

# Emission of Brominated Pollutants from Waste Printed Circuit Boards during Thermal Treatment: A Review

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## ABSTRACT

With the improvements seen in electronic equipment in recent years, the amount of electronic waste generated has increased dramatically. Researchers must find a way to treat waste printed circuit boards (WPCBs) without harming the environment as soon as possible. The disposal of WPCBs generates many brominated pollutants, which have a great impact on human health and the surrounding environment. At present, research on the WPCB disposal process has mainly been focused on efficient resource recovery; studies of pollutant emission have been limited to chlorinated dioxins, and research on brominated pollutants is relatively limited. This paper summarizes and analyzes the brominated pollutants emitted during the recycling of waste circuit boards, summarizes the sources and paths for the generation of brominated pollutants, and describes a series of studies performed to determine the emission characteristics and impact factors for brominated pollutants. The effects of raw materials, temperature, atmosphere, and reaction time on the emission of brominated pollutants are discussed, as are three methods of controlling the pollutants. It was found that when the temperature was controlled at 850–1200°C, the reaction time was greater than 2 s, the air flow rate was 3–4.5 m s<sup>-1</sup>, and the excess air coefficient was 1–2, the impact of pollutant emissions was relatively small. It is hoped that this paper will provide help to other researchers and practitioners on WPCB recycling.

**Keywords:** Waste circuit boards, Brominated pollutants, Pollutant control, Emission characteristics, Influence factor, Generate path

## 1 INTRODUCTION

With the continuous development of society and improvement in the economic level, the demand for various electronic products has increased sharply. In addition, with the rapid development of the electronic industry and information technology, electronic products are increasingly used in various fields, and the replacement cycle is also increasingly shorter. Service lives have been gradually shortened, resulting in a sharp increase in the production of electronic waste (WEEE) (Golmohammadzadeh *et al.*, 2018). Because of the serious pollution to the environment caused by electronic waste and the need to recycle and reuse it, it must be considered when charting a sustainable development path (Moltó *et al.*, 2011). According to statistics from the United Nations Environment Programme (UNEP) (Schluep *et al.*, 2009), the global income of the electronics industry is estimated at nearly one trillion yuan per year, and the global population will produce approximately 20–50 million tons of electronic waste every year. In 2019, a record 53.6 million tons of electronic waste was generated globally, an increase of 21% in just five years, and it is

## OPEN ACCESS



Received: June 8, 2023

Revised: September 23, 2023

Accepted: October 8, 2023

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Publisher:

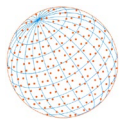
Taiwan Association for Aerosol  
Research

ISSN: 1680-8584 print

ISSN: 2071-1409 online

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expected to double by 2050 (Pariatamby and Victor, 2013; Masud *et al.*, 2019; Van Yken *et al.*, 2021). The growth rate for electronic waste is three times higher than those for other urban wastes, and it is one of the fastest growing solid waste types in the world (Cucchiella *et al.*, 2016). At present, the problem of electronic waste in China is particularly worrying. In 2015, the output of electronic waste in China reached 6 million tons, accounting for 14.4 wt.% of the total electronic waste output worldwide and ranking second in the world (Duan *et al.*, 2016).

Printed circuit boards (PCBs) constitute the foundation of modern electronic and electrical equipment and are used in almost all electronic equipment, and the content in common household appliances constitutes approximately 1–10 wt.% (Jaunich *et al.*, 2016). The printed circuit boards (WPCBs) usually contain 30% plastics (mainly organic polymers containing halogen flame retardants), 30% refractory oxides (ceramic and glass fibers mainly composed of silicon and aluminum oxide) and approximately 40% metals, almost all elements in the periodic table (Awasthi *et al.*, 2016b; Wang *et al.*, 2017a). Various heavy metals (such as lead, mercury, cadmium, hexavalent chromium) and halogen-containing substances (such as polybrominated biphenyls and polybrominated diphenyl ethers) contained in WPCBs cause serious nonpoint source pollution to the soil, groundwater and ecological environment, cause severe damage to the autonomic nervous system, blood system and kidneys and even induce carcinogenesis (Sohaili *et al.*, 2012; Hu *et al.*, 2013; Sanyal *et al.*, 2013; Tue *et al.*, 2013; Yoshida *et al.*, 2016). Additionally, the large amount of waste liquid, waste residue and waste gas generated in the process of WPCB recovery and treatment are very likely to cause serious pollution of the environment, and traditional combustion treatment methods generate dioxins (Ning *et al.*, 2017).

An increasing amount of research has been conducted in recent years. The disposal processes for WPCBs include mechanical and physical methods, pyrometallurgy, hydrometallurgy, biological treatment, etc. (Wang *et al.*, 2017b; Abdelbasir *et al.*, 2018; Li *et al.*, 2018; Nekouei *et al.*, 2018; Tatariants *et al.*, 2018; Park *et al.*, 2019). At present, the recycling and disposal processes for waste printed circuit boards (WPCBs) are mainly focused on recovering resources more effectively. Most of the research on pollution emission in the disposal process is focused on chlorinated dioxins. In this paper, the processes used in recycling waste circuit boards are considered, and studies on the emission characteristics, influencing factors and control methods for brominated pollutants have been carried out to address the problems of brominated pollutants. These studies include disposal methods for brominated dioxins, hydrogen bromide and other pollutants produced by circuit board treatment and recovery technology, and the goal is to lay a foundation for controlling the emission of brominated dioxins generated during the disposal of brominated waste.

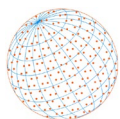
## 2 STATUS OF WASTE-PRINTED CIRCUIT BOARDS

### 2.1 Current Situation and Composition of the Circuit Board

Global informatization is advancing rapidly. With the gradual shortening of the replacement cycles for information and communications technology (ICT) devices, the amount of waste electrical and electronic equipment (WEEE) generated has increased sharply. In recent years, global WPCB output has increased by 8.7% annually. The average growth rates for Southeast Asia (10.8%) and mainland China (14.4%) are higher (Guanghan *et al.*, 2016). The output of WPCBs will continue to grow for a long time, and large-scale production of electronic products will lead to substantial waste.

WPCBs mainly come from the recycling of electronic waste and electrical equipment (WEEE). It was estimated that the total value of the raw materials contained in global electronic waste was approximately 55 billion euros in 2016, which exceeded the 2016 GDPs of most countries in the world (Baldé *et al.*, 2015). WPCBs are among the main products from scrapped electronic and electrical equipment disassembly, accounting for approximately 3–5% of the total amount of electronic waste (Faraji *et al.*, 2018; Mostafavi *et al.*, 2018). In some types of electronic equipment, WPCBs, as the core components of electronic and electrical equipment, account for a relatively large proportion of the total waste, such as 7.04% for color TVs, 18.8% for computer mainframes, and 21.3% for mobile phones (Duan *et al.*, 2011).

The printed circuit board is mainly composed of a substrate, conductive element and electronic components, etc. (Fig. 1). The substrate is prepared from copper foil, resin adhesive, glass fiber



cloth, etc., with high temperature and high pressure. The conductive pattern on the substrate supplies the connections between the electronic components (Canal Marques *et al.*, 2013; Choubey *et al.*, 2015; Gu *et al.*, 2016; Wang *et al.*, 2017b). There are many kinds of copper foil substrates. At present, brominated flame-retardant mixed epoxy resins are widely used in circuit board manufacturing.

