



Thermal Formation of Polybrominated Diphenyl Ethers in Raw and Water-Washed Fly Ash

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ABSTRACT

To the best of our knowledge, the formation mechanisms of PBDEs in the combustion system, fly ash, as well as their relation with temperature, have never been investigated. In this study, the formation mechanisms of PBDEs occurring in the processes of heating fly ash have been investigated using a laboratory furnace and graphite crucibles in order to clarify the effects of temperature. A dramatic increase in total PBDE content in the heated water-washed fly ash (3.33 ng/g) occurred, 17.7 times higher than that of the unheated one (0.188 ng/g), when the heating temperature reached 400°C. We speculate that at this temperature, PBDEs form through de novo synthesis as well as heterogeneous solid-phase reactions of the precursor mechanisms. Another dramatic increase in PBDE content in the water-washed fly ash occurred in the range 650°C–850°C, due to the enormous amounts of brominated aromatic precursors that are formed, and then condensed or are adsorbed onto the surface of the fly ash during the cooling down to 400°C, where PBDEs form through de novo synthesis and precursor mechanisms. The raw fly ash contains more alkaline compounds such as sodium and calcium compounds than the water-washed fly ash, which are inhibitors for the formation of precursors and PBDEs. Consequently, the water-washed fly ash has much greater PBDE formation potential than the raw fly ash.

Keywords: PBDEs; Fly ash; Formation; de novo; Precursor.

INTRODUCTION

Polybrominated diphenyl ethers (PBDEs), structurally similar to polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) and polychlorinated biphenyls (PCBs), have been extensively used as brominated flame retardants (BFRs) in a large variety of consumer products. However, due to health risks, commercial penta-BDE and

octa-BDE mixtures were banned within the European Union in 2004. In addition, the European Court of Justice ruled against the exemption of deca-BDE from the RoHS directive and decided that its use must be phased out by July 1, 2008 (European Court of Justice, 2008).

Our recent study found that besides controlling PBDE mixtures in commercial products, mitigation of PBDE emissions from combustion sources is required to reduce environmental PBDE levels and human exposure (Wang *et al.*, 2010b). The PBDE concentrations in the stack flue gases of combustion sources, including waste incinerators, metallurgical processes, power-heating systems and so on, range from 8.07 to 469 ng/Nm³ (Wang *et al.*, 2010b). Combustion sources possess high concentrations, huge flow rates of stack flue gases and in great numbers, revealing that they are not only major PCDD/F emission sources, but also important PBDE emitters to the

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atmosphere (Wang *et al.*, 2010b). But not like PCDD/Fs, comprehensive research have been conducted into their important sources, such as waste incinerators (Wang *et al.*, 2003b; Wang *et al.*, 2007; Wang and Chang-Chien, 2007; Lin *et al.*, 2010a; Wang *et al.*, 2010d), iron ore sintering (Wang *et al.*, 2003c; Wang *et al.*, 2008b), power plants (Lin *et al.*, 2007; Lin *et al.*, 2010b; Wang *et al.*, 2010e; Wu *et al.*, 2010), boilers (Chen *et al.*, 2011), electric arc furnaces (EAFs) (Lee *et al.*, 2004; Lee *et al.*, 2005; Li *et al.*, 2008; Wang *et al.*, 2010c; Chiu *et al.*, 2011), secondary aluminum smelters (ALS) (Lee *et al.*, 2005; Li *et al.*, 2007), crematories (Wang *et al.*, 2003a), vehicles (Chuang *et al.*, 2010a; Chuang *et al.*, 2010b), and the open burning of rice straw (Lin *et al.*, 2007) and wood (McNamara *et al.*, 2011). Furthermore, intensive studies have conducted to identify the formation mechanisms of PCDD/Fs that occur within the combustion system. Although we found the occurrence of PBDEs in the stack flue gases of sinter plants, revealing that PBDEs can form during the combustion processes through similar formation conditions of PCDD/Fs (Wang *et al.*, 2010c), the detailed formation mechanisms of PBDEs still need to be clarified to be able to control PBDE emissions from combustion sources.

