



Review

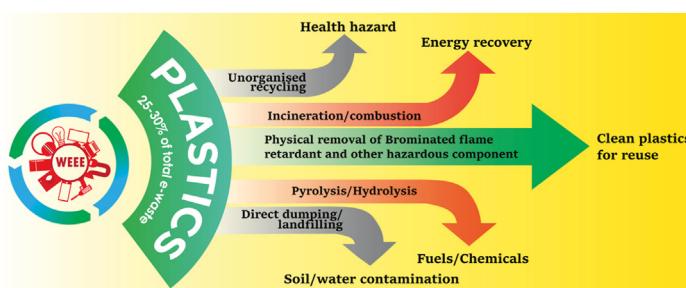
Value-added products from thermochemical treatments of contaminated e-waste plastics

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HIGHLIGHTS

- Recycling of e-waste plastic is essential for economical disposal of e-waste.
- Non-bonded brominated flame retardants can be extracted by organic solvents.
- Metal oxide catalysts are suitable for fixation of Br in the pyrolysis.
- Hydrothermal treatment can remove both Br and Sb from the e-waste plastics.

GRAPHICAL ABSTRACT



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ABSTRACT

The rise of electronic waste (e-waste) generation around the globe has become a major concern in recent times and its recycling is mostly focused on the recovery of valuable metals, such as gold, silver, and copper, etc. However, e-waste consists of a significant weight fraction of plastics (25–30%) which are either discarded or incinerated. There is a growing need for recycling of these e-waste plastics. The majority of them are made from high-quality polymers (composites), such as acrylonitrile butadiene styrene (ABS), high impact polystyrene (HIPS), polycarbonate (PC), polyamide (PA), polypropylene (PP) and epoxies. These plastics are often contaminated with hazardous materials, such as brominated flame retardants (BFRs) and heavy metals (such as Pb and Hg). Under any thermal stress (thermal degradation), the Br present in the e-waste plastics produces environmentally hazardous pollutants, such as hydrogen bromide or polybrominated diphenyl ethers/furans (PBDE/Fs). The discarded plastics can lead to the leaching of toxins into the environment. It is important to remove the toxins from the e-waste plastics before recycling. This review article gives a detailed account of e-waste plastics recycling and recovery using thermochemical processes, such as extraction (at elevated temperature), incineration (combustion), hydrolysis, and pyrolysis (catalytic/non catalytic). A basic framework of the existing processes has been established by reviewing the most interesting findings in recent times and the prospects that they open in the field recycling of e-waste plastics.

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Contents

1. Introduction	2
2. Recycling processes associated with e-waste plastics	4
2.1. Mechanical recycling (primary/secondary) recycling	4
2.2. Tertiary (chemical and thermal) recycling	5
3. Solvothermal processes to remove BFRs from e-waste plastics	5
4. Thermochemical degradation of e-waste plastics	7
4.1. Incineration	7
4.2. Hydrothermal treatment	7
4.3. Pyrolysis	7
4.3.1. Fixation of Br content during catalytic pyrolysis of e-waste plastics	11
4.3.2. Different reactor configurations for e-waste plastic pyrolysis	11
5. Conclusion and outlooks	16
6. Future scope	17
Declaration of competing interest	17
Acknowledgement	17
References	17

1. Introduction

The recycling and disposal of contaminated waste electrical and electronic equipment (WEEE) or electronic waste (e-waste) plastics are challenging environmental issues that need serious attention. Due to the rapid technological advancement in the field of household appliances, mobile communication and computers, the global generation of e-waste keeps on increasing and becomes a burden to the waste management systems (Ma et al., 2016a).

Definition of e-waste comprises of discarded electronic, electrical pieces of equipment, light bulbs, and batteries. The waste generator can be the manufacturer (due to defect or obsolescence) or the consumer when the equipment reached its end of their service life (Robinson, 2009). Fig. 1 shows the trends of the generation of electronic and electrical waste in recent years. An increase of 47% of e-waste generation has been observed from the year 2010–2018 (Baldé et al., 2017). The United Nation's recent assessment reported that about 44.7 million metric ton (MMT) of e-waste was generated globally in the year 2016 and only 20% (8.9 MMT) was documented to be collected and properly recycled (Baldé et al., 2017). The amount of e-waste generation reached 53.6 MMT in 2019 (Forti et al.), which exceeded the projection in the report for 2021 (Forum, 2019). Of the discarded e-waste, 80% goes either into

landfills or incineration.