There are various types of circuit boards, and rough measures of their components are given. WPCBs usually contain 50–70% nonmetallic components and approximately 30–50% metallic components. Among them, the nonmetallic materials in WPCBs are mainly composed of glass fibers, epoxy resins or phenolic resins, brominated flame retardants and inorganic fillers, and WPCBs contain most of the metals in the periodic table, including copper, iron, tin, nickel, lead and zinc (Table 1). WPCBs also contain small amounts of precious metals, such as gold, silver, and palladium (Schluep *et al.*, 2009; Akcil *et al.*, 2015; Fogarasi *et al.*, 2015).

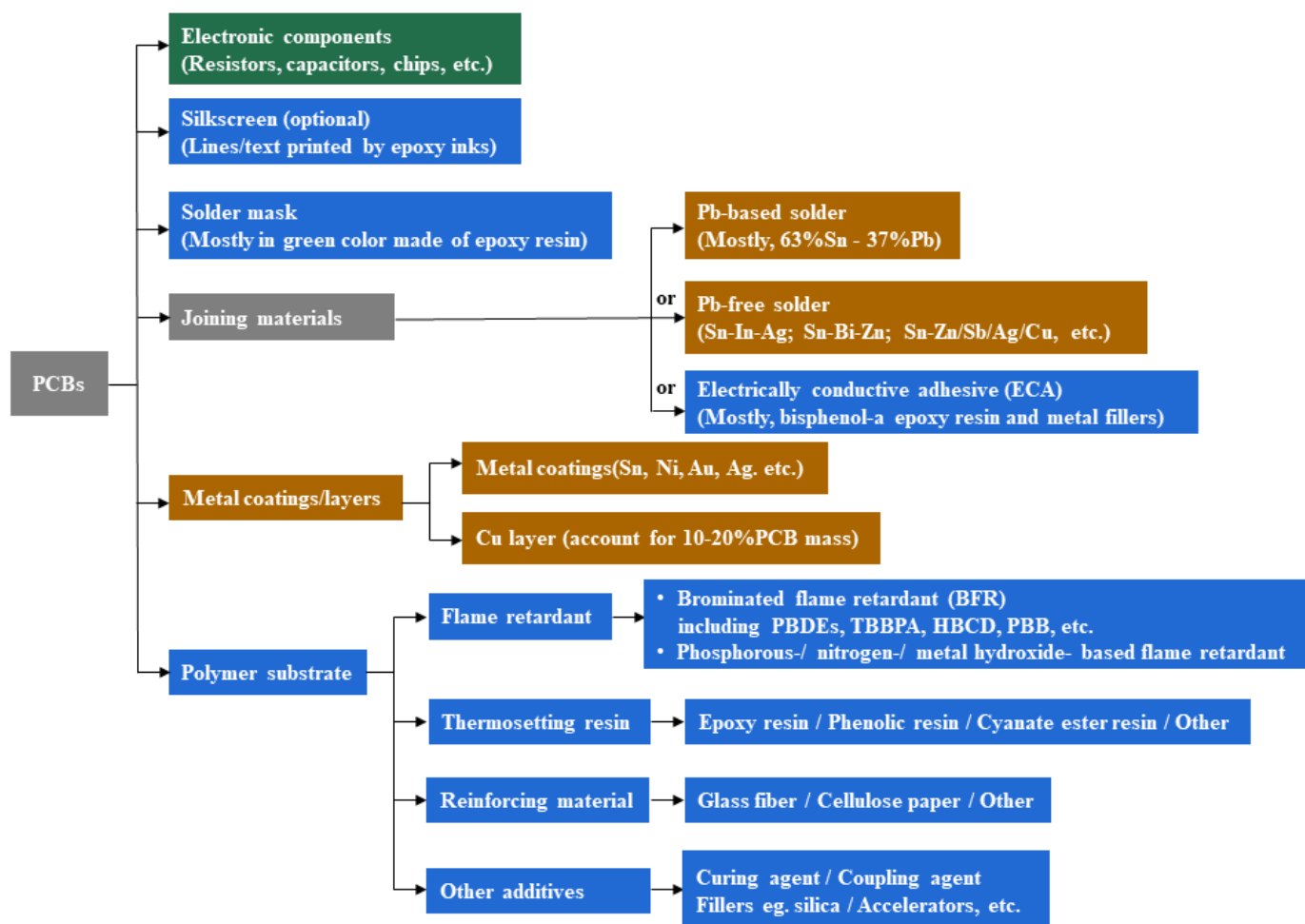
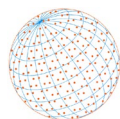


Fig. 1. Components and materials containing PCBs (Li *et al.*, 2018).

Table 1. Proportions of some heavy metals in WPCBs. (wt.%)

Al	Cu	Pb	Ni	Sn	Zn	Fe	Ref.
0.26	35.5	1.87	3.41	3.39	5.92	12.5	(Silvas <i>et al.</i> , 2015; Vats and Singh, 2015)
1.47	39.2	1.54	0.64	-	20.3	1.98	(Arshadi and Mousavi, 2014)
-	34.3	1.11	0.43	-	-	2.45	(Kumari <i>et al.</i> , 2016)
5.20	34.0	0.75	-	1.30	11.0	2.80	(Saidan <i>et al.</i> , 2012)
-	26.8	-	0.20	-	0.26	5.36	(Li <i>et al.</i> , 2015)



**Table 2.** Advantages and disadvantages of different disposal methods (Chen *et al.*, 2015; Estrada-Ruiz *et al.*, 2016; Wang *et al.*, 2017b; Abdelbasir *et al.*, 2018; Li *et al.*, 2018; Nekouei *et al.*, 2018; Tatariants *et al.*, 2018).

Technology	Advantage	Disadvantage
Mechanical physics method	<ul style="list-style-type: none"><li>➤ Low operational cost</li><li>➤ Absence of chemical usage</li><li>➤ No solvent contamination</li><li>➤ Minimal cost for consumables</li></ul>	<ul style="list-style-type: none"><li>➤ Time consuming</li><li>➤ Labor intensive process</li><li>➤ Low efficiency</li><li>➤ Incomplete metal recovery</li></ul>
Pyrometallurgy	<ul style="list-style-type: none"><li>➤ Applicability to any type of electronic waste</li><li>➤ No need for pretreatment</li><li>➤ Less process steps</li></ul>	<ul style="list-style-type: none"><li>➤ Plastic cannot be recycled</li><li>➤ High costs and energy consumption</li><li>➤ Low product purity</li></ul>
Hydrometallurgy	<ul style="list-style-type: none"><li>➤ High recovery</li><li>➤ No gaseous pollutants emission</li><li>➤ Less sensitivity to base metals</li><li>➤ Recyclable</li></ul>	<ul style="list-style-type: none"><li>➤ High cost of reagents</li><li>➤ Limited availability</li><li>➤ High toxicity</li><li>➤ Need for mechanical pretreatment of the scrap</li></ul>
Biological treatment	<ul style="list-style-type: none"><li>➤ Economic and simple</li><li>➤ Ecofriendly process</li><li>➤ Less energy requirement</li></ul>	<ul style="list-style-type: none"><li>➤ Relatively slow process</li><li>➤ Dependent on operational conditions</li><li>➤ Unstable processing effect</li></ul>

## 2.2 Disposal Technology and Environmental Hazards

At present, waste printed circuit boards are generally recycled using physical and chemical methods (Table 2). Physical recovery involves crushing and separating waste circuit boards, separating metals and nonmetals, and recycling recyclable products. Chemical methods mainly include pyrolytic recovery, solvent recovery and supercritical fluid recovery; that is, chemical methods are used to decompose WPCBs into small molecular compounds or low molecular weight resins. In addition, the methods also include the use of nonmetallic scraps of waste circuit boards as fillers for the preparation of composite materials used in recycling.