Fly ash is typically composed of an alumina-silicate construct, with 20% of the weight of fly ash being carbon, and 5 to 10% being silicon, inorganic chlorides, sulfur and potassium (NATO, 1988). The particles of fly ash have specific surface areas in the range of 200 to 400 m²/kg (NATO, 1988). Furthermore, fly ash contains transition metal compounds and incompletely combusted products which are necessary for the formation of PCDD/Fs, PBDD/Fs, PCBs and polychlorinated diphenyl ethers (PCDEs) (Wang *et al.*, 2003b; Wang *et al.*, 2010a; Liu *et al.*, 2011). The formation of PCDD/Fs which is relevant to the precursor mechanisms and de novo synthesis, that is, the interaction of chlorine, precursors, fly ashes, metal catalysts, and a favorable range of temperatures, have been well studied. The temperature is perhaps the most important factor in PCDD/F formation (Addink *et al.*, 1991).

To the best of our knowledge, however, the formation mechanisms of PBDEs in the combustion system, fly ash, as well as their relation with temperature have never been investigated. Although air pollution control devices (APCDs), such as bag filters (Lin *et al.*, 2008; Li *et al.*, 2010; Yamada *et al.*, 2011), electrostatic precipitators (EP) (Ruttanachot *et al.*, 2011), activated carbon injection (Li *et al.*, 2010) and other measures (Lai *et al.*, 2007; Wang *et al.*, 2008a), could be used to remove pollutants, clarifying the mechanisms of PBDEs may lead to methods that prevent their formation and their release into the environment. In this study, the formation mechanisms of PBDE occurring in the processes of heating fly ash have been investigated using a laboratory furnace with graphite crucibles in order to clarify the effects of temperature. Two kinds of fly ash, raw and water-washed, are selected to conduct the experiments of PBDE thermal formation to clarify the influence of the bromide and calcium contents in the fly ash on PBDE formation.

MATERIALS AND METHODS

Fly Ash Samples and Thermal Process

Fly ash collected from the bag filter of a medical waste incinerator equipped with a dry scrubber and bag filter as air pollution control devices was used to investigate the formation mechanisms of PBDEs occurring in the processes of heating fly ash. The fly ash samples were put in a graphite crucible and placed in the furnace. The experiments were performed by increasing the furnace temperature by 5 °C/min from room temperature. For water-washed fly ash, the heating temperature was maintained at 50, 100, 150, 200, 250, 300, 400, 500, 600, 650, 750, 850, 900, 1000, 1100, 1200, 1300, 1350 and 1450°C, respectively, for 1 h, and then cooled down to room temperature naturally, while for raw fly ash, the heating temperature was kept at 50, 100, 150, 200, 250, 300, 400, 500, 600, 650, 750 and 850°C, respectively.

Analytical Procedures of PBDEs

The analyses of thirty PBDE congeners were performed following U.S. EPA Method 1614. PBDE internal standards were spiked to the samples before Soxhlet extraction with toluene, and were used to monitor the extraction and cleanup procedures. The detailed analytical procedures for PBDEs are given in our previous works (Wang *et al.*, 2003a; Wang *et al.*, 2010c). In brief, the concentrated extract was treated with concentrated sulfuric acid, and this was followed by a series of sample cleanup and fractionation procedures, including a multi-layered silica column and alumina column. The eluate was concentrated to approximately 1 mL and transferred to a vial. The concentrate was further concentrated to near dryness, using a stream of nitrogen. Immediately prior to analysis, the standard solution for recovery checking was added to the sample.

Instrumental Analysis of PBDEs

A high-resolution gas chromatograph/high-resolution mass spectrometer (HRGC/HRMS) was used for PBDE analyses. The HRGC (Hewlett-Packard 6970 Series gas chromatograph, CA) was equipped with a DB-5HT capillary column ($L = 15$ m, i.d. = 0.25 mm, film thickness = 0.1 μm) (J & W Scientific, CA), and with a splitless injection (injector at 250°C, transfer glass line at 280°C). The injection volume was 2 μL. The column oven temperature program was set according to the following parameters: initial oven temperature began at 100°C (held for 4 min), then increased at 40 °C/min to 200°C (held for 3.5 min), and finally increased at 10 °C/min to 325°C (held for 2.5 min). Helium at a flow rate of 1.0 mL/min was the carrier gas. The HRMS (Micromass Autospec Ultima, Manchester, UK) mass spectrometer was equipped with a positive electron impact (EI+) source. The selected ion monitoring mode was used with a resolving power of 10,000. Detailed instrumental analysis parameters of the PBDEs are given in our previous work (Wang and Chang-Chien, 2007; Wang *et al.*, 2010a; Wang *et al.*, 2010c).

