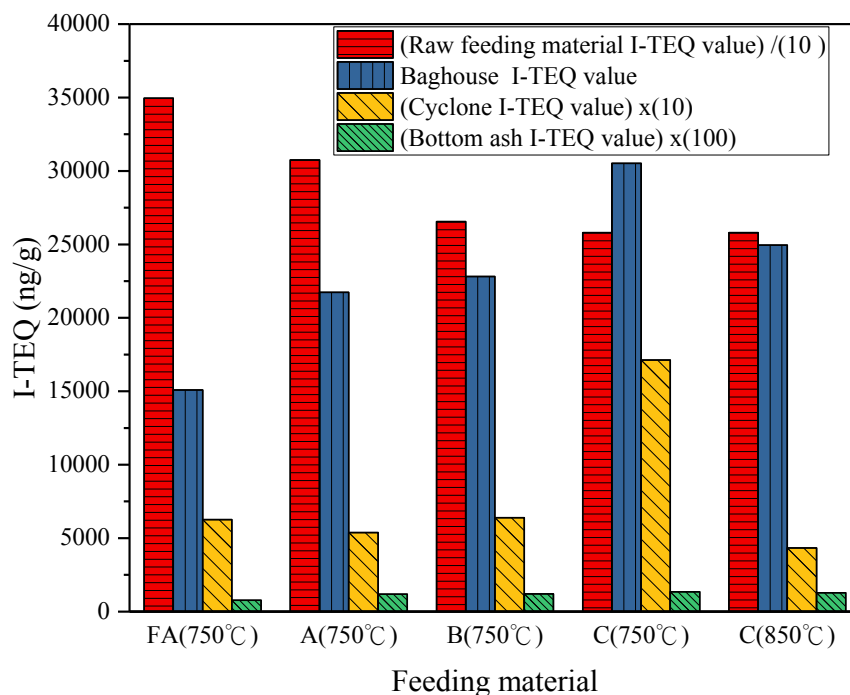


Fig. 10. Comparison of the I-TEQ values of the feeding materials and the residues collected from the baghouse, cyclone, and bottom ash after burning.

adding clay to the pellets can increase the quantity of bottom ash, and the destructive efficiency can be increased with a higher bed temperature. To obtain a higher destructive efficiency, further research should be conducted to develop a suitable pelletized fly ash formula.

PCDD/F Homolog and Congener Profiles of the Fly Ash

Fig. 11(a) shows the congener profiles of the seventeen PCDD/Fs, which were similar for each experimental group. The most prevalent congeners in the fly ash were 1,2,3,4,6,7,8-HpCDF (9–35%), OCDD (6–31%), and OCDF (6–22%), which was similar to the results of Liu *et al.* (2017) and Ma *et al.* (2019). The most prevalent distributions before and after burning were HpCDD, OCDD, HxCDF, HpCDF, and OCDF. According to the data shown in Fig. 11(a), the homologs of 1,2,3,4,6,7,8-HpCDF (9–21%), OCDF (6–13%), 1,2,3,4,6,7,8-HpCDD (7–15%), and OCDD (6–13%) are prevalent in the raw materials. Shin and Chang (1999) investigated the distribution characteristics of fly ash homologs produced by a waste incinerator in Korea, and the results showed that both PCDD and PCDF existed with a higher degree of chlorination. This agreed with the results obtained in this study. Based on the results from sixteen incinerators in Taiwan, Chang *et al.* (2011) found that OCDD was the most abundant type of PCDD, followed by HpCDD. This is also consistent with the experimental results of this study. In summary, most studies demonstrated that PCDD is present in fly ash with

a high number of chlorine bonds, while PCDF is present with a lower degree of chlorination.

