Green Energy and Fuel Research

Review

Thermal Reactions and Byproducts from the Waste-to-Energy Process of Flame Retardant-Containing Wastes—A Review

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Abstract: The increasing global concern about global warming has spurred researchers and industries to actively explore low-carbon energy alternatives to reduce carbon emissions and lessen dependence on traditional energy sources. Waste-to-energy (WTE) conversion has emerged as a promising solution in this pursuit. However, the prevalence of flame retardants (FRs) in various household materials poses a challenge to WTE processes. FRs, commonly added to prevent fire hazards, include chlorine-, phosphorus-, and nitrogen-based variants, each with specific applications and fire suppression mechanisms. Thermal treatment technologies, such as incineration, pyrolysis, gasification, and hydrothermal treatment, are currently employed for energy conversion. While effective in reducing waste volume and degrading most FRs, these processes can generate secondary pollutants, including polychlorinated dioxins, with complex reaction pathways that are difficult to control. This necessitates stringent management measures to mitigate the associated environmental risks. In contrast, non-thermal degradation techniques, such as chemical degradation, photocatalysis, biodegradation, and electrochemical methods, offer more environmentally friendly alternatives. However, current technological limitations constrain their application scope and efficiency. This review aims to comprehensively examine the pollutant emission behaviors of FRs during thermal treatment processes for energy conversion, highlight the associated environmental risks, and assess the potential of non-thermal degradation techniques. By analyzing these aspects, the review seeks to provide scientific insights and technological support for achieving waste valorization and low-carbon sustainability.

Keywords: waste-to-energy; persistent organic pollutants; flame retardant; thermal treatment; non-thermal degradation

1. Introduction

In recent years, global warming has become an increasingly critical issue. To minimize carbon emissions and decrease reliance on traditional energy sources, efforts are being made worldwide to explore low-carbon energy alternatives. Renewable energy, which refers to naturally occurring energy sources that can be directly utilized or processed for reuse, is key in reducing carbon emissions while mitigating air and water pollution. This category encompasses various technologies, including solar energy, bioenergy, geothermal energy, wind energy, and ocean energy. Among these, waste-to-energy (WTE) conversion has gained significant attention as a viable form of renewable energy. On the other hand, to enhance fire resistance and prevent flame propagation, thereby ensuring safety and protecting lives and property, FRs are now widely incorporated into everyday products. These include textiles, electronic devices, building materials, automotive components, and plastics [1]. With the widespread use of these products, the demand for FRs has also increased.

With the widespread use of various products, the consumption of FRs has increased significantly. Due to their high stability and persistence in the environment, FRs have gradually become a serious environmental concern. Numerous studies have indicated that these compounds are present in the air, water, soil, and even living organisms, posing potential risks to biological systems. Exposure to FRs has been linked to disruptions in the endocrine, nervous, reproductive, immune, and cardiovascular systems [2–4]. Moreover, when flame-retardant-containing waste undergoes thermal treatment for energy conversion, the process inevitably generates by-products, many of which also exhibit high stability. Organic chemicals that exhibit stability, toxicity, bioaccumulation, and long-range environmental transport are classified as persistent organic pollutants (POPs). In the past, POPs did not receive sufficient attention; however, their widespread use has increased, their environmental hazards have become more apparent. In 1995, the United Nations Environment Programme (UNEP) called for global action to address POPs and compiled a list of the 12 most environmentally hazardous chemicals. In 2001, countries signed the Stockholm Convention, initially regulating only 12 chemicals. As research advanced and awareness of hazardous chemicals grew, the convention has been progressively updated, with 39 chemicals currently under regulation. Among the regulated substances are several brominated FRs, such as polybrominated biphenyls (PBBs), polybrominated diphenyl ethers (PBDEs), tetrabromobisphenol A (TBBPA), and hexabromocyclododecane (HBCD), as well as chlorinated FRs like short- and medium-chain chlorinated paraffins. The convention classifies chemicals into three categories based on their properties: Annex A (Elimination), Annex B (Restriction), and Annex C (Unintentional Production). All regulated FRs are listed under Annex A, requiring signatory countries to take measures to eliminate their production and use.

FRs can be classified into two categories based on their application: reactive and additive types. Reactive FRs form stable bonds by chemically reacting with the polymer matrix to create covalent links (e.g., TBBPA). In contrast, additive FRs are physically blended into the polymer without chemical bonding, making them more prone to leaching (e.g., PBDE and HBCDD) [5]. FRs can be categorized by composition into inorganic and organic types, with organic FRs generally exhibiting higher persistence and toxicity than inorganic ones. Organic FRs are further divided into three main types based on their chemical makeup: halogenated FRs (containing bromine or chlorine), phosphorus-based FRs (PFRs), and nitrogen-based FRs (NFRs). Initially, halogenated FRs, especially brominated FRs (BFRs), were the most popular due to their thermal stability, minimal impact on polymers, and lower cost compared to other FRs, leading to their widespread use. Studies show that most halogenated chemicals persist, bioaccumulate, and are toxic to the environment, animals, and humans. As a result, specific flame retardant components have since been regulated [6].

Although some BFRs have been banned or voluntarily phased out, specific emerging and existing BFRs continue to be used in industrialized countries. With increasing concerns over the hazards posed by halogenated FRs, interest in halogen-free alternatives has grown. PFRs have become widely adopted as substitutes for BFRs [7,8]. A Stapleton et al. [9] study illustrates this trend of PFRs replacing BFRs. They collected and analyzed 102 polyurethane foam samples from residential sofas purchased in the U.S. between 1985 and 2010. In samples purchased before 2005 (n = 41), polybrominated diphenyl ethers (PBDEs) related to the penta-bromodiphenyl ether mixture were the most common FRs, followed by tris(1,3-dichloro-2-propyl) phosphate (TDCPP; 24%). In samples purchased after 2005 (n = 61), TDCPP was the most frequently detected flame retardant, and mixtures of non-halogenated organic phosphate FRs, such as triphenyl phosphate (TPhP) and tris(4-butylphenyl) phosphate (TBPP), were also found. In 2017, the global consumption of organic phosphate esters (OPEs) was approximately 2.5 million tons [9].

PFRs are more biodegradable than BFRs and cause less environmental harm, making them a comparatively more eco-friendly option [10]. Subsequent research has found that some PFRs and their transformation products exhibit moderate to high persistence [11]. Increasing evidence suggests that these compounds pose health risks and can also induce biological effects, potentially causing significant environmental harm over time [7,12].

FRs enter the environment primarily through three pathways: (i) Emissions during the manufacturing process. Some products are heated during production, releasing trace amounts of FRs. (ii) Release from flame retardant-containing products during use. Products containing FRs can emit small amounts over time. In a study by Kajiwara, Desborough, Harrad, Takigami [13], it was found that decabromodiphenyl ether (DecaBDE) in flame-retardant textiles undergoes photodegradation when exposed to natural sunlight, indicating that these textiles may be a potential source of BFRs in dust. (iii) Waste disposal. This is the primary source of FRs in the environment. Both municipal and electronic waste contain FRs, and improper disposal poses a significant environmental risk [14]. FRs are commonly found in municipal waste, and improper disposal of these materials poses an essential environmental risk. Thermal treatment is one of the most widely used waste management techniques. However, this approach conflicts with the properties of FRs, raising concerns about its suitability. Although several existing reviews have examined the environmental risks or degradation pathways of specific FRs, most have been limited to a single type of FR or a specific treatment technology. Moreover, the behavior of FRs during high-temperature conversion processes remains insufficiently explored. In particular, the formation of toxic by-products and the variability in thermal decomposition mechanisms among different types of FRs have yet to be comprehensively investigated. In contrast, this review adopts an integrative perspective from a WTE standpoint, offering a comparative analysis of various types of FRs and major thermal treatment technologies. In addition, it explores the potential applications of non-thermal treatment methods to address current gaps in the literature. The novelty of this review lies in three key contributions: (i) It presents the first systematic comparison of the decomposition pathways and by-product formation mechanisms of different FR types, including halogenated, phosphorus-based, and nitrogen-based compounds, under thermal treatment conditions, a topic often overlooked in energy conversion research. (ii) Beyond focusing on halogenated FRs, it also highlights the environmental risks and potential toxic by-products associated with phosphorus- and nitrogen-based FRs, areas that have received comparatively limited attention. (iii) While discussing thermal treatment strategies, the review further examines the complementary role and advantages of non-thermal degradation techniques in pollution control. It proposes the feasibility of integrated treatment approaches tailored to the characteristics of different waste streams.