Analyses of Bromide and Calcium in the Ashes

For bromide measurement, the ash sample (3–5 g) was extracted with deionized water for 48 h. The extract was then separated by centrifugation, and the bromide concentrations were determined by ion chromatography. For the analyses of metal contents in the ash samples, the ash sample (0.5 g) was pulverized, and then hermetically digested with an acid mixture in a Teflon vessel by a microwave digester. The digested mixture was added up to 25 mL with deionized water, and then analyzed by inductively coupled plasma mass spectrometry (ICP-MS) to determine the metal contents.

RESULTS AND DISCUSSION

Bromide, Calcium and PBDE Contents in the Raw and Water-Washed Fly Ash

Fig. 1 illustrates the contents and congener profiles of PBDEs in the raw and water-washed fly ash. The PBDE content in the raw fly ash of the MWI is comparable to those in the superheater, economizer, dry scrubber and bag filter ashes (0.332–1.87 ng/g) of the municipal solid waste

incinerators with 20% of the feeding waste as industrial waste (Wang et al., 2010a). The PBDE content decreased from 0.750 ng/g to 0.188 ng/g after water washing treatment, revealing the PBDE reduction resulted from the finer ash being dissolved or suspended in the water, and later being removed. Furthermore, after water washing treatment, the fraction of BDE-209 in the fly ash dramatically decreased from 76% to 39%, suggesting the combustion-originated ash, which should be finer than ash from waste incompletely incinerated, contained more BDE-209.

The contents of inorganic bromide and calcium in the raw fly ash also reduced from 1.61 and 26.3 mg/g to 0.0483 and 1.32 mg/g after water washing treatment, that is, there was about 97% and 95% removal of the original contents, respectively. (Zhu et al., 2008) found that approximately 15% of the chlorine in the raw fly ash of MSWI was in the form of NaCl, 10% was in the form of KCl, 51% was CaCl₂, and the remainder was in the form of Friedel’s salt. In ash washing experiments, the mole percentage and the amount of soluble chlorides including NaCl, KCl and CaCl₂ decreases quickly with the increase

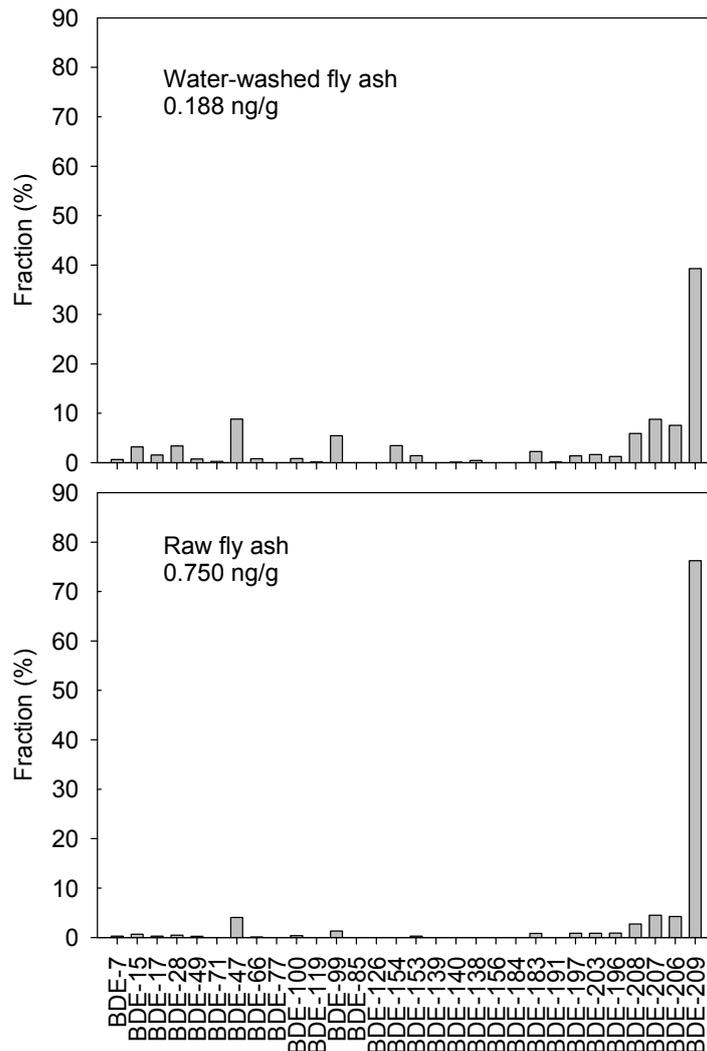


Fig. 1. The contents and congener profiles of PBDEs in the water-washed and raw fly ash.

in liquid to solid (L/S) ratio or washing frequency. However, those of insoluble chlorides decrease slower (Zhu *et al.*, 2010).