For the raw material, the congener profiles of the 17 PCDD/Fs are evenly distributed. After burning, a significant change can be observed, as the homologs of 1,2,3,4,6,7,8-HpCDF (25–35%), OCDF (16–22%), and OCDD (11–31%) in the fly ash collected from the baghouse or cyclone are more prevalent than the homologs of 1,2,3,4,6,7,8-HpCDD (4–9%). The proportion of 1,2,3,4,6,7,8-HpCDF is higher in the fly ash collected from both the cyclone and baghouse. This can be attributed to the higher thermal stability of these homologs. The PCDFs are less prone to oxidative degradation than PCDDs, and more stable in fly ash (Luijk *et al.*, 1994). As shown in Fig. 11, the prevalence of other types of PCDD congeners decreased greatly, and 1,2,3,4,6,7,8-HpCDF, OCDF, and OCDD were reduced to a lesser extent than the other homologs, resulting in higher percentages of these congeners after burning.

The greatest contributor to the total I-TEQ values was 2,3,4,7,8-PeCDF (36.72–38.66%), as shown in Fig. 11(b). This agrees with the results of previous studies (Han *et al.*, 2017; Hsieh *et al.*, 2018; Lin *et al.*, 2018; Zhan *et al.*, 2018). The second-greatest contributor was HxCDF homologs (30–50%). For the raw material, most of the total toxicity originates from PCDF, and a small amount originates from PCDD. In fly ash collected from the cyclone and baghouses, most of the toxicity originates from PCDF.