2. Organic Flame Retardants

From past to present, commonly used organic FRs include BFRs, CFRs, PFRs, and NFR [15]. Table 1 summarizes the most representative compounds among various classes of organic flame retardants, along with a brief overview of their typical applications and current usage status. Figure 1 further illustrates the molecular structures of these compounds and their corresponding application areas. The development trends and practical applications of each type of flame retardant will be discussed in greater detail in the following sections.

Table 1. Different Types of Organic FRs.

Category	Examples	Remarks
BFRs	TBBPA, HBCD, PBDE, PBB	Formerly widespread in plastics and electronics, its use has significantly declined due to persistence and bioaccumulation concerns, though it’s not entirely banned.
CFRs	Dechlorane Plus (DP) and chlorinated paraffins (CPs)	
PFRs	TDCPP, TPhP, tert-Butylated Triarylphosphate (TBPP), 9,10-dihydro-9-oxa-10-phosphaphenanthrene-10-oxide (DOPO)	Widely adopted halogen-free alternatives in foams, coatings, and electronics face some toxicity concerns.
NFRs	Pure melamine, melamine derivatives	Typically combined with PFRs, they work through endothermic decomposition and inert gas release. Effectiveness varies by formulation.

BFRs and CFRs are often collectively referred to as halogenated FRs due to their similar flame-retardant mechanisms and structures. Among these, bromine dominates in application because of its higher efficiency. Efficiency and stability are the two main factors determining which halogenated compounds can be used as FRs. Fluorinated compounds are too stable (C–F bond: 467 kJ/mol), while iodinated compounds are insufficiently stable to withstand processing temperatures (C–I bond: 228 kJ/mol). Therefore, only chlorine (C–Cl bond: 346 kJ/mol) and bromine (C–Br bond: 290 kJ/mol) are suitable for use as FRs [16]. Structurally, halogenated FRs can be divided into three major categories: aliphatic, aromatic, and alicyclic compounds. Due to their superior thermal stability, aromatic compounds are more commonly used.

Halogenated FRs (HFRs) are highly effective in flame retardation, do not significantly alter the properties of polymers when added, and are more cost-effective compared to other types of FRs. These advantages have made HFRs highly favored in industries such as electronics, plastics, and rubber. In the past, the most widely used BFRs included tetrabromobisphenol A (TBBPA), hexabromocyclododecane (HBCD), and polybrominated diphenyl ethers (PBDEs). However, the environmental and health concerns associated with traditional BFRs have drawn increasing attention in recent years, leading to the implementation of restrictions and bans on some BFRs.

Abbasi et al. [17] highlighted that although PBDEs have been gradually phased out, their emissions are expected to persist until 2050 due to the temporal and spatial lag in chemical management practices. The restrictions on traditional BFRs have, in turn, driven the development and adoption of next-generation BFRs [6], including novel brominated FRs (NBFRs). Among these, decabromodiphenyl ethane (DBDPE) is the most widely used, directly replacing commercial decabromodiphenyl ether mixtures. DBDPE is marketed for use in various applications such as plastics, resins, rubbers, adhesives, and textiles [18]. Studies indicate that NBFRs share similar physicochemical properties and hazard characteristics with traditional BFRs [19]. However, no comprehensive regulations currently govern the use of NBFRs.

For CFRs, Dechlorane Plus (DP) and chlorinated paraffins (CPs) are the most widely used. In January 2018, DP was listed as the 18th “Substance of Very High Concern (SVHC)” by the European Chemicals Agency (ECHA) [20]. CPs are categorized based on carbon chain length into short-chain (C10-13, SCCPs), medium-chain (C14-17, MCCPs), and long-chain (C > 17, LCCPs) chlorinated paraffins. SCCPs exhibit the highest potential toxicity, particularly to aquatic organisms [21]. In 2017, SCCPs were included in Annex A of the *Stockholm Convention* and are now regulated globally. While halogenated FRs have been widely used in the past, concerns over their environmental impact have led to a gradual shift towards halogen-free FRs in recent years.

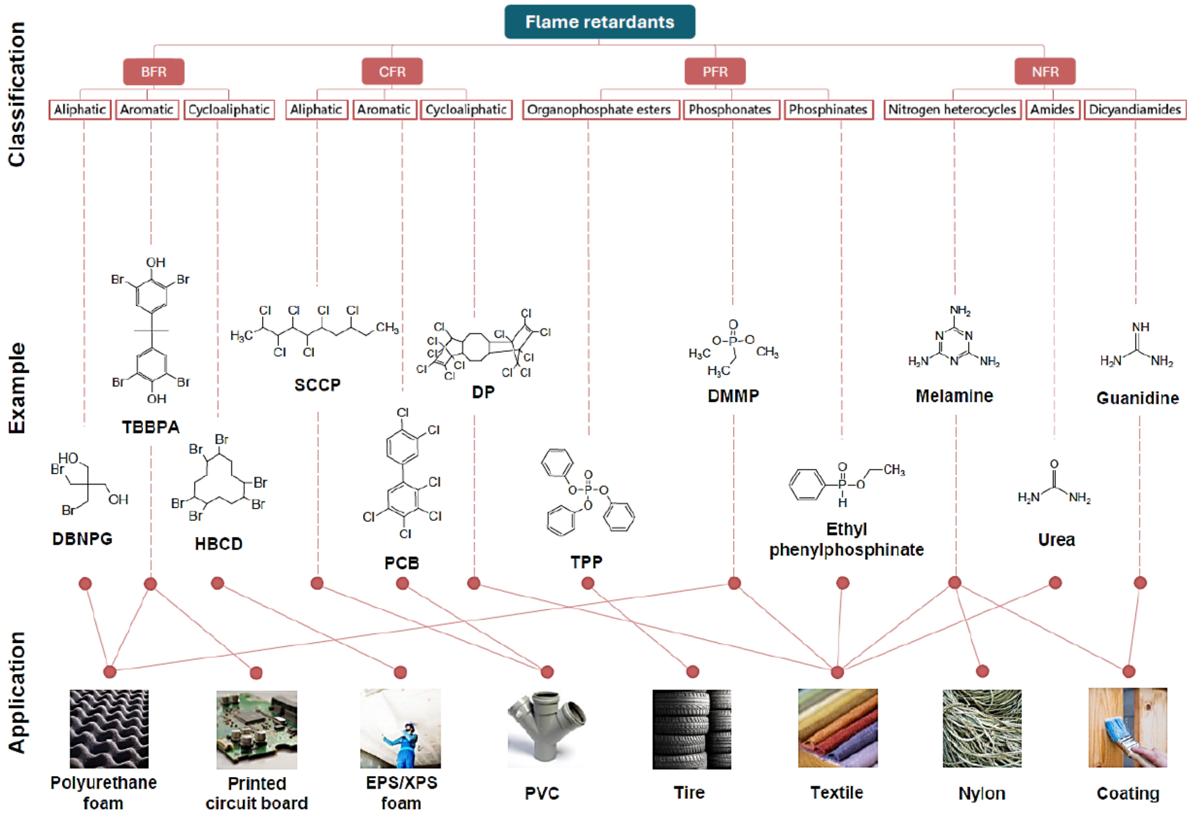

Figure 1. Flame retardant classification. (Dibromoneopentyl Glycol(DBNPG); Polychlorinated biphenyls(PCB); Triphenyl Phosphate (TPP); Dimethyl methyl phosphonate (DMMP)).