Thermal Formation of PBDEs Occurring in the Water-Washed Fly Ash

The trends of the PBDE homologue contents in the water-washed fly ash under various heating temperatures are illustrated in Fig. 2(a), while those of Σ_{2-8} BDE (the summation of di-BDE to octa-BDE contents), nona-BDE and deca-BDE (BDE-209) at five different temperature ranges of $< 300^\circ\text{C}$, 400°C , 500°C – 600°C , 650°C – 850°C and $> 900^\circ\text{C}$ are shown in Fig. 2(b). Furthermore, Fig. 2(c) compares the trends of the ratios of the PBDE contents in the heated water-washed fly ash to those of the unheated one. The total PBDE contents in the heated water-washed fly ash were between 0.184–0.296 ng/g within low temperature range (50°C – 300°C), close to that of unheated water-washed fly ash (0.188 ng/g).

A dramatic increase in total PBDE content in the heated water-washed fly ash (3.33 ng/g) occurred, 17.7 times

higher than that of the unheated one, when the heating temperature reached 400°C (Fig. 2). Only a few studies have also observed PBDE formation during combustion system (Wang *et al.*, 2010a). Our previous study found that when the flue gases of municipal solid waste incinerators (MSWIs) flowed from the superheaters to the economizers, the PCDD/F contents in the ashes of the economizers whose the operational temperatures (339°C – 396°C) were within the region of optimal PCDD/F formation were much higher than those in the ashes of the superheater whose temperatures (480°C – 537°C) were not favorable for PCDD/F formation. The PBDD/F and PBDE contents in the ashes of the economizers were also six times higher than those in the ashes of the superheater, suggesting that de novo syntheses could also occur among PBDD/Fs and PBDEs (Wang *et al.*, 2010a).

We speculate that at this temperature range the PBDE formation pathway is similar to the de novo synthesis of PCDD/Fs, that is, PBDEs form the oxidative breakdown of macromolecular carbon structures of fly ash catalyzed by a transition metal in the presence of bromine. PBDEs are