Electronic waste contains many harmful substances, such as lead (Pb), copper (Cu), chromium (Cr), tin (Sn), polybrominated biphenyls (PBBs), polybrominated biphenyl ethers (PBDEs), tetrabromobisphenol A (TBBPA), and decabromodiphenylethane (DBDPE) (Chen *et al.*, 2012). In the past, the storage and handling of WPCBs have caused very serious environmental problems and posed an enormous threat to human health (Fig. 2). Potential carcinogenic and harmful substances, such as toxic metals and persistent organic pollutants, found in electronic waste disassembly areas have received considerable attention. These pollutants accumulate readily in the surrounding dust, soil, water, atmosphere and other environmental media (Sepúlveda *et al.*, 2010; Pradhan and Kumar, 2014; Wu *et al.*, 2014; Awasthi *et al.*, 2016a; Pathak *et al.*, 2017).

Prior studies have shown that the toxicities of brominated pollutants are mainly reflected by interference with thyroid hormones, hepatotoxicity, immunotoxicity, neurotoxicity, and potential carcinogenicity (Eng *et al.*, 2019; Jing *et al.*, 2019; Wang *et al.*, 2019; Sun *et al.*, 2020). Some reports on human pollution indicate that the living environments in electronic waste dismantling areas have been seriously polluted (Chen *et al.*, 2019; Ohoro *et al.*, 2021; Zhao *et al.*, 2021). High concentrations of brominated pollutants have been found in the hair, blood, breast milk and umbilical cord blood of residents living around electronic waste dismantling areas, which indicates that the dismantling and recycling activities have created serious health risks (Whitehead *et al.*, 2013; Arita *et al.*, 2018; Guo *et al.*, 2019; Luan *et al.*, 2019). These signs and research results show that improper disassembly and disposal of electronic waste has seriously affected the lives and health of residents.

## 3 FORMATION OF POLLUTANTS

### 3.1 Generation Paths and Mechanisms

#### 3.1.1 Sources

PBDD/Fs are unintentionally generated by precursors and in so-called de novo syntheses involving chemical reactions, photochemical reactions or thermal reactions (Barontini and Cozzani, 2006). Source and characteristics of some PBDD/Fs are shown in Table 3. PBDD/Fs are present in organic

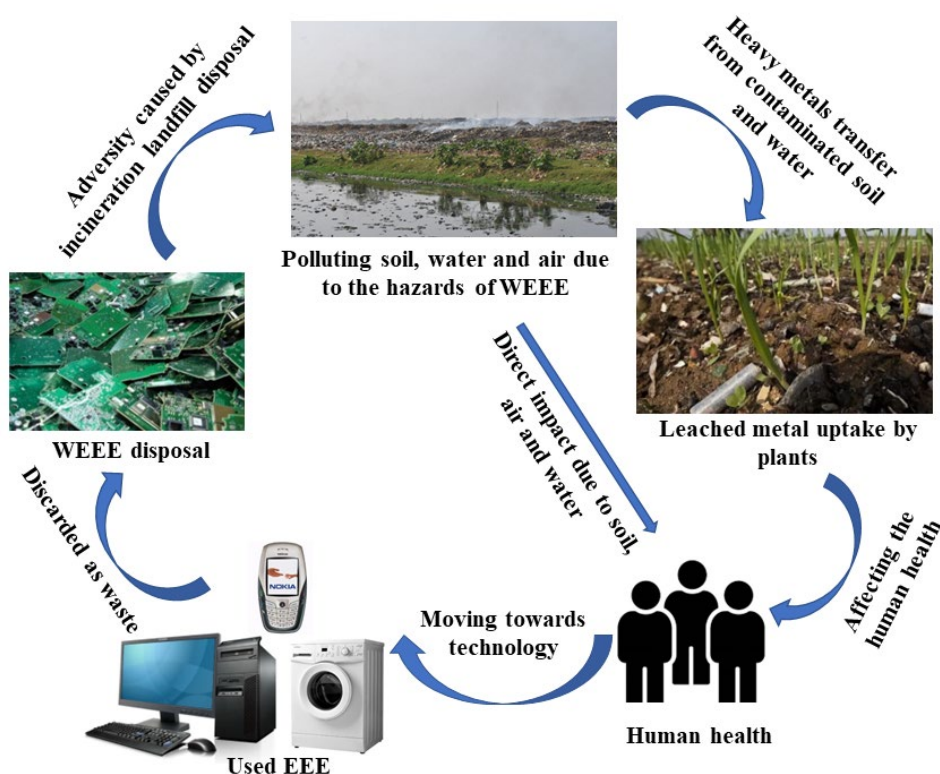
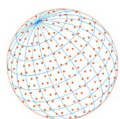


Fig. 2. Impacts of electronic waste on humans and the environment (Pathak *et al.*, 2017).

compounds containing bromine, especially bromine-containing flame retardants, such as PBDEs, decabromobiphenyl, and TBBPA. PBDFs and smaller PBDDs can be produced by photodegradation of organic bromides such as PBDEs and bromophenols.

The results of laboratory pyrolysis experiments showed that brominated flame retardants such as PBDEs, PBBs, TBBPA, and bromophenols have high potential for producing PBDD/Fs, and production is affected by factors such as temperature, polymers and synergistic reactants. Production is also affected by metals, metal oxides, water, oxygen and combustion equipment or reaction conditions.

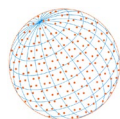
PBDD/Fs are generated during the production of polymers containing brominated flame retardants, such as acrylonitrile butadiene styrene (ABS) and polyethylene terephthalic acid (PBT), at temperatures of 150–300°C. Moreover, the generation capacities of plastics containing brominated flame retardants (BFRs) during production, processing and recycling have been confirmed (Hanari *et al.*, 2006; Ren *et al.*, 2011).

The incineration of brominated compounds is one of the sources of PBDD/Fs, and this includes municipal domestic waste, medical waste, electronic and electrical waste, hazardous waste, and fire retardants (Du *et al.*, 2010; Wang *et al.*, 2010). In particular, PBDD/Fs were detected in the fly ash and flue gas resulting from incineration of municipal domestic waste, medical waste and hazardous waste. Leaded gasoline is also a source of PBDD/Fs.