Halogen-free organic FRs can be broadly classified into PFRs and NFRs. These two types are often used synergistically to enhance the flame retardancy of materials. Based on the structure of phosphorus, PFRs can be categorized into three subgroups: organophosphate esters (OPEs), phosphonates, and phosphinates [8], with OPEs being the most commonly applied. In addition to structural classification, many studies prefer to categorize PFRs based on the presence or absence of halogens, dividing them into halogenated or non-halogenated PFRs. Generally, halogenated PFRs pose greater hazards, driving a trend toward replacing them with non-halogenated PFRs. PFRs are widely used in materials such as epoxy resins, unsaturated polyesters, polyurethane (PU) foams, and textiles.

The classification of NFRs is less defined and is rarely discussed in the literature. This review categorizes NFRs into three groups based on their structure: nitrogen heterocycles, amides, and dicyandiamides. Among these, melamine-based compounds are the most established NFRs and are commonly combined with PFRs. NFRs are characterized by their low toxicity, corrosiveness, and low smoke production, making them suitable for circuit breakers, public transportation applications, polyurethane flexible foams, nylon, and textiles [22–24]. While NFRs are generally safer than halogenated FRs, their applications are limited. They demonstrate excellent flame-retardant performance in certain materials, particularly nylon-based plastics, which are well-suited to the action mechanism of NFRs. However, in other materials, the flame-retardant efficacy of NFRs may be significantly reduced, failing to achieve comparable performance levels [25].

2.1. Flame Retardant Mechanisms

The flame-retardant mechanisms of organic FRs can be divided into two primary approaches: gas-phase action and condensed-phase action. Gas-phase active FRs decompose at high temperatures to release free radicals, inhibiting the free-radical chain reactions in the flame reaction zone. In contrast, condensed-phase active FRs suppress pyrolysis by promoting charring, intumescence, and the formation of protective barriers[26]. Figure 2 illustrates the flame-retardant mechanisms of different FRs. For BFRs, when exposed to heat or fire, the material releases gaseous bromine (Br_2, HBr, and Br), which reacts with free radicals such as OH· and H·. This reaction interrupts the combustion chain reaction, effectively suppressing the flame propagation[27,28]. CFRs like BFRs belong to the category of halogenated FRs. Their flame-retardant mechanism is similar, relying on gas-phase action to inhibit the combustion reaction.

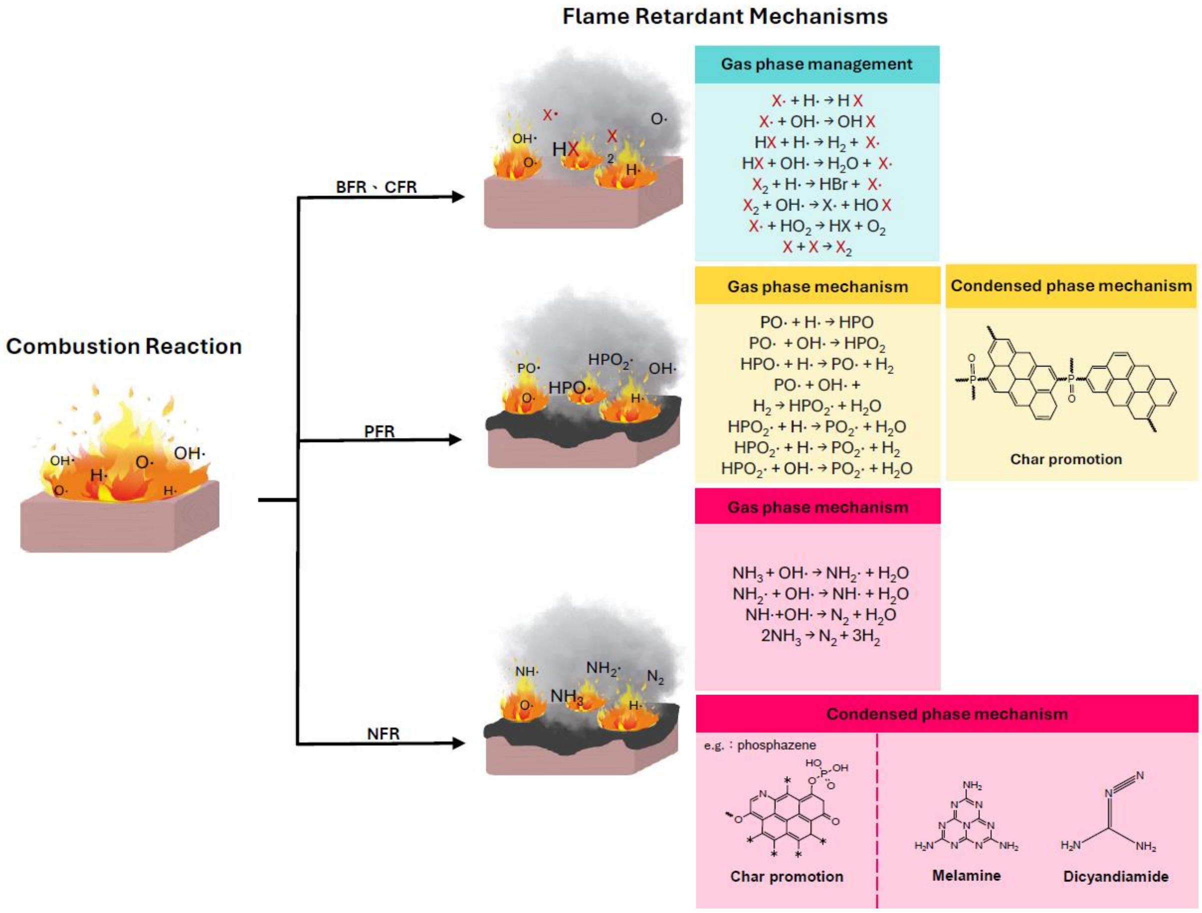

Figure 2. Flame Retardant Mechanisms of Different FRs.

The flame-retardant mechanisms of PFRs operate through two primary modes: (i) gas-phase action and (ii) condensed-phase action. In the condensed phase, phosphates promote dehydration reactions in polymers, leading to char formation. The resulting char acts as a physical barrier to heat and mass transfer, significantly impeding the flow of heat and combustible fragments between the underlying polymer and the combustion zone [29–31]. In most cases, the flame-retardant mechanisms of PFRs involve gas-phase and condensed-phase actions simultaneously. This dual mechanism enhances flame retardancy and reduces toxic gas emissions. The volume of harmful gases released by PFRs is significantly lower than that of BFRs, leading to advocacy for PFRs as safer alternatives to BFRs.

NFRs can function in both the gas phase and the condensed phase. At high temperatures, NFRs release non-flammable gases, such as nitrogen and ammonia, which dilute combustible gases and inhibit the chain reactions of combustion [32,33]. In the condensed phase, two main mechanisms are observed: (i) Formation of a char layer. This typically occurs in phosphorus/nitrogen synergistic FRs. Phosphates create a dehydrating environment, while the presence of nitrogen enhances the charring reaction [34]. (ii) Formation of high-stability products. When melamine decomposes above 350 °C, it releases non-flammable ammonia gas and generates thermally stable compounds such as melamine and dicyandiamide, which improve the thermal stability of polymers [35].