The results of this study were compared in Fig. 12 with

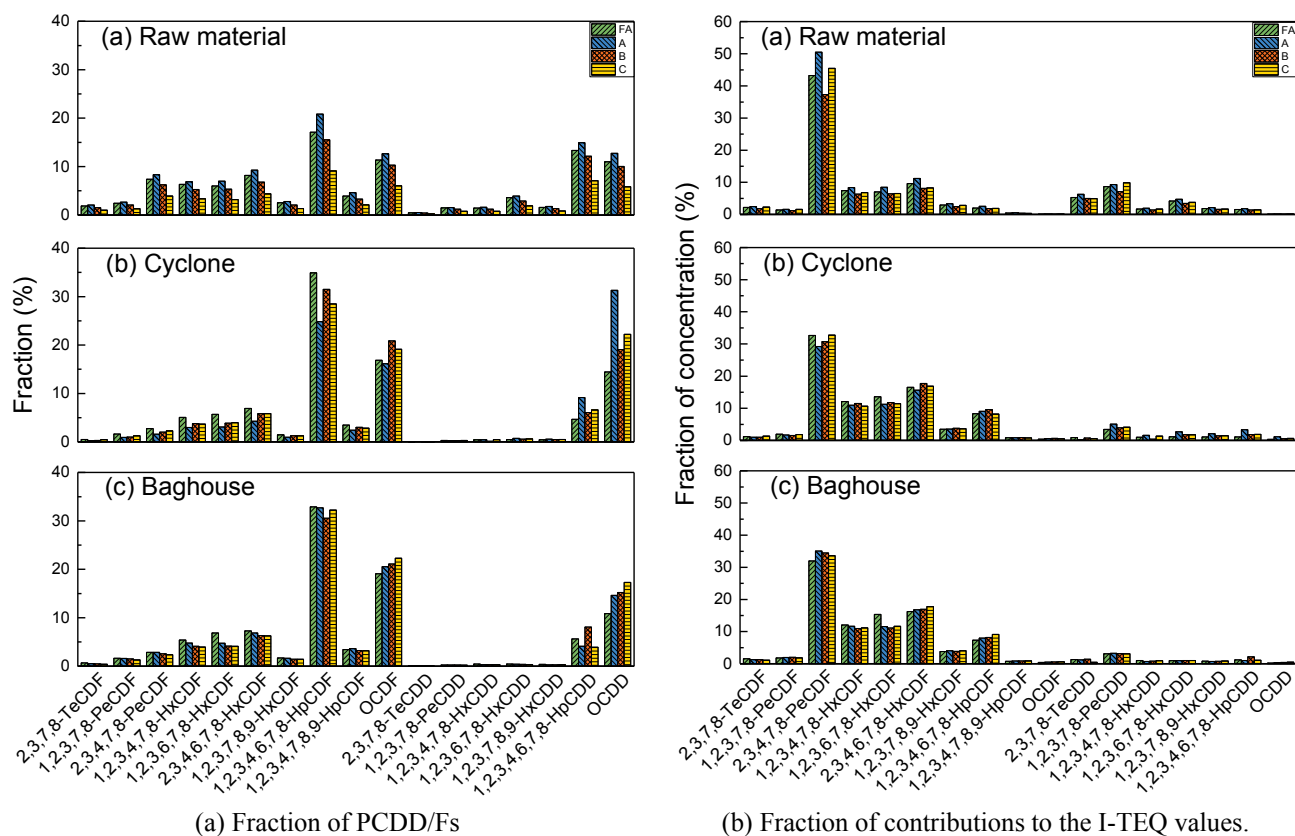


Fig. 11. Congeners of the 2,3,7,8-substituted PCDD/Fs of the fly ash obtained from the combustion of various feeding materials at a bed temperature of 750°C; (a) fraction of PCDD/Fs, (b) fraction of their contributions to the I-TEQ values.

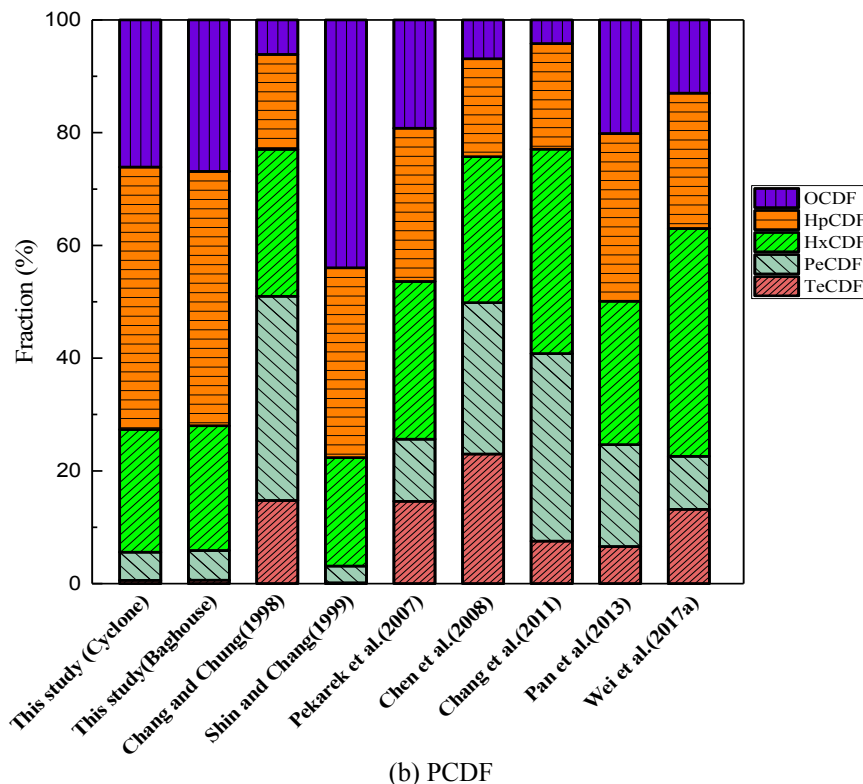
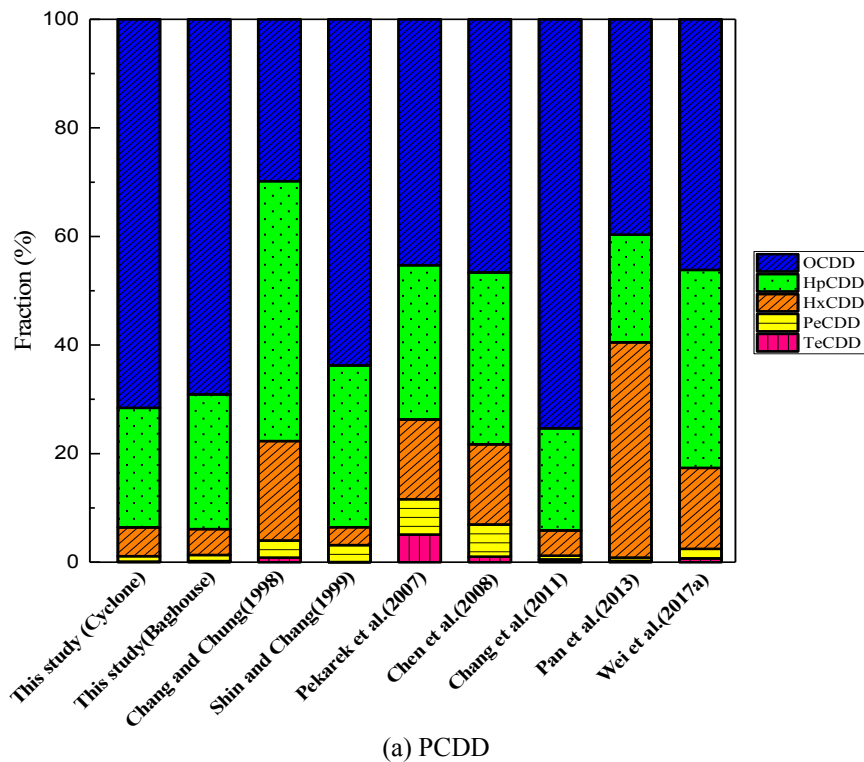