### 3.1.2 Formation mechanism

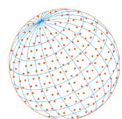
As with the generation of PCDD/Fs, PBDD/Fs are also mainly generated from precursors and de novo syntheses. The difference is that in the generation of PCDD/Fs, most of the bromine released into the environment comes from the production and use of brominated flame retardants, while brominated flame retardants very easily generate PBDD/F precursors such as bromobenzene and bromophenol during flame retardancy or pyrolysis and combustion. Therefore, the formation of PBDD/Fs is mainly related to the use of brominated flame retardants. The pyrolysis test results for brominated flame retardants show that brominated flame retardants such as PBDEs, PBBs, TBBPA and bromophenols have high potential for producing PBDD/Fs, and the process of producing





**Table 3.** Source and characteristics of some PBDD/Fs.

Source	Matrices	Furnace type	Levels of PBDD/Fs	Major congeners	Main sources	Air pollution control devices	Ref.
Disposal of e-waste and plastics	Air from vehicle parking area	-	0.0734–3.35 pg m <sup>-3</sup>	1,2,3,7,8-PeBDF, 2,3,4,7,8-PeBDF, 2,3,7,8-TBDF	PBDE-treated auto parts, auto exhaust, photolysis of BFRs, etc.	-	(Li <i>et al.</i> , 2016)
	Air around electronic waste dismantling area	-	8.12–461 pg m <sup>-3</sup>	Lower brominated congeners	BFR and heating recovery process in electronic equipment	-	(Li <i>et al.</i> , 2007)
Partial industrial activities	Farmland soils in e-waste recycling area	-	2.5–17 pg g <sup>-1</sup>	1,2,3,4,6,7,8-HpBDF, OBDF	Legacy of electronic waste recycling and disposal	-	(Xu <i>et al.</i> , 2017)
	Commercial polybrominated diphenyl ether mixtures	-	257–49605 ng g <sup>-1</sup>	Hexa-octa BDFs	Formed during the production of commercial PBDE	-	(Hanari <i>et al.</i> , 2006)
Combustion process	Stack gas from municipal solid waste incineration	Circulating fluidized bed	0.45–5.46 pg Nm <sup>-3</sup>	1,2,3,4,6,7,8-HxBDD, 2,3,4,7,8-PeBDF, 2,3,7,8-TeBDF	Bromine containing solid waste	Selective noncatalytic reduction denitrification + semidry desulfurization	(Wang and Chang-Chien, 2007)
	Stack gas from industrial waste incineration	Grate-type	4.5–77.3 pg Nm <sup>-3</sup>	2,3,7,8-TeBDF, 1,2,3,7,8-PeBDF, 2,3,4,7,8-PeBDF	Bromine containing industrial waste	Selective noncatalytic reduction denitrification + semidry desulfurization	(Wang and Chang-Chien, 2007)
Stack gas from waste incineration and metallurgical plants	Stack gas from waste incineration and metallurgical plants	Circulating fluidized bed	0.17–4.13 ng Nm <sup>-3</sup>	1,2,3,4,6,7,8-HpBDF	Composition of feedstock and APCDs	Activated carbon + fabric filter + wet deacidification	(Song <i>et al.</i> , 2019)
	Fly ash from cement kiln coprocessing solid waste	Cement kiln	0.032–0.22 ng g <sup>-1</sup>	HpBDF	Bromine containing solid waste	Electrostatic precipitation	(Yang <i>et al.</i> , 2019)
	Stack gas from secondary copper smelting	Anode furnace	0.459–1.41 ng Nm <sup>-3</sup>	1,2,3,4,6,7,8-HpBDF	Smelting process	Fabric filter + wet deacidification + activated carbon	(Yang <i>et al.</i> , 2021)



PBDD/Fs is related to the mechanism for synthesizing the precursor PBDD/Fs. The partial formation pathways of PBDD/Fs are shown in Fig. 3.

(1) PBDE-PBDD/Fs: PBDE-PBDD/Fs mainly remove Br<sub>2</sub> or HBr from within the molecule. The fact that mildly brominated PBDD/Fs produce more than highly brominated PBDD/Fs may indicate that the HBr removal reactions of mildly brominated PBDE have lower energies than those of the highly brominated PBDE. The other explanation involves obstruction of C-C bond formation in highly brominated PBDFs: for example, when 1,9-PBDFs are generated, the bromines substituted at the 1 and 9 positions generate "site crowding" in the PBDFs. The "bit crowding" effect for 1,9-halogenation of OCDFs has been reported. For brominated PBDFs, because they have a greater van der Waals force than chlorinated PBDFs, the position is more important. Buser *et al.* (1986) believed that the process of PBDE-PBDD decomposition involved intramolecular oxygen insertion; others believed that PBDE first decomposed into PBP and PBBz and then formed via dimerization. That is, PBDD was generated from PBDE via bromophenol via metal catalysis.

(2) PBBs-PBDF-oxygen insertion process: this conclusion was drawn from an experiment using decaBB and polybutylene terephthalate to generate PBDFs, and it is believed that the oxygen insertion process in this process proceeded via two mechanisms: a concentrated phase and a gas phase mechanism (Luijk and Govers, 1992). After studying the process by which PCB formed PCDF during pyrolysis, it was shown that PCDF was formed by intramolecular cyclization, including the removal of HCl molecules and two Cl or H atoms during oxygen insertion. There was a small difference between bromine and chlorination. The energies of the bonds led to a difference in the choice between the removal reactions and the optimal temperature range.

(3) PBP-PBDD/F-concentration reaction: The thermal generation of PBDD/F from PBP has great potential. It has been reported that in the pyrolysis processes of 2,4,6-triBP, the yields of PBDD/F reached 56% (500°C) and 89% (600°C). Sakurai and Weber (2002) studied the mechanism for the production of PBDD/Fs from 2,4-tiBP at low temperature (300°C). In the first concentration reaction, bromine or hydrogen was replaced; in the second concentration reaction, bromine was selectively removed. Notably, the production of PBDD/Fs by PBP was not limited by whether it originally existed in the material. Often, other BFR materials (such as PBDE or TBBPA) produce PBP during pyrolysis, and the generated PBP plays a role in the production of PBDD/Fs during pyrolysis.

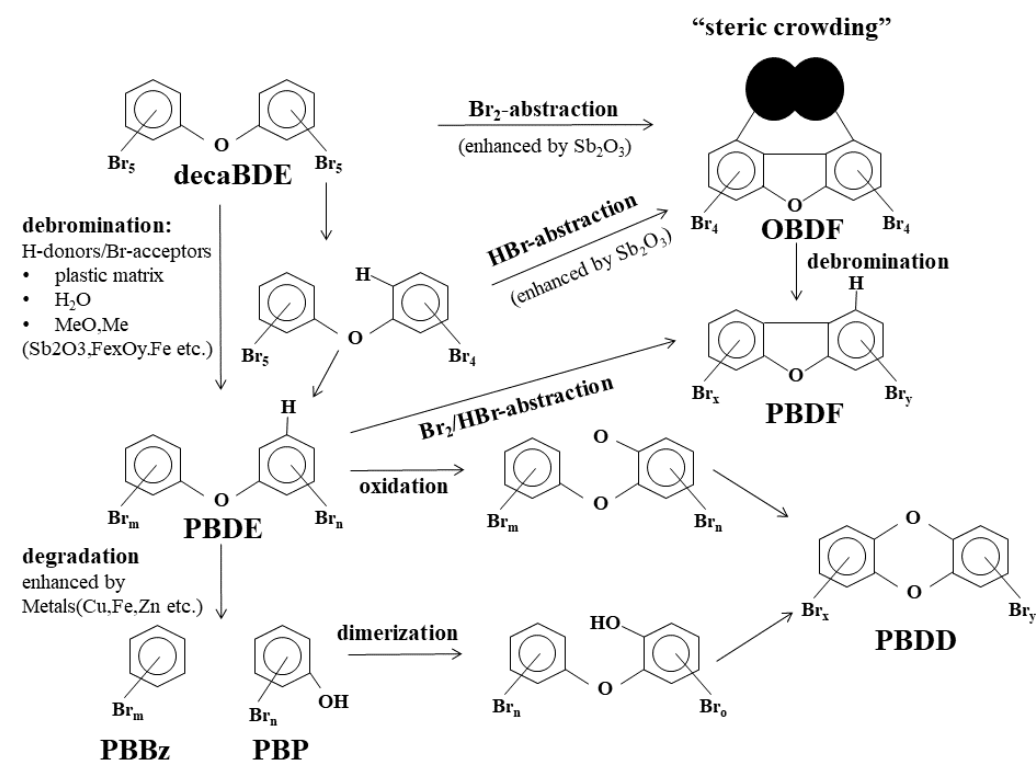
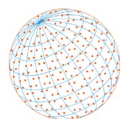


Fig. 3. Pathways for the formation of brominated dioxins (Weber and Kuch, 2003).