2.2. Release of FRs during Normal Use

FRs are widely used in various consumer products and are released in trace amounts during use. Generally, the concentration of FRs indoors is higher than outdoors, and the types of FRs detected indoors are often associated with the products present. For example, in a study by Vojta, Melymuk, Klánová [36], significant increases in the concentrations of hexabromobenzene (HBB) and tris(2-chloropropyl) phosphate (TCIPP) were observed during computer installation and operation. Similarly, the addition of certain furniture and carpets significantly affected the levels of tris(1,3-dichloro-2-propyl) phosphate (TDCIPP) and tris(methylphenyl) phosphate (TMTP). However, this correlation is not absolute. The types of FRs used in similar products may vary, and a single product can often contain multiple FRs [37,38]. Furthermore, the formulation details of FRs in consumer products are typically not disclosed. As a result, pinpointing the exact sources of FRs in indoor environments is often challenging.

Regarding the mechanism of flame retardant migration from products to dust, Rauert, Lazarov, Harrad, Covaci, Stranger [39] proposed three hypotheses: (i) FRs volatilize from the product and settle into dust, (ii) wear and tear during product use cause the physical transfer of FRs, and (iii) FRs are removed from products through direct contact with dust on the product surface. Depending on the physicochemical properties of the FRs, these mechanisms apply to varying degrees for all FRs.

There are many factors that influence the extent of flame retardant release, and these may vary depending on usage methods, environmental conditions, and product age. Generally, additive FRs are more likely to be released into the environment because their bond with the polymer is less stable than reactive FRs. Additionally, the working temperature of the product has a significant impact. Higher emission rates are often observed in high-temperature indoor environments or when electronic products operate [40]. Kemmlein et al. [41] pointed out that an increase in temperature (to 60 °C) can lead to a 500-fold increase in the emission of various FRs. In a study by Carlsson et al. [42], a relationship between flame retardant emissions, operating temperature, and product age was also observed. After operating a video display unit at a normal temperature (50 °C) for one day, the concentration of triphenyl phosphate in the air rose to nearly 100 ng/m^3. However, after 183 days, it decreased to approximately 10 ng/m^3. Although the concentration decreased significantly, it was still ten times higher than the background value.

In indoor environments with high flame-retardant materials, such as public facilities, emissions may be even greater than in typical households. Takigami et al. [43] conducted a study in a hotel, collecting eight dust samples from different floors and analyzing substances such as BFRs and PFRs. The results showed that PBDEs and HBCD dominated the BFRs, with concentrations ranging from 9.8 to 1700 ng/g (median: 1200 ng/g) and 72 to 1300 ng/g (median: 740 ng/g). Additionally, the study highlighted that the concentration of flame retardant compounds varied across different areas, suggesting a link between the location of the source products and the concentration of FRs in the dust. Similarly, substantial differences in flame retardant concentrations and indoor environmental conditions were observed across different countries.

Among the four types of FRs discussed in this review, NFRs are less frequently mentioned in the literature regarding their emissions in indoor environments. Specifically, most studies focus on the food sector, melamine, and its derivatives. This is due to incidents in 2007–2008 when multiple countries experienced outbreaks of kidney stones and acute kidney injuries in pets and humans as a result of melamine ingestion [44]. Although there is relatively less research on environmental emissions of NFRs, it is not absent. Zhu and Kannan [45] investigated melamine and its derivatives in indoor dust across 12 countries, finding detectable levels in all dust samples. Among these, melamine was the dominant substance, followed by cyanuric acid. The global median concentrations were as follows: melamine 1800 ng/g, cyanuric acid 1100 ng/g, ammeline 48 ng/g, and ammelide 45 ng/g.

In conclusion, many factors influence the concentration of FRs in indoor environments, making it difficult to define the exact emission levels of various FRs. However, we can confirm that even under normal indoor temperature and usage conditions, FRs are released in trace amounts.

2.3. Reaction Temperature and Products

Different types of FRs have varied applications, which may be related to their properties and the temperatures at which they are used. Generally, BFRs are suitable for temperatures ranging from 105 °C to 300 °C [46,47]; CFRs are effective at temperatures below 285 °C [48], PFRs are appropriate for temperatures between 275 °C and 450 °C [49,50], and NFRs are used within a range of 250 °C to 450 °C, with the majority concentrated around 300 °C [23,51,52].

When FRs are heated, other by-products are formed in addition to the target products of the flame-retardant mechanism. Some of these byproducts can be particularly undesirable. Balabanovich et al. [53] mentioned that before HBr is released from BFRs, aliphatic compounds and ketones are first formed, followed by the release of phenols and bromophenols, and eventually, HBr is emitted. Furthermore, heating BFRs does not completely convert them into HBr; in addition to the possible conversion of highly BFRs into lower-brominated ones, brominated phenols, benzene compounds, and by-products such as PBDD/F can also be generated [54,55]. Some studies suggest that BFRs can act as precursors for PBDD/F. Due to their structural characteristics, certain BFRs (such as PBDEs) can directly convert into PBDD/F through simple condensation or other elimination steps. In contrast, most BFRs require more complex mechanisms to form these compounds [27]. The by-products generated during thermal degradation are equally toxic to living organisms, which is one of the reasons why some researchers have proposed replacing BFRs with PFRs. Different types of BFRs can result in varying proportions of by-products. Liang et al. [56] pointed out that highly brominated PBDEs favor the cleavage of ether bonds to form polybrominated benzenes, whereas less brominated PBDEs are more likely to transform into PBDD/Fs. Wang et al. [57] studied the formation mechanisms of PBDD/Fs across 65 PBDE congeners. The authors noted that the presence of polymers lowers the optimal formation temperature of PBDFs from 600 °C to 350–400 °C. This phenomenon may be attributed to: (i) the decomposition of polymers providing unsaturated brominated hydrocarbons, eliminating the need for de novo synthesis of PBDD/Fs; and (ii) the free radicals generated during polymer chain scission, which facilitate the initial degradation of PBDEs. Both CFRs and BFRs belong to the halogenated FRs, sharing similar characteristics. The combustion of CFRs produces small toxic gas molecules (e.g., HCl and phosgene), chlorinated aromatic compounds (e.g., trichlorobenzene and chlorobenzene), and highly toxic substances such as PCDD/Fs [58].

The combustion of the other two types of FRs poses significantly less harm. PFRs generate phosphoric acid derivatives that may cause respiratory irritation but are generally not highly toxic upon heating. Purser [59] review mentioned that any phosphorus source combined with trimethylol-based polyols can produce bicyclic phosphates with strong neurotoxic effects in combustion by-products. However, the associated risks are relatively low since trimethylol-based polyols are not widely used. Among PFRs, halogen-containing variants are more hazardous, as they can generate chlorine-containing by-products like those derived from CFRs.

Upon heating, NFRs primarily produce nitrogen gas, ammonia, and certain stable condensed-phase products during combustion. However, they may also generate NOx and HCN, which are rarely discussed in the literature. HCN is highly toxic, inhibiting the cellular respiratory chain, ultimately leading to organismal death. That said, the amount of HCN produced can be significantly reduced. In their review, Singh and Jain [60] highlighted that adding ammonium polyphosphate (APP) to polyurethane foams can significantly reduce HCN emissions.