Fig. 12. Comparison of the distribution of PCDD/Fs homologs in fly ash among the results of various studies. (a) PCDD and (b) PCDF.

those obtained from previous studies (Chang and Chung, 1998; Shin and Chang, 1999; Pekarek *et al.*, 2007; Chen *et al.*, 2008; Chang *et al.*, 2011; Pan *et al.*, 2013; Wei *et al.*, 2017a). According to the data shown in Fig. 12 the

distribution of the PCDD homologs was dominated by OCDD, while the dominant contributors to the PCDF distribution varied. Other than the combustion of fuels, this may have been caused by the injection of activated carbon

into the flue gas for the purpose of incineration. In the absence of activated carbon, low-chlorinated PCDD and PCDF were not easily adsorbed by the fly ash in the baghouse, resulting in a high chloride bond distribution in PCDD and PCDF. After the injection of activated carbon, the concentration of low-chlorine bonds adsorbed on the activated carbon increased. The distribution of the PCDF homologs changed significantly due to *de novo* synthesis

CONCLUSIONS

In this study, a bench-scale fluidized bed combustor was used to re-burn dioxin-containing pelletized fly ash. From the experimental data obtained, we can draw the following conclusions:

1. The dioxin concentrations of the fly ash collected from the cyclone (0.326–0.926 ng g⁻¹) was much lower than that collected from the baghouse (50.3–70.3 ng g⁻¹), and they met the regulations of the Taiwan EPA (< 1 ng g⁻¹). The total dioxin toxicity can be reduced by 88–95% after re-burning. *De novo* synthesis did not occur at the bottom of the cyclone. The temperatures in the post-combustion zone were suitable for *de novo* synthesis, resulting in high dioxin concentrations in the fly ash collected from the baghouse. The addition of a cyclone to the system can not only reduce the quantity of fly ash in the baghouse, but also reduce the total dioxin toxicity. The addition of a quench tower is suggested to reduce the *de novo* synthesis of dioxins in the post-combustion zone.
2. The amount of residual carbon in the fly ash is strongly positively correlated with the dioxin concentration. Adding coal to the pelletized fly ash leads to an increase in the incomplete particulate matter combustion products, resulting in an increase in the concentration of dioxins. A higher temperature is recommended to reduce the amount of residual carbon for lower dioxin formation.
3. The pellet strength can be enhanced by adding clay, which leads to the formation of aggregated ash in the bed that is then discharged as bottom ash. Erosion and friction between the pellets and bed material lead to breakage, resulting in an increase in fly ash. A bed temperature of 800°C is suitable for sintering pellets with a higher hardness. In order to reduce the fly ash generated in the combustion chamber during re-burning, further research is required to develop a more suitable pellet formula.
4. The profiles of the distributions of PCDD/Fs homologs in the fly ash collected from the baghouse in each experiment were generally similar. The most prevalent congeners in the ash were 1,2,3,4,6,7,8-HpCDF, OCDD, and OCDF. The predominant contributor to the I-TEQ g⁻¹ was 2,3,4,7,8-PeCDF.

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