(4) Presynthesis of the TBBPA-PBDD/Fs precursor: PBDD/Fs have been detected during TBBPA pyrolysis, but the amount of PBDD/Fs present was several orders of magnitude lower than that of bromophenol, and only some mildly brominated ( $\text{Br}_1\text{-Br}_3$ ) PBDD/Fs were generated in the presence of the polymer. [Wichman \*et al.\* \(2002\)](#) reported that TBBPA and TBBPA polymers produced PBDDFs at the ppm level, in which a brominated compound occupied the main position.

The formation of bromophenol and bromobenzene via pyrolysis of TBBPA and its analogs showed that the production of bromophenol and bromobenzene had an important relationship with the formation of PBDD/Fs. It was formed via the following two steps: first, the formation of precursors (PBP, PBBz) during pyrolysis and incineration of TBBPA polymers; second, dimerization and concentration of precursors ([Weber and Kuch, 2003](#)). At this level, it is possible to predict the potential of brominated flame retardants (BFRs) to generate PBDD/Fs by estimating the potential of BFRs to generate bromophenol during pyrolysis.

## 3.2 Influencing Factors

### 3.2.1 Raw materials

The contents of bromine, brominated flame retardants and brominated dioxins in raw materials have a great impact on the generation of pollutants during subsequent treatment. Fly ash containing S is unlikely to generate dioxins. The printed circuit board and its shell were incinerated and pyrolyzed at 500°C with a low oxygen content, and their emitted pollutants were explored ([Moltó \*et al.\*, 2009](#)). The results showed that the volatile compounds released were closely related to the polymer content in the sample. Some brominated compounds have been identified as the precursors of brominated dioxins. The identification of dibromophenol and tribromophenol indicates the emission of PBDD/Fs. Moreover, the yields seen during pyrolysis and combustion are similar, but the toxic equivalent of the dioxin generated during pyrolysis is higher.

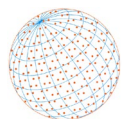
Brominated flame retardants (BFRs) are widely used to inhibit the ignition of combustible materials and prevent accidental fires. Waste printed circuit boards contain much brominated flame retardants. However, this can stimulate the emission of dioxins and mixed chlorinated/brominated dioxins produced by combustion with biomass ([Tange and Drohmann, 2005](#)). The material flow rates for PBDD/Fs in incineration flue gas and incineration residue were studied for three kinds of waste containing brominated flame retardants. When the concentration of bromine in the sample increased, the concentrations of brominated dioxins in the flue gas increased. Under proper combustion control and flue gas treatment conditions, the amount of PBDD/Fs released by incineration of a brominated flame retardant resin is lower than the input amount. The concentration of PBDD/Fs in the sample before incineration reached 45000 ng g<sup>-1</sup>. After incineration, the content of PBDD/Fs in the flue gas was 15–43 ng g<sup>-1</sup>, the content of PBDD/Fs in the solid residue was 2400–4400 ng g<sup>-1</sup>, and the destruction rate for all brominated dioxins was 89–99.7%. The PBDEs in the incineration residue dominated the total release of dioxins ([Sakai \*et al.\*, 2001](#)).

### 3.2.2 Temperature

Temperature is an important factor affecting the generation and emission characteristics of brominated pollutants. At low temperatures, brominated pollutants mainly exist in the form of HBr. At medium temperatures, brominated organics, such as bromoform, bromophenol and dibromophenol, are produced. At sufficiently high temperatures and with excess air, organic bromine can be converted into inorganic bromine.

The low-temperature pyrolysis range (290–450°C) is mainly used to treat circuit boards. Thermogravimetric analysis showed that the 300–360°C temperature range resulted in rapid weight loss from circuit boards and was the main production range for HBr. Further analysis showed that the HBr from the waste circuit board was mainly produced at 290–406°C during incineration. The concentration range for PBDD/Fs in the original circuit board was high, 3–130 µg g<sup>-1</sup>, and increases in the temperature reduced the amount of HBr emitted ([Chiang and Lin, 2014](#)). Some researchers have also used the two-stage pyrolysis method to remove bromine from electronic waste plastic mixtures ([Bhaskar \*et al.\*, 2002](#)). In the first stage, the reaction was run at 330°C for 2 h, and the second stage was carried out at 430°C. This two-stage pyrolytic method meant that most of the bromine found in the first pyrolysis oil was contained in hydrogen bromide and organic





bromides. The pyrolysis oil obtained from the second step accounted for 60% of the total but only contained a small amount of bromine. By studying the thermal decomposition processes and pyrolytic pathways converting waste circuit boards into macromolecular monomers and benzofuran products, it was shown that HBr was generated in the initial stage of pyrolysis, then the resin body was depolymerized into macromolecular monomers, and then random fracture and free radical reactions formed the pyrolysis products. Over the whole thermogravimetric process, HBr was mainly produced at the maximum thermogravimetric stage, and the peak value corresponded to the maximum thermogravimetric peak, indicating that the formation of HBr and fracture of the main chains of the brominated epoxy resin occurred simultaneously, and in addition to HBr, methyl bromide, vinyl bromide, etc., were produced (Marongiu *et al.*, 2007; Grause *et al.*, 2008).

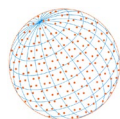
In the medium temperature range (450–800°C), due to limited diffusion of the resin matrix, an increase in the pyrolysis temperature did not improve the conversion rates of the waste circuit boards, and bromobenzene and bromophenol were found in both the gas and liquid products. The pyrolysis products generated in the middle temperature range contained many oxides and halogenated compounds. Methanol, ethyl acetate, acetone, dichloromethane, tetrachloromethane and acrylonitrile were the main oxides and chlorides produced during the pyrolysis of WPCBs, and their total emissions reached 846–5269  $\mu\text{g g}^{-1}$  at 500°C. The chlorides mainly came from the residual chlorination reagent left from the manufacture of the PCBs. When the pyrolysis temperature exceeded 400°C, higher contents of brominated organic compounds, namely, bromoform, bromophenol and dibromophenol, were produced. Pyrolyses of high-impact polystyrene were carried out at 450°C to 550°C in a fluidized bed reactor. It was found that when the temperature was raised from low temperature to medium temperature, the amounts of inorganic bromides emitted were significantly reduced, and the contents of hydrogen bromide and bromine in the pyrolysis oil decreased from 120 ppm to 20 ppm and 0 ppm, respectively (Hall and Williams, 2006).