E-wastes are complex and contain almost 69 elements from the periodic table including precious metals (Au, Ag, Cu, Pt, Pd, Ru, Rh, Ir and Os), critical metals (Co, Pd, In, Ge, Bi and Sb) and non-critical metals, such as Al and Fe (Forti et al., 2020). Metals that have strategic importance and might have a high risk of supply in near future due to the fast depletion of their reserves are considered as critical, whereas metals with abundant stocks are non-critical (Supanchaiyamat and Hunt, 2019). In the era of the circular economy, e-waste is considered as a secondary resource for the mining of these precious metals (Zeng et al., 2018; Awasthi et al., 2019). It was roughly estimated that plastic in e-waste could be around 25–30% of the total e-waste (Gramatyka et al., 2007). The plastics present in e-waste are usually burned during metal extraction (Vehlow et al., 2003; Duan et al., 2011) or end up in landfills despite their intrinsic value, which was estimated to be around 17.6 Billion USD in 2016 (Baldé et al., 2017). A proper plastic recycling scheme could greatly enhance the realization of the circular economy regarding e-wastes. Fig. 2 represents different types of plastics in e-waste streams in the UK. Most common types of plastics in e-wastes are styrene-based polymers, such as acrylonitrile butadiene

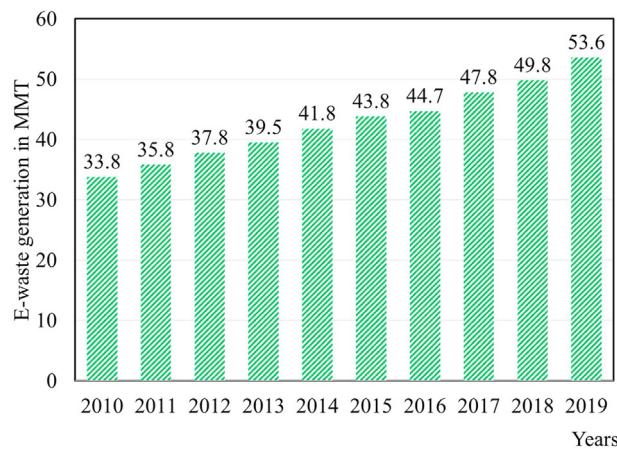


Fig. 1. Trend of worldwide e-waste generation in recent years (Forti et al. Wang, 2018).

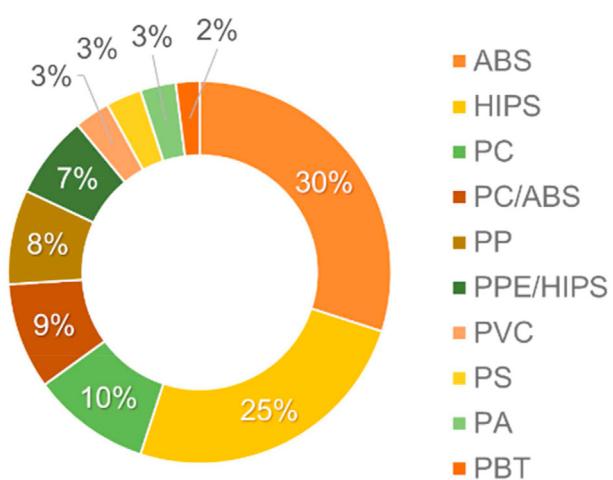


Fig. 2. Typical composition of e-waste plastics in UK, 2006 (Keith Freegard; Alston et al., 2011).

styrene (ABS), polystyrene (PS), high impact polystyrene (HIPS) and epoxy resins (from printed circuit boards). Presence of ABS and HIPS represent more than 55% of all e-waste's plastics. Other polymers, such as polycarbonate (PC), polypropylene (PP), polyethylene (PE), polyvinyl chloride (PVC), polybutylene terephthalate (PBT), polyamide (PA) and poly(methyl methacrylate) (PMMA), can also be found in different e-wastes. The plastics (polymers) used in electronic and electrical equipment (EEE) are products of the petrochemical industry and depolymerisation or control degradation can revert them back to hydrocarbon feedstock or fuel.