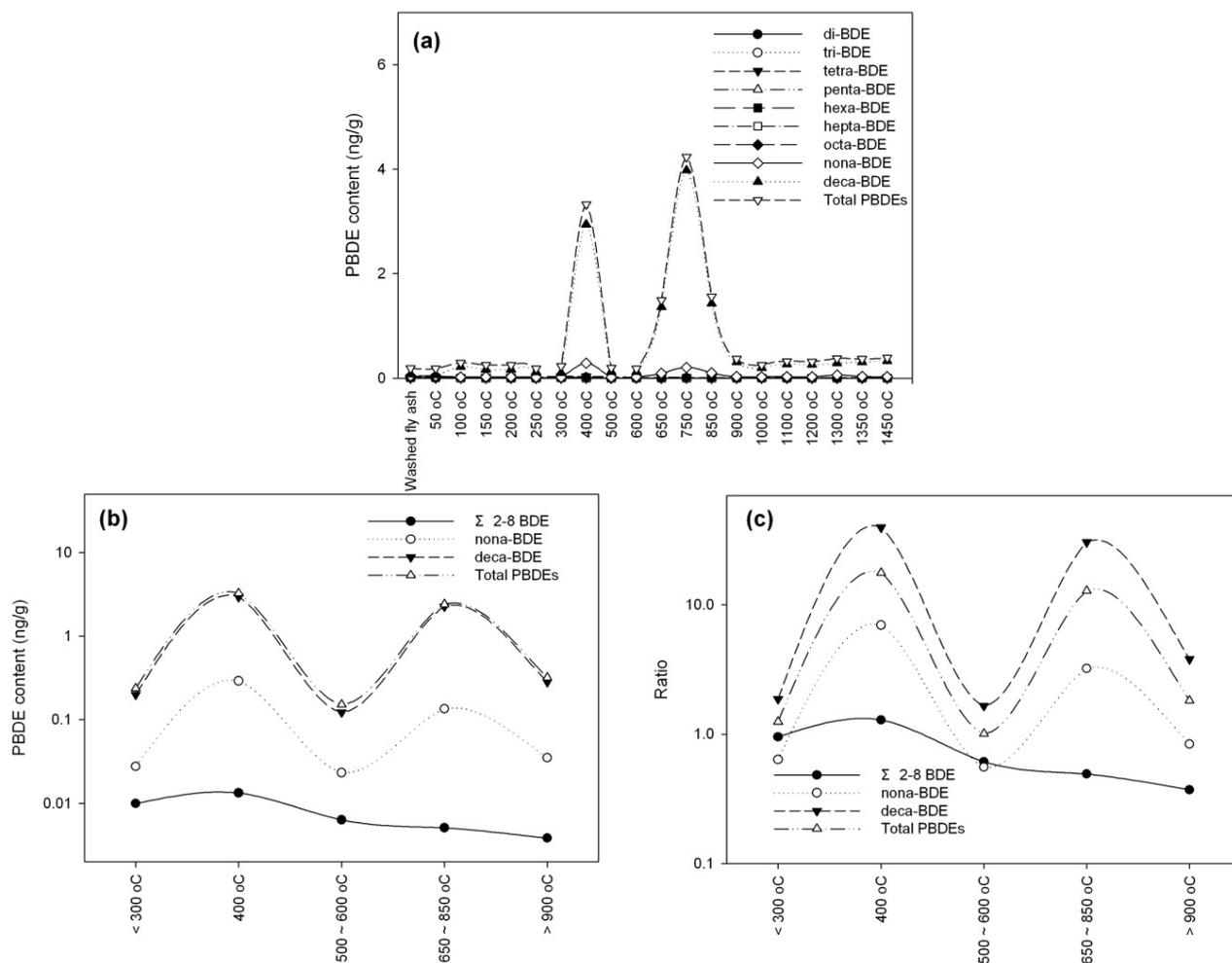


Fig. 2. (a) The trends of the PBDE homologue contents in the heated water-washed fly ash under various temperatures. (b) The trend of the contents of Σ_{2-8} BDE, nona-BDE and deca-BDE in the heated water-washed fly ash at five different temperature ranges. (c) The trends of the ratios of the PBDE contents in the heated water-washed fly ash to that in the unheated one.

formed partially from the aromatic carbon-oxygen functional groups embedded in the carbon skeleton. Furthermore, PBDEs could also form from precursor mechanisms, in other words, heterogeneous reactions on the reactive fly ash surface involving aromatic precursor compounds and bromine promoted by a transition metal catalyst. The precursor compounds include brominated benzenes, brominated phenols, phenol, and benzene.

Furthermore, at 400°C the contents of Σ_{2-8} BDE, nona-BDE and deca-BDE were 1.3, 7.0 and 39 times higher than those of the unheated water-washed fly ash, respectively (see Fig. 2(c)). Deca-BDE has the greatest formation compared to other PBDE congeners. Similarly, our previous study found that PBDE and PBDD/F formations during the combustion processes prefer highly brominated congeners (Wang *et al.*, 2010b; Wang *et al.*, 2010c). PBDEs are available as precursors for the formation of PBDFs in the combustion of sinter plants, and from a mechanistic point of view, the formation of PBDFs from PBDEs requires only an intra-molecular elimination of Br₂ or HBr (Weber and Kuch, 2003).

This phenomenon has also been observed in the formation of PCDEs (Liu *et al.*, 2011). The PCDE formation was investigated using laboratory-scale flow reactors under air and under nitrogen at 350°C, and the results showed that PCDEs formed from precursors on the fly ash surface. OCDE was the dominant PCDE homologue product formed on the surface of SiO₂, whereas deca-CDE was the predominant product formed on the SiO₂/FeCl₃ and SiO₂/CuCl₂ surfaces. The PCDE homologue distribution shifted towards the more chlorinated species in the presence of FeCl₃ and CuCl₂ (Liu *et al.*, 2011).