3. Thermal Treatment Technology

Under high-temperature conditions, the thermal degradation of BFRs generally progresses through several stages. Initially, the molecular structure of BFRs begins to break down, forming small molecules and free radicals. As degradation continues, a debromination reaction occurs, where bromine atoms are removed from the structure, resulting in debrominated intermediates such as aromatic hydrocarbons or other organic compounds. During this process, highly toxic dioxin-like compounds may form, and a small amount of partially degraded residues may remain. Figure 3 illustrates the thermal degradation pathways of pure TBBPA [60]. Seven primary dissociation pathways are identified: (1) cleavage of the C-CH3 bond, (2) monomolecular transfer of the phenolic hydrogen leading to bromine (Br) removal, (3) direct cleavage of the C-Br bond, (4) rupture of isopropylidene linkages accompanied by hydrogen migration, (5) cleavage of phenolic O-H bonds, (6) breaking of aromatic ring bonds, and (7) loss of hydrogen atoms from methyl groups. Among these, the cleavage of the C-CH3 bond requires the least energy and is thus the predominant pathway.

CFRs and BFRs belong to the halogenated flame retardant category, and their decomposition processes are broadly similar. During the thermal degradation of highly chlorinated paraffins (CP70) (Figure 4), a variety of substances are generated under different thermal conditions, including significant amounts of SCCPs, MCCPs, unsaturated analogs, and toxic chlorinated aromatic compounds [61]. SCCPs readily undergo further breakdown through dehydrochlorination, followed by cyclization or aromatization. Between 200 °C and 400 °C, this process predominantly breaks down PCBs. At higher temperatures (above 500 °C), asymmetric chain cleavage becomes the dominant reaction, generating more small molecules or free radicals, accelerating the addition reactions, and forming larger chlorinated aromatic compounds.

Currently, there is limited research summarizing the thermal degradation pathways of PFR and NFR, likely because their associated hazards are less significant compared to halogenated FRs. Nevertheless, at high temperatures, PFR and NFR undergo several critical thermal degradation steps. Initially, chemical bonds break, leading to the formation of small molecules and free radicals. Phosphorus compounds like phosphates may decompose into phosphoric acid, nitrogen oxides, and other organic by-products. During dephosphorylation or denitrogenation, phosphorus or nitrogen atoms may be removed from the molecular structure, producing degradation products that are more susceptible to further breakdown.

Thermal treatment technology is currently the most common method for waste disposal, offering a direct, rapid, and practical approach to significantly reduce the mass (by approximately 70–80%) and volume (by about 80–90%) of solid waste [62]. General thermal treatment processes include incineration, pyrolysis, gasification, and hydrothermal processes. However, a significant drawback of these thermal treatment techniques is that FRs may not be destroyed after heating. Some FRs can persist in the gas phase, particulate phase, or liquid phase, and under high-temperature conditions, they may transform into other harmful by-products, particularly derivatives of PCDD/F.

Table 2 provides a comparative overview of four major thermal treatment technologies, focusing on key parameters such as operational conditions, expected products, and post-treatment requirements. These factors are critical for evaluating flame retardants' behavior and ultimate fate under various waste-to-energy scenarios.

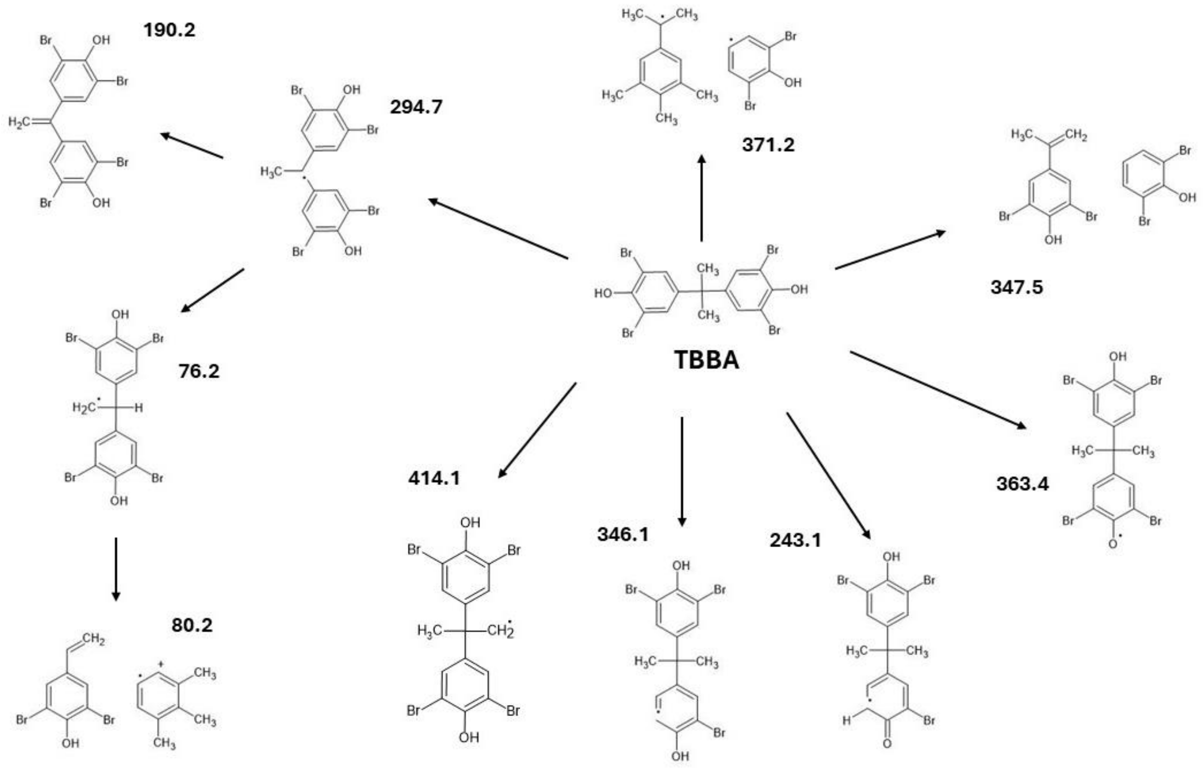

Figure 3. TBBA thermal degradation pathway.