The high-temperature section (800–1200°C) was used for incineration research. The experiments showed that a sufficiently high temperature and excess air converted the organic bromides into inorganic bromides, mainly HBr and Br<sub>2</sub>. When the temperature was increased from 800°C to 1400°C, the proportion of inorganic bromine in the flue gas increased from 90% to 100%. Additionally, the proportion of Br<sub>2</sub> in the flue gas decreased from 0.1% to 0.04%, and the proportion of HBr increased from 0.01% to 0.18%. At higher temperatures, Br<sub>2</sub> was more likely converted into HBr. In a study of organic bromides, the experimental results showed that the yields of PBDD/Fs increased from 809.33 to 1220.36  $\text{pg g}^{-1}$  for pyrolyses at 800°C or 1000°C. When the temperature was higher than 1100°C, the PBDD/Fs generated in the incineration process were completely decomposed, and the amount of PBDD/Fs produced was reduced to 717.79  $\text{ng g}^{-1}$  (Lai *et al.*, 2007). Overall, the toxicity equivalent of PBDD/Fs was continuously reduced by heating at 800–1200°C. Some scholars have used rotary kilns and incinerated electronic waste containing brominated flame retardants such as polyethylene with polybrominated diphenyl ethers (PBDEs), discarded television sets and printed circuit boards. After high-temperature incineration, the destruction rates for halogenated dioxins brought into the incinerator with fuel reached 89%–99.7%. The discharged halogenated dioxins were mainly concentrated in the slag and fly ash. Therefore, high-temperature incineration effectively destroyed the brominated dioxins.

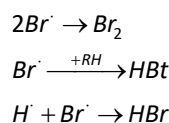
Additionally, the incineration temperature is also an important factor affecting the state of bromine. With increasing temperature, the conversion of NaBr-HBr in the combustion process increased greatly, and the bromine content in the residue showed a downward trend. At low temperatures, bromine was found in the residue. At high temperatures (above 800°C), bromine was volatilized in the form of inorganic bromide salts. At high temperature, the presence of oxygen significantly increased the conversion rate of bromine. The temperature was important in determining the emission characteristics of the pollutants. Based on the literature and the influence of temperature on the generation of brominated compounds, the high-temperature treatment of circuit boards effectively reduced the emission of halogenated dioxins.

### 3.2.3 Atmosphere

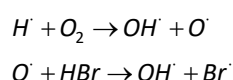
The atmosphere is also an important factor affecting the generation of pollutants. The study found that an increased O<sub>2</sub> concentration in flue gas increased the rate of HBr conversion to Br<sub>2</sub>, and the concentration of HBr in the flue gas decreased. When excess air was added, the HBr



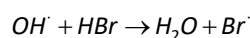
concentration decreased, and in the presence of oxygen, the proportion of bromine contained as inorganic bromine was very high. It is widely believed that when the polymer is heated to a certain temperature, the chemical chain breaks, and the chemical bonds between H and other elements (C, O) break, producing H free radicals. At the same time, the bonds connecting the main parts of Br and TBBA molecules are also disrupted, producing Br radicals. Then,



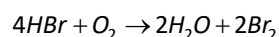
In the above reactions, Br radicals are mainly converted into HBr (Noto *et al.*, 1998; Barontini and Cozzani, 2006). Meanwhile, in the presence of oxygen,



The above reactions are the most important producers of OH radicals (Kuo, 1986).



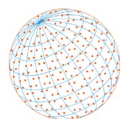
The above reactions are the most important free radical scavengers (Julien *et al.*, 1996; Wang and Anthony, 2009; Ni *et al.*, 2012). All the above reactions can be summarized as the following overall reactions:



The conversion of HBr to Br<sub>2</sub> consumes a large amount of OH and H radicals. Small amounts of excess air lead to high CO and HBr contents in the flue gas; more air causes heat loss and forms more SO<sub>2</sub> and NO<sub>x</sub>. In the high-temperature incineration experiment with the continuous feed dropper combustion system, it was found that when the temperature was above 1200°C, the excess air factor was 1.3, and the high-temperature residence time was 0.75 s, 99.9% of the bromine was discharged into the flue gas as HBr and Br<sub>2</sub> (Ni *et al.*, 2012). Therefore, the excess air coefficient should be kept at 1–2 to control the formation of brominated organic compounds and reduce the emission of SO<sub>2</sub> and NO<sub>x</sub>.

### 3.2.4 Reaction time

The reaction time is an important factor affecting the extent of the reaction. The research showed that the halogenated aromatic compounds were not completely destroyed due to insufficient combustion. However, with controlled secondary combustion (at temperatures higher than 850°C), the BFRs were decomposed efficiently, and a retention time of 2 s ensured destruction of the brominated ether precursors with destruction rates exceeding 99.9% (Weber and Kuch, 2003). Moreover, the reaction time also affected the conversion of bromine. If the flue gas remained in the furnace for less than 5 s, the proportion of bromine molecules in the inorganic bromine decreased because oxygen in the air converted the hydrogen bromide into molecular bromine. With increased time, the bromine content in the residue decreased to approximately 2.5%. This occurred because the increased reaction time of the residue meant that bromine was separated in gaseous form. There was a relationship between the sintering time and the formation of dioxins (Barontini and Cozzani, 2006). Compared with the self-condensation reactions of chlorophenol/chlorophenoxy radicals, the ab initio synthetic reaction took more time. The time required for complete combustion and decomposition of the dioxins was directly related to the temperature, as well as to the mixing generated by turbulence, vortex mixing and a strong velocity gradient. At 1200°C (a typical temperature in hazardous waste incinerators), it took a



few milliseconds to achieve complete combustion in air, while at 800–900°C (a higher temperature range for fluidized bed incineration), it took a few seconds. Therefore, the waste circuit boards were fully combusted in a few milliseconds in a high-temperature smelting furnace and then disposed.

## 4 CONTROL TECHNOLOGY OF POLLUTANTS

### 4.1 Source Control

PBDEs and HBCDs are mainly used in electronic products, textiles and thermal insulation materials. Due to bioaccumulation, biological toxicity, environmental durability and long-distance mobility, they have been listed as persistent organic pollutants by the Stockholm Convention. The World Health Organization (WHO) recommended that the maximum amount of PBDD/Fs inhaled by the human body per day should be 1–4 pg TEQ day<sup>-1</sup> kg<sup>-1</sup>. The Directive on the Restriction of the Use of Hazardous Substances in Electronic and Electrical Products requires EU member states to exclude lead, mercury, cadmium, hexavalent chromium, polybrominated diphenyl ethers (PBDEs) or polybrominated biphenyls from the electronic and electrical products put on the market after July 7, 2006.

The Regulations on the Administration of the Recycling and Disposal of Waste Household Appliances and Electronic Products, which were issued by China, preliminarily determined an overall management framework for recycling and disposal of waste household appliances. The limiting value for PBDD/Fs contained in waste circuit boards was specified in the national standard GB 5085.6-2007 as 15 µg TEQ kg<sup>-1</sup>.

To avoid generating large amounts of pollutants during the disposal of waste circuit boards, waste circuit boards can be pretreated before processing to control the release of organic pollutants during disassembly (Fig. 4).