E-wastes are generally treated separately from municipal solid waste (MSW) due to their toxic content. Toxic substances, such as Hg, Pb, Se, Cd, hexavalent chromium (Cr(IV)), polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyl, and polybrominated diphenyl (PBD), were found in the area near e-waste recycling and storage facilities across the world (Tang et al., 2010; He et al., 2012; Damrongcharoensathien et al., 2016; Tue et al., 2017). Table 1 listed different toxic compounds in e-waste with their sources.

E-waste plastics often found contaminated with heavy metals (Pb, Hg, Cd etc.) and brominated flame retardants (BFRs). (Tange and Dohmann, 2005; Das et al., 2009; Li et al., 2009; Peng et al., 2010; Damrongcharoensathien et al., 2016; Debnath et al., 2018). The directives of EU on e-waste (commission regulation (eu) no 1342/2014 amendment regulation no – 850/2004) consider the presence of Br in excess of 2000 ppm as persistent organic pollutants (POPs). Over the years, the European Union regulated and restricted various substances in plastics particularly in the case of electronics and electrical goods (E.U., 2015). Table 2 lists restricted substances and their permissible limits in such goods. The science of removal of the toxic chemicals from e-waste plastics has been explored and a number of technologies were investigated that optimize energy and material consumptions. The removal of metal contamination at the molecular level is difficult by mechanical and physical means (Kurose et al., 2006; Li et al., 2007). Although hydrometallurgical processes like leaching assisted with cyanide, thiourea, thiosulfate

Table 2
Restricted substances used in the electronic and electrical goods by EU (E.U., 2015).

Toxic substance	Permissible limit (wt%) ^a
Pb	0.1
Hg	0.1
Cd	0.01
Cr (IV)	0.1
Polybrominated biphenyls (PBB)	0.1
Polybrominated diphenyl ethers (PBDE)	0.1
Bis(2-ethylhexyl) phthalate (DEHP)	0.1
Butyl benzyl phthalate (BBP)	0.1
Dibutyl phthalate	0.1
Diisobutyl phthalate (DIBP)	0.1

^a 0.1 wt% = 1000 ppm and 0.01 wt% = 100 ppm.

or halide have been developed mainly to recover precious metals from waste printed circuit boards (PCBs) (Cui and Zhang, 2008; Zhang et al., 2012), they can also be used for the removal of contaminated metal from e-waste plastics. The non-metallic part of the PCBs is most toxic as it contains traces of solders made from Pb/Sn and BFRs. Although, recent PCBs are lead free solder but most old PCBs have 2–5% of Pb in their solder (Ghosh et al., 2015).

Flame retardants (FRs) are chemical substances added to consumer goods to prevent the start or slow the growth of the fire. These are essential to electronic and electrical pieces of equipment due to the requirements from fire safety codes. BFRs are the most common FR used in electronic and electrical equipment (EEE), which are made from the family of organobromine compounds. The BFRs are relentless, bio-accumulative and toxic in nature, and their decomposition pathways during thermal treatment have been reported previously (Altarawneh et al., 2016; Ma et al., 2016a). The treatments to recover BFR free plastics or products are discussed in the subsequent sections. Most abundantly used commercial BFRs were polybrominated diphenyl ethers (PBDEs), tetrabromobisphenol A (TBBPA), hexabromocyclododecane (HBCD),

Table 1
Chemical classification of toxins from e-waste and their source (Grant et al., 2013).