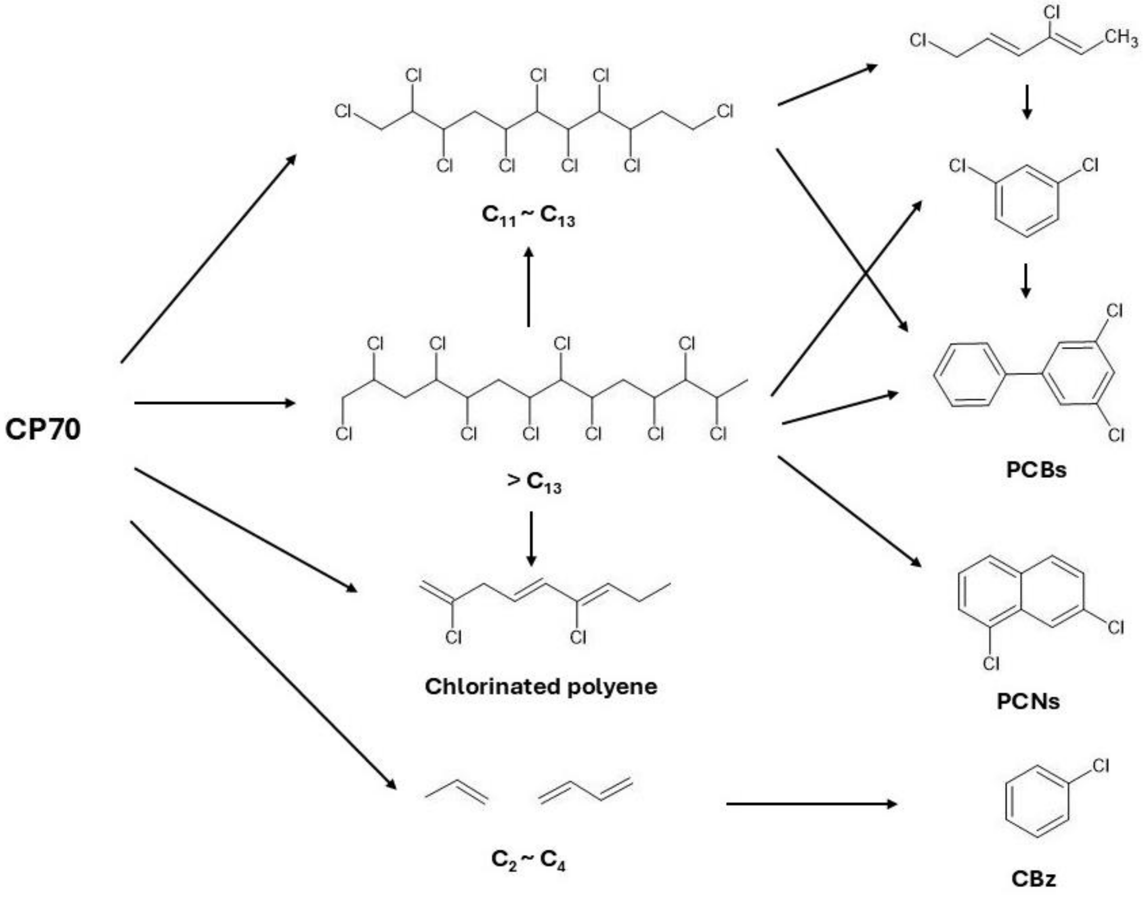

Figure 4. Thermal degradation of highly chlorinated paraffin (CP70).

Table 2. Heat treatment technology.

Treatments	Operating Conditions	Feed Conditions	Products	Post-Treatment Requirements
Incineration	• T [a] = 800–1200 °C • O_2 present • P [a] = 0.1 MPa • RT [b] = seconds	• Solid particles • Moisture < 20% wt • Calorific value above 8–10 MJ/kg	• Flue gas • Ashes	• Flue gas control • Ash stabilization and disposal
Pyrolysis	• T [a] = 300–900 °C • Nearly no O_2 • P [a] = 0.1–5 MPa • RT = seconds	• High-carbon feedstock • Solid particles • Moisture < 20% wt	• Flue gas • Biochar • Bio-oil* • Ashes	• Oil purification • Biochar utilization
Gasification	• T [a] = 700–1400 °C • O_2-deficient (limited O_2, CO_2, steam, or air) • P [a] = 0.1–5 MPa • RT = seconds	• High-carbon feedstock • Solid particles • Moisture < 20% wt	• Syngas* • Ashes • Tar	• Product purification • Tar recycling
Hydrothermal	• T [a] = 180–350 °C • Aqueous reaction • P [a] = 5–22 MPa • RT = seconds	• Low-salinity wet feedstock • Solid particles	• Wastewater • Biochar • Bio-oil • Gas	• Product purification • Wastewater treatment • Biochar/bio-oil utilization

Note: [a] T: operation temperature; [b] P: operation pressure; [c] RT: resident time; * Major product.

3.1. Incineration

Incineration is the most common thermal treatment method and a well-established commercial process, making it one of the primary waste management techniques today. It operates at high temperatures of approximately 800 to 1200 °C to ensure the complete combustion and decomposition of the most harmful substances. However, the inevitable generation of substances such as polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), and polychlorinated dioxins (PCDDs) presents significant challenges, particularly in the control of pollutant gas emissions and the management of ash residues. Many studies indicate that the destruction rate of FRs during incineration can typically exceed 99%. Any undestroyed FRs are usually detected in the residual ash after combustion, while their presence in the flue gas is relatively minimal.

For instance, Sakai, Watanabe, Honda, Takatsuki, Aoki, Futamatsu, Shiozaki [63] conducted a study on the combustion of BFRs, using actual waste materials (such as the casings of discarded televisions and waste printed circuit boards) and custom samples (polyethylene resin containing PBDEs and ABS resin containing TBBP-A). After processing, they analyzed the concentration of PBDEs, finding residual concentrations in the ash ranging from 2.9 to 180 μg/g, which is 1 to 3 orders of magnitude lower than the concentrations in the original samples. PBDEs were detected in the flue gas only in one instance, with other measurements falling below detection limits. While PBDEs were almost nonexistent in the flue gas, by-products such as PBDD/DF, PCDD/DF, and PXDD/DF were detected.

The high destruction rates of FRs during incineration are also evident in the research conducted by Matsukami et al. [64]. They tested refuse-derived fuel (RDF) containing four different PFRs in a pilot-scale incinerator. They found detectable PFRs in the flue gas and ash, with overall destruction efficiencies exceeding 99.999%. The authors noted that PFRs were primarily destroyed during the initial combustion stage, indicating that the conditions in the primary combustion chamber are critical for the degradation and emission control of PFRs. Kwon et al. [65] conducted incineration of waste containing CFRs and found that CFRs were not detected in the flue gas at both 1100 °C and 850 °C. They also investigated dioxin formation under these two conditions, revealing that at a reaction temperature of 1100 °C, emissions were generally below the current limit value (5 ng I-TEQ/Sm^3). Consequently, the authors recommended adoption of strategies that minimize the risk of unintentionally generating dioxins and other persistent organic pollutants, operating temperatures should be maintained above 1100 °C. Research on the incineration of NFRs remains relatively limited, likely due to their lower environmental risk and certain application constraints. Current studies focus on developing novel NFRs to enhance their flame-retardant efficiency, applicability, and thermal stability, enabling broader use.

An essential aspect of thermal treatment technology is controlling by-product formation. Various methods are available to manage concentration, such as adding specific substances during the thermal process. For instance, adding chlorine, nitrogen, CaO, or coal can influence the formation of polychlorinated dibenzodioxins/furans (PCDD/Fs) [66]. However, the potential for these additives to trigger other adverse effects and their applicability in practical operations requires further investigation. Additionally, research by Sakai et al. [63] has shown that mixtures of metals and higher cooling temperatures (300 °C) may promote the formation of PBDD/DF.

Waste composition is highly complex, and completely removing metal compounds necessitates screening and pretreatment, often posing practical challenges and significantly increasing treatment costs. Therefore, control measures must be implemented from other perspectives. For example, Yang et al. [67] successfully reduced the emissions of brominated persistent organic pollutants by over 97%, including PBDD/F and PBB emissions, and achieved a 61% reduction in PBDE emissions through controlling the startup of waste-to-energy incinerators.

While incineration technology can effectively destroy most FRs, some residues may still pose environmental threats due to the characteristics of the FRs. Additionally, the by-products generated during incineration require careful attention to prevent further environmental contamination. Kwon et al. [65] incinerated waste containing CFRs and did not detect CFRs in the exhaust gases at 1100 °C and 850 °C. They also investigated dioxin formation under these two conditions, and the results showed that at a reaction temperature of 1100 °C, the dioxin levels were generally below the current emission limit (5 ng I-TEQ/Sm^3). Therefore, the authors recommended that to reduce the potential for unintentional formation of persistent organic pollutants such as dioxins, the operating temperature should be above 1100 °C.

3.2. Pyrolysis

Unlike incineration, pyrolysis is conducted under anaerobic conditions, typically at 200 to 700 °C. It requires lower energy and converts biomass/biosolids into valuable products during heating. Compared to incineration, it does not emit large amounts of greenhouse gases, reducing gas generation by approximately 5% to 10% [68]. The generation of PCDD/F can also be reduced through pyrolysis. A study by Chen, Sun, Li, Lin, Xiang, Yan [69] indicated that the PCDD/F yield from the pyrolysis of waste tires was significantly lower than that in combustion flue gas, nearly reducing it by 100%. Due to these characteristics, pyrolysis has attracted considerable attention in recent years.