When the heating temperature increased to 500°C–600°C, the total PBDE contents in the water-washed fly ash decreased to 0.182–0.198 ng/g, but rose again dramatically to 1.49–4.23 ng/g in the temperature range of 650°C–850°C. The highest PBDE content (4.23 ng/g) that occurred at 750°C was 22.5 times larger than that of the unheated water-washed fly ash. We speculate that enormous amounts of key brominated aromatic precursors for PBDEs are formed in the higher-temperature region (about 750°C), and subsequently condense or are adsorbed onto the surface of fly ash during the cooling down to 400°C, where PBDEs form through de novo synthesis as well as heterogeneous solid-phase reactions of the precursor mechanisms. Similar phenomena concerning the formation of chlorinated precursor of PCDD/Fs have been found in Froese and Hutzinger (1996), with chlorinated benzene and phenols in fly ash catalyzing heterogeneous reactions with ethylene and ethane at temperatures of 300°C–600°C in the presence of HCl, oxygen, and a metal catalyst. Reaction with fly ash at 600°C formed hexachlorobenzene that was 1000 times higher than those at 400°C and 500°C, with similar results for chlorophenols. No chlorobenzene congener precursors were formed from ethylene and ethane at 300°C.

When the operational heating temperature of the water-washed fly ash rose to 900°C, the PBDE contents decreased to 0.37 ng/g, and remained at the level of

0.253–0.389 ng/g at 900°C–1450°C. At this high temperature range, the organic carbon and precursors in the fly ash can achieve more complete combustion, so the PBDE formation during cooling down is suppressed. At this high temperature range, although the destruction of the original PBDEs in the fly ash also could reduce the PBDE contents, we found that the suppression of PBDE formation is mainly responsible for the observed phenomena by comparing the PBDE contents in the heating temperature range of 500°C–600°C.

Thermal Formation of PBDEs Occurring in the Raw Fly Ash

The trends of the PBDE homologue contents in the raw fly ash from 50°C–850°C are illustrated in Fig. 3(a), while those of the contents of Σ_{2-8} BDE, nona-BDE and deca-BDE are shown in Fig. 3(b). Furthermore, the trends of the ratios of the PBDE contents in the heated raw fly ash to that of the unheated one are depicted in Fig. 3(c). When compared to the trends of the heated water-washed fly ash (see Fig. 2), we find that they are basically similar, with PBDE formation at heating temperatures of 400°C and 650–850°C. However, one of the differences is that for raw fly ash at 100°C, Σ_{2-8} BDE contents increased with the decline of deca-BDE, revealing that the debromination of the original PBDEs occurred due to its higher PBDE content than that of water-washed fly ash. Another difference is the formation of Σ_{2-8} BDE and deca-BDE at a heating temperature of 150°C–200°C found for the raw fly ash. We speculate that at this temperature range, the loss of water content and the decomposition of some combustible organic compounds further concentrated the bromine, precursors and catalytic metal contents in the raw fly ash, which were originally higher than those in the water-washed fly ash. This concentration could lower the temperature of PBDE formation, and so additional PBDE formation was observed.

Furthermore, the most significant difference between the results of heating the raw fly ash and the water-washed fly ash is that the latter possessed much greater PBDE formation potential. (Fujimori *et al.*, 2010) reported that more than 95% of the formation of chlorinated aromatic compounds, such as chlorobenzenes and PCBs in MSWI fly ash, could be inhibited by the addition of NaOH, Ca(OH)₂, or NaHCO₃ in comparison with formation in heated raw MSWI fly ash. Our previous study also found that adding CaO to adsorb HBr inhibits the formation of polybrominated dibenzo-p-dioxins and dibenzofurans (PBDD/Fs) by more than 90% (Lai *et al.*, 2007). Consequently, we think that most of the alkaline compounds, which are inhibitors for the formation of precursors and PBDEs, such as sodium and calcium compounds, are removed by water washing treatment, and thus the thermal formation of PBDEs occurring in the water-washed fly ash is greater than that in the raw fly ash. Although water washing treatment also removed 97% of inorganic bromide in the raw fly ash, the retained bromide (0.0483 mg/g) and organic bromine contents are still sufficient for the formation of PBDEs and their precursors.