The temperature for disassembly of the circuit board can be controlled to reduce the amount of pollutant emitted at the source. Circuit board fragments were thermally disassembled at temperatures of 100–260°C to study the factors leading to PBDE emission (Liu *et al.*, 2019). When the temperature was increased from 100°C to 260°C, the level of PBDEs emitted rose from

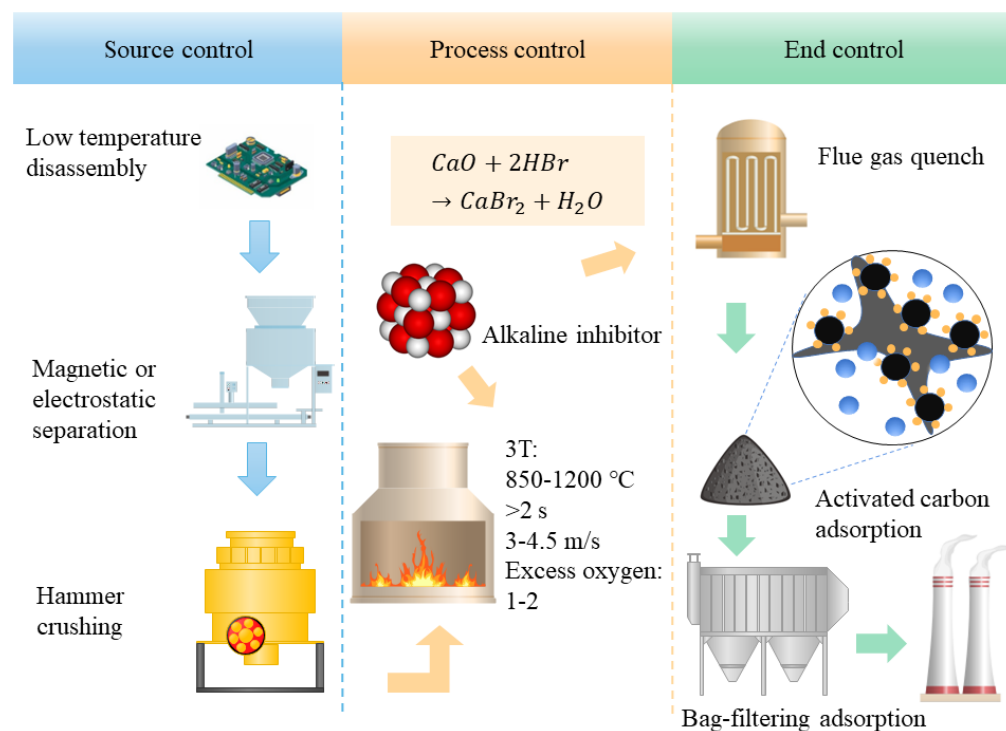
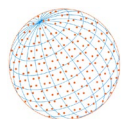


Fig. 4. Schematic diagram for the control method.



8.38 ng g<sup>-1</sup> to 8238.10 ng g<sup>-1</sup>. Wang and Xu (2015) used a sealed system to disassemble a circuit board and found that the critical temperature at which toxic PCBs were produced was 270–280°C. Therefore, low-temperature crushing or reduced disassembly temperatures should be adopted to reduce the release of organic pollutants during thermal disassembly.

The degree of circuit board breakage during pretreatment is also an important factor. When the crushing particle sizes were increased from 2.0 cm to 5.0 cm, the copper recovery rate decreased significantly from 98.77 to 80.25 wt.%, which constituted a substantial metal loss. When the crushing particle size increased from 2.0 to 5.0 cm, the organic bromine content in the solid residue increased from 40 to 45 wt.%, indicating that the larger particle sizes during pretreatment of the crushed circuit board reduced bromine pollution and the migration of bromine components from the solid phase to the liquid and gas phases. The results showed that when the particle size of pretreated particles was 4.0 cm, metal recovery was efficient, and the brominated pollutants were easily controlled.

There are also new processes used to pretreat waste circuit boards. Supercritical CO<sub>2</sub> and aqueous solutions were used to dissolve waste circuit boards at different temperatures and pressures, and then the delamination mechanism was studied with Fourier transform infrared spectroscopy and other material characterization methods. The experimental results showed that the waste circuit board was layered and easily divided into copper foil, glass fibers and polymer. At 180°C and 138 bar, the glass transition temperature, the crosslinking density and the FTIR spectra of the polychlorinated biphenyls did not change significantly, and no new hydrocarbons or toxic gases were produced, which indicated an environmentally friendly process.

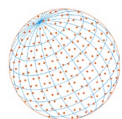
Suggestions for effectively reducing and preventing the discharge of pollutants during thermal disassembly of waste circuit boards include the following:

- (a) before thermal disassembly, the components (capacitors, resistors, slots) in the circuit board can be removed without heating and treated separately in an appropriate way to reduce the total PBDE emissions;
- (b) before heating and disassembling the waste circuit boards, structural damage and aging of the materials by environmental factors should be avoided, which would reduce the amount of PBDEs emitted during heating and disassembly;
- (c) to maintain high disassembly rates for the electronic components, a lower disassembly temperature should be used to reduce the pollution of the surrounding air by PBDEs and heavy metals;
- (d) the concentrations of PBDEs, heavy metals and fine particles present during the disassembly process are high. A particle purification device should be added to the thermal disassembly equipment or at the disassembly site; Zn, Cr, Sb and other heavy metals produced during thermal disassembly of waste circuit boards are typically adsorbed on coarse particles. Safety equipment such as anti-particle masks effectively prevents injury to the respiratory systems of workers caused by Zn, Cr, Sb and other heavy metals.

#### 4.2 Process Control

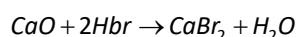
During the treatment process, an alkaline inhibitor can be added to control pollutant discharge (Fig. 4). In some studies, waste circuit boards were mixed with sulfur powder and calcium oxide powder before combustion. These experiments showed that after mixed combustion, the efficiency for conversion of the bromine in the raw materials into inorganic bromine was increased. The presence of CaO promotes the conversion of organic bromine into inorganic bromine, and the SO<sub>2</sub> generated during sulfur powder combustion can also reduce Br<sub>2</sub> in the flue gas to HBr, reducing the proportion of Br<sub>2</sub> in inorganic bromine, thus effectively controlling the formation of brominated pollutants (Söderström and Marklund, 2002; Jin *et al.*, 2011).

The effects of alkaline inhibitors such as CaO, CaCO<sub>3</sub> and Ca(OH)<sub>2</sub> on the generation and emission of PBDD/Fs during high-temperature incineration of circuit boards have also been studied by scholars (Åslander and Modig, 1987; Blazsó *et al.*, 2002; Liu *et al.*, 2005; Lu *et al.*, 2007). CaO, CaCO<sub>3</sub> and Ca(OH)<sub>2</sub> inhibited the formation of PBDD/Fs when waste circuit boards were incinerated at high temperatures. When the mass ratio of CaO was 1:1, it reduced PBDD/F formation by 97%, and the toxic equivalent of the PBDD/Fs was reduced by 66%. The added CaO enabled conversion of the PBDD/Fs from highly brominated to mildly brominated. When the mass ratio of CaCO<sub>3</sub> was



1:3, the amount of PBDD/Fs formed was reduced by 84%, but the toxic equivalent of the PBDD/Fs was increased by the addition of CaCO<sub>3</sub>. When the mass ratio was 1:2, the toxic equivalent of the PBDD/Fs was 40% higher compared to conversion without CaCO<sub>3</sub>. When the mass ratio of Ca(OH)<sub>2</sub> was 1:3, PBDD/F formation was reduced by 89%, and the toxic equivalent of the PBDD/Fs was reduced by 36%. Overall, the inhibition efficiencies of CaO, CaCO<sub>3</sub> and Ca(OH)<sub>2</sub> for PBDD/F formation decreased in the order CaO > Ca(OH)<sub>2</sub> > CaCO<sub>3</sub> (Lai *et al.*, 2007; Lu *et al.*, 2007; Terakado *et al.*, 2011). In mixed pyrolyses of alkaline additives and circuit boards, it was found that the Br removal efficiency exceeded 95%, and the amount of PBDD/Fs formed was related to the content of HBr in the atmosphere. Therefore, the addition of alkaline substances inhibited PBDD/F formation during circuit board incineration and pyrolysis.