The products of pyrolysis can be categorized into three types: flue gas, biochar, and bio-oil. Among these products, oil constitutes the majority (50 wt%–70 wt%), followed by biochar (13 wt%–25 wt%), while gas production is the least (12 wt%–15 wt%) [69]. Therefore, pyrolysis easily leads to the accumulation of pollutants in the oil phase [70,71]. Ye et al. [72] pyrolyzed waste-printed circuit boards (WPCBs) containing BFRs at 500 °C and measured the organic bromine content in the oil phase at 151.13 mg/g. However, by adding certain substances to reduce the bromine content, varying proportions of composite additives (Fe_3O_4 and Si-Al zeolite) effectively lowered the bromine levels in the pyrolysis oil (10.45 to 20.24 mg/g). However, by adding different proportions of composite additives (Fe_3O_4 and Si-Al zeolite), the bromine content in pyrolysis oil was effectively reduced to between 10.45 and 20.24 mg/g [73]. Conducted pyrolysis studies on cotton fibers with and without phosphorus-nitrogen FRs. The results showed that at a pyrolysis temperature of 600 °C, the pyrolysis rate of cotton fibers without FRs reached 97.23%, while that of fibers containing FRs was only 76.53%. This indicates that the presence of FRs inhibited the pyrolysis reaction. Additionally, it was found that the yields of compounds such as ketones, aldehydes, esters, and ethers in the pyrolysis products significantly decreased, with mainly cyclic compounds like furan and glucan being produced. In a study on the pyrolysis of polyurethane (PU), Eschenbacher et al. [74] investigated the effects of TCPP (tris(1-chloro-2-propyl) phosphate) on the pyrolysis process. The results showed that when the pyrolysis temperature exceeded 700 °C, TCPP was no longer present in the products, indicating that it completely decomposed at high temperatures. Additionally, the study found that the formation of chlorine-containing products peaked at around 600 °C. Chen et al. [75] examined the distribution of nitrogen-containing products during PU pyrolysis and discovered that hydrogen cyanide (HCN) and ammonia (NH_3) were predominant in the gas phase, while the liquid phase contained amines, nitrogen heterocycles, and nitriles. Nitrogen oxides were nearly undetectable in the solid phase. Adding metal oxides or catalysts like CaO during pyrolysis promoted the formation of nitrogen gas (N_2) while suppressing the generation of NH_3 and HCN, contributing to a more environmentally friendly pyrolysis process. Recent advancements in flame retardant treatment through pyrolysis have also emerged. Cho et al. [76] demonstrated a method to increase the value of melamine using pyrolysis. Under CO_2 and Ni-catalysis conditions, melamine was completely converted into gaseous pyrolysis products, with an increased concentration of carbon monoxide (CO). This experiment provides an efficient approach to transforming melamine waste into valuable energy resources, such as syngas. Kumagai et al. [77] investigated the effects of calcium hydroxide ($Ca(OH)_2$) on the thermal decomposition of both phenol and epoxy resin paper-laminated printed circuit boards (PCBs) containing TBBPA. Their pyrolysis experiments revealed a maximum removal of 94% HBr, 98% brominated phenols, and 98% phosphorus from the gaseous and liquid products. Additionally, the inhibition of Br-induced metal volatilization improved the recovery rate in the solid fraction.

Charitopoulou et al. [78] evaluated the pyrolysis performance of waste electrical and electronic equipment containing TBBPA using five different catalysts: Al_2O_3, Fe/Al_2O_3, MgO, Fe/MgO, and zeolite. The study found that all the catalysts promoted the formation of phenolic compounds, valuable products for the chemical industry. Among the catalysts, Fe/Al_2O_3 was identified as the most effective, achieving a bromine reduction rate of over 75%. However, using catalysts introduces additional costs and poses the risk of secondary pollution, making catalytic methods less optimal for practical applications. Chen et al. [79] explored the co-pyrolysis of two types of waste, red mud and waste printed circuit boards, successfully fixing 89.55 wt% of the bromine in the solid residue and increasing the yield of light tar by 44.29%. Beyond the use of co-pyrolysis and catalysts, adjusting pyrolysis conditions may further enhance pollutant degradation, such as employing staged pyrolysis. Staged pyrolysis shows significant potential in regulating the formation of nitrogen-containing gaseous pollutants from agricultural biomass waste. Zhan et al. [80] demonstrated that, compared to single-stage pyrolysis at the appropriate temperature, two-stage pyrolysis reduced total nitrogen-containing pollutant yields by 57% to 60% for three different samples. Currently, research on the effects of staged pyrolysis on flame-retardant materials is still limited, but this approach may facilitate more efficient degradation of FRs, improving environmental performance.

3.3. Gasification

Gasification is similar to pyrolysis. It occurs at high temperatures (700–1000 °C) in a partial oxygen environment, converting materials into various gaseous components such as H_2, CO, CO_2, CH_4, and C_2H_6, which can be used as fuel. In addition, by-products like biochar and tar are also produced [81].

Research on the gasification of flame-retardant-containing materials is noticeably less prevalent compared to studies on pyrolysis and incineration. Sodium phytate, a potential material for PFRs, was investigated by Lidman Olsson et al. [82]. Their study examined the release behavior of sodium phytate during thermal treatment. It was found that at 1000 °C under an inert atmosphere, approximately 30% of phosphorus was released in the gaseous phase. This finding provides valuable insights into the characteristics of phosphorus release during the gasification process. In 2003, Yamawaki et al. [83] employed high-temperature (over 1200 °C) treatment combined with rapid cooling to suppress PBDDs/PBDFs emissions to very low levels. Although the authors did not explore the degradation of FRs, studies on incineration and pyrolysis indicate that thermal treatment technologies generally achieve high removal rates for FRs, though the generation of by-products is often unavoidable. Yamawaki et al.'s methodology significantly reduced the formation of PBDDs/PBDFs potentially generated by FRs, highlighting the potential of gasification in minimizing harmful by-products. In 2024, Lo et al. [84] suppressed hydrochloric acid and dioxin emissions through the co-gasification of calcium-containing waste and automobile shredder residue. Most studies on gasification focus on gas yield, tar formation [85–87], and the emission of certain pollutants, such as HCl and polychlorinated dibenzodioxins/furans (PCDD/F) [88]. Few investigations have delved deeply into the relationship between FRs and gasification. One reason for this gap may be that gasification requires higher temperatures than pyrolysis, which results in a lower risk of pollutant release and by-product formation from FRs. Additionally, gasification technology is considered an effective method for reducing pollutants, as it primarily aims to convert waste into syngas. This focus on waste-to-energy conversion has contributed to the relative scarcity of research on FRs in gasification processes.

3.4. Hydrothermal Treatment

Hydrothermal technology operates at medium to low temperatures (180–375 °C) and under high-pressure conditions, converting biomass into valuable products. The primary products can be solid fuels or bio-oil, depending on the operating conditions. Compared to traditional thermal treatment methods, Hydrothermal carbonization technology has the significant advantage of processing high-moisture feedstocks without requiring dewatering, which substantially reduces pretreatment costs [89].

Uddin et al. [90] used hydrothermal treatment to remove decabromodiphenyl ether (DBDE) from plastics. Their study found that at 280 °C, the process achieved high plastic recovery rates and optimal debromination, with most bromine extracted into the water phase as HBr.