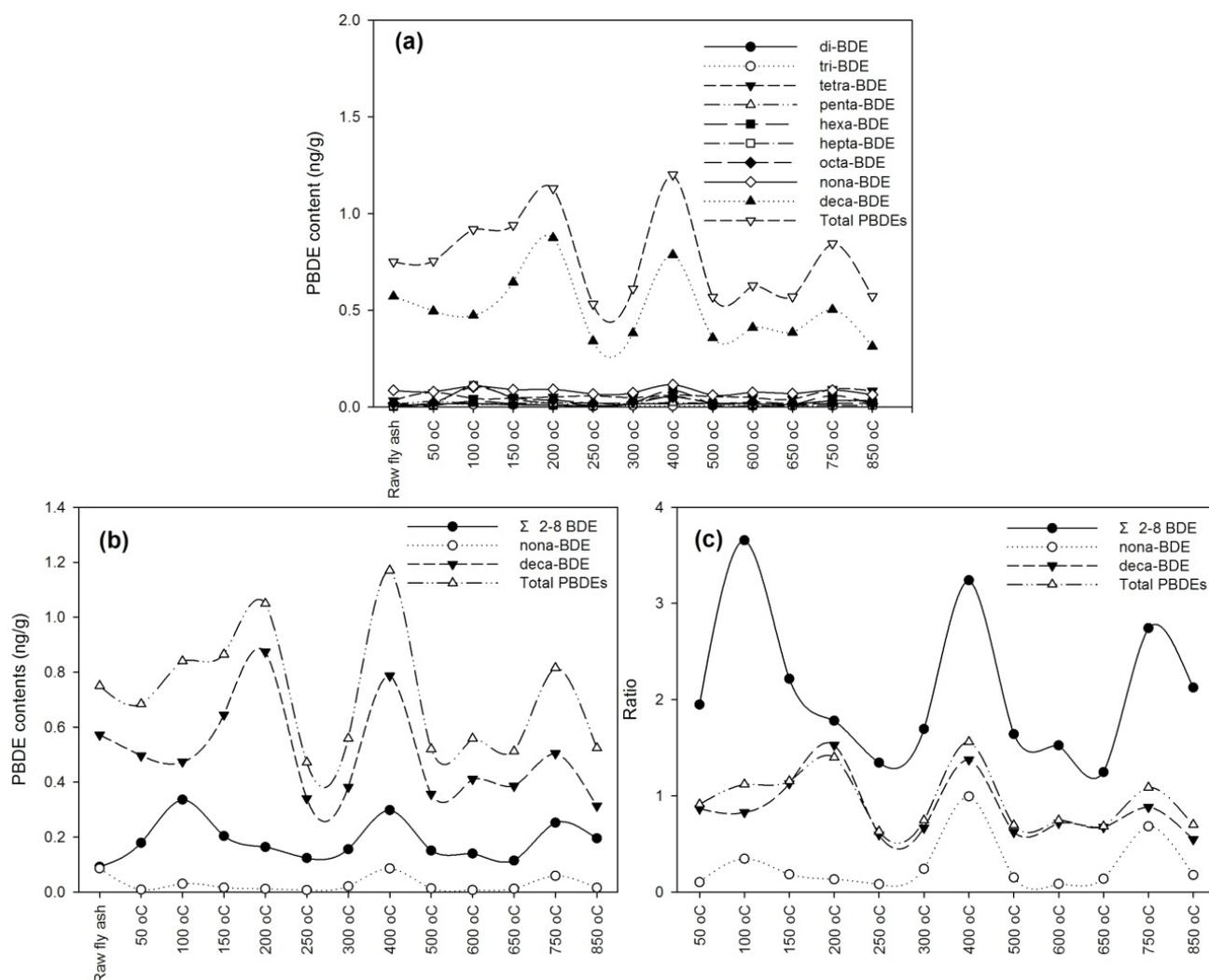


Fig. 3. (a) The trends of the PBDE homologue contents in the heated raw fly ash under various temperatures. (b) The trend of the contents of Σ_{2-8} BDE, nona-BDE and deca-BDE in the heated raw fly ash under various temperatures. (c) The trends of the ratios of the PBDE contents in the heated raw fly ash to that in the unheated one

CONCLUSIONS

The trends of PBDE thermal formation in the heated water-washed and raw fly ash are basically similar. Both see PBDE formations at heating temperatures of 400 °C and 650–850 °C, and the most significant difference is that the water-washed fly ash possessed much greater PBDE formation potential, due to its most removal of alkaline compounds which are inhibitors for the formation of precursors and PBDEs.

At a heating temperature of 400 °C, deca-BDE has the greatest formation compared to other PBDE congeners, and this has also been observed in the formation of PCDEs when fly ash contains FeCl_3 and CuCl_2 . At temperatures of 650–850 °C, we speculate that enormous amounts of brominated aromatic precursors are formed in fly ash catalyzed heterogeneous reactions, resulting in another dramatic increase in PBDE contents through de novo synthesis and precursor mechanisms when the temperature slowly falls to 400 °C.

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