Generally, bromine decomposes at high temperatures to produce an acidic gas (HBr), which is neutralized by an alkaline inhibitor. For example, a possible reaction between CaO and HBr is as follows (Lai *et al.*, 2007):



The equation shows that the reaction between HBr and CaO formed the solid product CaBr<sub>2</sub>, which is nonvolatile and was collected in the bottom ash. Therefore, the added CaO effectively adsorbed HBr, thus inhibiting the synthesis of PBDD/Fs. The mechanisms for other alkaline inhibitors are similar to that of CaO.

### 4.3 End Control

In addition to source control and process control, the tail end must also be controlled. Flue gases contain many pollutants and must be purified. Tail control can be realized by adding a purification device or adsorbent or by quenching the tail gas (Fig. 4).

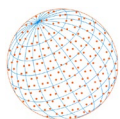
The impact of activated carbon used to control dioxin/heavy metal emissions was studied. When the amount of activated carbon was increased from 0 to 50 kg h<sup>-1</sup>, the average toxic equivalent concentration of dioxin decreased from 0.360 ng I-TEQ Nm<sup>-3</sup> to 0.113 ng I-TEQ Nm<sup>-3</sup>, and the removal efficiency increased from 87.97% to 96.22%. The adsorption capacity of the activated carbon was reflected in the removal of low-halogenated dioxins from the flue gas. However, the impact of added activated carbon on the emission of the heavy metals Hg and Cd was investigated. Because activated carbon was used in the production process, activated ash particles were deposited on the surfaces of the cloth bag, which led to highly efficient heavy metal removal even if it was not used in the test stage (Hutson *et al.*, 2009; Hu and Cheng, 2016).

The most inexpensive way to remove dioxins is to spray adsorbents such as activated carbon in front of particulate matter capture devices, such as cloth bags. When the adsorbent reacts physically with the dioxin in the flue gas, the dioxin is absorbed and then enters the cloth bag. The cloth bag is used to separate the adsorbent and flue gas and to intercept the fly ash and other substances in the semidry system.

The removal efficiency for dioxins depends on the quality of the spray adsorbent, the mixing efficiency of the adsorbent and the flue gas, the type of filter bag and the operation of the system. The efficiency of the cloth bag during the operation should be guaranteed because the cloth bag ensures that the reaction time between the adsorbent and the flue gas is extended due to air flow through the filter bag. Due to the low prices of adsorbents, bag dust collectors are more commonly used in foreign countries than electrostatic precipitators. However, the disadvantage of this technology is that, in general, and especially in refurbished incinerators, the temperature at the bag outlet is often higher than 200°C. To ensure adsorbent adsorption efficiency and the safety of the activated carbon, a maximum allowable temperature should be established for this method. If efficient byproduct recovery is considered when flue gas purification equipment is installed, adsorbent injection is obviously beneficial. The most common adsorbents used are coke and powdered activated carbon. Currently, some special materials, such as alumina, are also being used due to their high specific surface areas.

Injecting limestone into an SDA (spray dryer absorption) system reduces the amount of dioxin in the flue gas (Nielsen *et al.*, 1986). The test results showed that the removal efficiency reached





84–99.7%. Suzuki *et al.* (2003) used the waste generated by the CaC<sub>2</sub> production process (the waste contained much oxides of Ca, Mg, Si, Fe, etc.) to test the efficiency for dioxin adsorption from the flue gas and obtained 74–95% PBDD/F adsorption efficiencies in practical applications.

Other scholars have studied the use of quenching equipment to control the emission of dioxin/heavy metals. These experiments showed that the levels of dioxin emitted were reduced to 0.114 and 0.223 ng I-TEQ Nm<sup>-3</sup> with the quenching equipment, which were significantly lower than the 0.619 and 0.387 ng I-TEQ Nm<sup>-3</sup> emission rates seen without the quenching equipment, so the installation of quenching equipment in the waste heat boiler section effectively reduced the emission of PBDD/Fs. The toxic equivalences for the dioxin isomers produced with and without quenching facilities were compared. The total toxic equivalence of the low-halogenated dioxins was 42% with the quenching equipment, which was significantly lower than the 61% value generated without quenching, indicating that quenching facilities inhibit the generation of low-halogenated dioxins (Schlummer *et al.*, 2007; Aurell *et al.*, 2009; Dong, 2021).

## 5 CONCLUSIONS

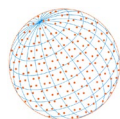
This article introduces the current situation, treatment methods, policies and regulations for electronic waste in circuit boards as well as the sources, generation mechanisms and research progress on brominated dioxins. The factors influencing waste circuit board treatments were discussed and summarized. The emission characteristics and factors influencing brominated pollutants, including the temperature, atmosphere, time, and raw materials, were comprehensively discussed. Research has found that (i) below 450°C, brominated pollutants mainly exist in the form of HBr; Between 450°C and 1000°C, brominated organic compounds such as dioxins, bromophenol, and dibromophenol will be produced; At temperatures ranging from 800 to 1300°C, the toxic equivalent of PBDD/Fs continuously decreases, and organic bromine can be converted into inorganic bromine. (ii) At high temperatures, the presence of oxygen significantly increases the conversion rate of bromine. The excess air coefficient should be maintained at 1–2 to control the formation of brominated organic compounds and reduce the emissions of SO<sub>2</sub> and NO<sub>x</sub>. (iii) The time required for complete combustion and decomposition of dioxins is directly related to temperature, turbulence, vortex mixing, and mixing generated by strong velocity gradients. If the reaction time is kept up to two seconds, the destruction rate of the brominated precursor ether will exceed 99.9%. An appropriate treatment range was selected to reduce the emission of brominated pollutants, which provides a foundation for subsequent research.

This article discusses the control measures for brominated pollutants for the source, process and end. This article believes that waste circuit boards can be treated at the source before processing, and the factors affecting the release of organic pollutants during the disassembly process, such as the disassembly temperature used during preprocessing is below 260°C or the disassembly particle size of the circuit board is about 4 cm. Alkaline inhibitors can be added to control the discharge of brominated pollutants. Pay attention to the 3T+E principle during the disposal process. The tail end emissions can be controlled by adding a purification device or adsorbent, or the tail gas can be quenched. Further study should gather supplemental data to (i) better understand the processes responsible for brominated pollutant formation and (ii) take measures for reducing the emissions formed over an entire cycle.

## ADDITIONAL INFORMATION AND DECLARATIONS

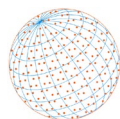
### Conflict of Interest Statement

We declare that we have no financial and personal relationships with other people or organizations that can inappropriately influence our work. There is no professional or other personal interest of any nature or kind in any product, service and/or company that could be construed as influencing the position presented in, or the review of.

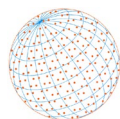


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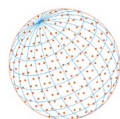
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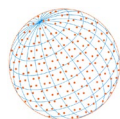


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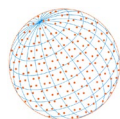


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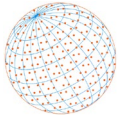




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