Although PBDD/DFs formation can occur during hydrothermal treatment, the associated risks are relatively low. The process can be improved by adding certain catalysts, alkaline agents, or extending the treatment time [91]. Previous studies have shown that an alkaline environment facilitates debromination reactions, a phenomenon also observed by Yin et al. [92]. At temperatures above 300 °C, with a residence time over 30 min and the presence of alkaline additives, more than 80% of brominated epoxy resin can be broken, primarily into phenol. Zhan et al. [93] further enhanced hydrothermal debromination by introducing an alkaline sulfide system, achieving extraction efficiencies of 85.60% for Sb and 90.13% for Br from flame-retardant plastics. Moreover, the plastic structure remained largely intact under optimal hydrothermal conditions, allowing for potential reuse.

However, the transformation of Cl during the hydrothermal process remains less understood. While co-hydrothermal technology has successfully removed Cl from the solid-phase products, challenges persist in managing Cl in the liquid phase to advance the practical application of this technique [94]. Xiu et al. [95] proposed an efficient dechlorination method for short-chain chlorinated paraffins using subcritical water with NaOH. This method achieved 100% dechlorination at low temperatures within a short reaction time (250 °C, 5 min), producing high-value hydrocarbons. However, the study did not use plastic waste as feedstock, indicating potential challenges in real-world applications. Thus, further exploration is required to optimize dechlorination processes for waste materials.

Currently, research on the hydrothermal treatment of PFR and NFR remains limited, likely because the degradation products of these substances pose lower environmental risks and are relatively easier to manage. Although studies specifically on PFR and NFR are scarce, considerable research has examined the transformation of phosphorus and nitrogen in waste materials. Phosphorus typically first converts to water-soluble phosphate ions (e.g., orthophosphate), which then precipitate by reacting with metal ions present in sludge or waste under high-temperature, high-pressure hydrothermal conditions [96–98]. Nitrogen compounds convert into inorganic nitrogen ions (e.g., NH_4^+, CN^-, NO_2^- and NO_3), with some nitrogen retained in the solid phase [99,100].

4. Non-thermal Treatment Technologies

Aside from thermal treatment, flame retardant degradation can also be achieved through chemical methods, photodegradation, microbial degradation, and electrochemical methods, each with its own advantages and drawbacks.

(1) Chemical Degradation

This method efficiently breaks down FRs and allows for some flexibility in controlling the by-products. Common approaches include Fenton reactions, persulfate oxidation, hydrogen peroxide, and ferrate treatments [101–104]. The effectiveness of these chemical agents can vary depending on the specific FRs being treated. For example, Ma et al. [105] tested six organic solvents to degrade phosphonate-based epoxy resins and found that methanol in an alkaline environment provided the best results. However, challenges such as high reagent costs, risks of secondary pollution, and limited selectivity hinder large-scale application.

(2) Photocatalysis

This promising technique uses visible or ultraviolet light to activate catalysts, generating free radicals that decompose pollutants. Despite its potential, photocatalysis faces challenges, such as high material costs and complex modification methods. Additionally, if photocatalysts are not promptly recovered, there may be issues with leaching new contaminants [106]. While this technique is commonly applied for pollutant removal in aqueous and gaseous phases, advancements in nanotechnology have enabled applications in soil, sediments, and waste treatment [107]. Preliminary photocatalytic studies on NBFRs and phosphorus FRs have laid a foundation [108–110]. However, factors such as identifying degradation by-products and changes in solution toxicity remain unresolved.

(3) Biodegradation

This method uses microorganisms or enzymes to break down organic FRs, making it particularly suitable for phosphorus- and nitrogen-based retardants. Hou et al. [111] reported aerobic biodegradation of three halogen-free phosphorus FRs, achieving removal rates between 29.3% and 89.9% after 25 days, with Klebsiella identified as a key degrader. Although environmentally friendly and effective, biodegradation is time-consuming, sensitive to environmental conditions, and may produce more toxic intermediates [112], complicating large-scale implementation. Understanding the biochemical pathways, involved microorganisms, and potential by-products is crucial [113].

(4) Electrochemical Methods

These can be divided into electrochemical reduction and oxidation pathways [114]. Due to the high electronegativity of halogenated compounds, reduction processes are generally more suitable [115]. Electrochemical techniques demonstrate high efficiency in removing organic pollutants Oturan et al. [116] and have also been applied to phosphorus FRs. For instance, Tang et al. [117] used electrochemical oxidation to degrade TDCPP, achieving effective degradation with low-toxicity intermediates. Despite their potential, electrochemical methods require further improvements, especially in electrode materials and efficiency, and they still face challenges related to high application costs and energy consumption.

Dong et al. [118] reviewed emerging catalytic methods for degrading BFRs, noting that most studies are conducted under controlled laboratory conditions with the addition of co-solvents such as methanol, acetone, or acetonitrile, leading to high degradation efficiencies. Compared to traditional thermal treatment, non-thermal methods are often more environmentally friendly and energy efficient. However, limitations include narrow applicability, slow reaction rates, stringent conditions, and incomplete degradation. Further optimization and validation are needed for the practical implementation of non-thermal techniques.

5. Comprehensive Discussion

In practice, thermal treatment techniques are indeed effective for rapidly degrading FRs. Although the purpose of adding FRs seems at odds with thermal degradation, in reality, FRs have minimal impact on the decomposition temperature and mainly influence the composition of gases and the quality of residues during decomposition [119]. Boro and Tiwari [120] suggest that FRs could be removed before thermal degradation. However, given the wide variety of FRs and the complex composition of waste materials, removing them before thermal treatment is challenging.

Thermal treatment effectively reduces waste volume and many pollutants but can also result in pollutants redistributing into other media. Therefore, combining multiple techniques is recommended to minimize the drawbacks of single methods. Among several thermal treatment methods, incineration is generally not the most valuable approach [121]. In contrast, pyrolysis, gasification, and hydrothermal processes are more feasible alternatives, producing fewer pollutants and valuable by-products. Under specific conditions, hydrothermal processing can even desorb pollutants without compromising material properties, providing an additional recovery pathway.

For more complex waste compositions, several treatment approaches are recommended: (i) performing initial thermal degradation, followed by non-thermal techniques for pollutant removal, or concentrating pollutants for further treatment through adsorption; (ii) reducing pollutant emissions by reintroducing waste ash or sludge into the combustion system [122]; (iii) conducting hydrothermal pre-treatment to remove pollutants, then using other thermal processes to generate energy [123].

6. Conclusions

This review provides a comprehensive comparison of thermal and non-thermal technologies for the treatment of flame retardants (FRs), focusing on incineration, pyrolysis, gasification, and hydrothermal treatment. While thermal methods are effective in degrading FRs, they may produce toxic by-products. Non-thermal approaches such as photocatalysis, biodegradation, and electrochemical treatment offer environmentally friendly alternatives but face limitations in efficiency and scalability. This review identifies several key findings:

(i) Flame retardants exhibit markedly different thermal decomposition behaviors under various treatment conditions, with diverse by-product profiles;
(ii) Thermal technologies are generally effective in reducing FR residues but may result in incomplete mineralization and secondary pollution.
(iii) Non-thermal approaches offer lower environmental burdens and serve as valuable supplements to thermal methods, though their practical implementation remains limited by technological constraints.

Based on these insights, this review offers three main contributions:

(i) It presents the first integrative analysis comparing the behavior of multiple FR types—including both halogenated and non-halogenated compounds—across thermal and non-thermal treatment routes;
(ii) It evaluates the applicability and limitations of each method from a waste-to-energy perspective, and proposes the feasibility of multi-technology integration based on waste characteristics;
(iii) It emphasizes the importance of understanding by-product toxicity and transformation mechanisms as key factors in selecting and optimizing treatment strategies.

Future research should focus on bridging the gap between laboratory-scale studies and full-scale applications, optimizing pollution control systems for emerging flame retardants, and evaluating the long-term environmental impacts of transformation products to support safer and more sustainable waste management practices.

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