Chemical species/toxins	Source in e-waste
POPs	
BFRs	Fire retardants for electronic equipment
Polybrominated diphenyl ethers	Dielectric fluids, lubricant and coolants used in different equipment with moving parts
Polychlorinated biphenyls	
Dioxins	
Polychlorinated dibenzodioxins and dibenzofurans	Released during heat treatment (combustion/pyrolysis) as a by-product
Dioxin-like polychlorinated biphenyls	Combustion/pyrolysis by-product but also found in dielectric fluid and lubricants
Perfluoroalkyls	Fluoropolymers
PAHs	
Naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benzo(a)anthracene, chrysene, benzo(a)pyrene, benzo(b)fluoranthene, benzo(k)fluoranthene, indeno(1,2,3-cd)pyrene, dibenzo(a,h)anthracene, benzo(g,h,i)perylene	Released during heat treatment (combustion/pyrolysis) as a by-product
Elements	
Pb	PCBs, cathode ray tubes (CRTs), light bulbs, television, batteries, electric wires and insulations
Cr (IV)	Anticorrosion coatings, floppy disc, data tapes
Cd	PCBs, switches, semiconductor chips, CRTs, batteries, copy machine and mobile phones
Hg	Thermostats, sensors, LCD monitors, PCBs, fluorescent lamps
Zn	PCBs, CRTs, coatings, plastic additives
Ni	Batteries
Li	Batteries
Ba	CRTs and fluorescent lamps
Be	power supply boxes, computers, x-ray machines, ceramic components of electronics

decabromodiphenyl ether (deca-BDE), etc., however, due to environmental safety, the use of these BFRs was restricted and they have been replaced by less harmful ones based on nitrogen, phosphorous and other inorganics (Singh and Jain, 2009). However, BFR is still extensively used and found in the e-waste plastics. BFRs such as PBDEs have been linked with thyroid cancer as its structure resembles thyroid's hormones (triiodothyronine and thyroxin) (McDonald, 2002; Li et al., 2018). During any heat treatment (pyrolysis/combustion), the BFRs in the e-waste plastics can produce brominated aromatic compounds which are precursors to the polybrominated dibenz-p-dioxins (PBDD) and polybrominated dibenz furans (PBDF) (Weber and Kuch, 2003; Zhang et al., 2016). A trace amount of PBDD and PBDF can cause severe health hazards. Toxicity profiles/relative potency of different isomers of PBDD/Fs depend on the number of Br atoms and what position it is attached. (Li et al., 2018; Fernandes and Falandysz, 2020). Basic PBDE and PBDF structures are given in Fig. 3.

Western countries tend to export e-waste to Asia and Africa for recycling. The unorganized recycling of e-waste in those developing countries causes serious health hazards to the workers and the lives around the sites. (Zhu et al., 2008b; Muenhor et al., 2010; He et al., 2012; Malliari and Kalantzi, 2017; Li et al., 2018). Metal recovery by open burning of e-waste is a common practice in many African countries (Cao et al., 2020), resulting the emission of harmful dioxins, furans, polycyclic aromatic hydrocarbon (PAHs), polyhalogenated aromatic hydrocarbon (PHAHs) and hydrogen halides into the environment. E-waste can also cause soil and ground water contamination due to lixiviation and leaching of toxins as well as air pollution due to burning. Tang et al. (2010) investigated the soil sample near Wenling, an e-waste recycling city of Taizhou province in China and reported 371.8–1231.2 µg/kg of PAHs and 52–5789.5 µg/kg of polychlorinated biphenyls, which are significantly higher than the permissible limit set by the governmental bodies and legislatures. Tue et al. (2017) reported high concentration of Cl-PAH and Br-PAH in the range 160–220 ng/g and 19–40 ng/g, respectively in the soil samples collected from the open e-waste burning area of Agbogbloshie's e-waste recycling site in Ghana. Reports of soil contamination with heavy metals at the e-waste recycling sites are extensively published in recent years(Luo et al., 2011; Wu et al., 2015; Damrongsiri et al., 2016; Zhang et al., 2019).

In this critical review, we focus on the processes that can recover

clean plastics from the e-waste and their thermochemical conversion into the alternative sources of useful mono- or oligomers or fuel. The thermochemical processes that are discussed in this review include the solvothermal process, incineration, hydrothermal process and pyrolysis. The solvothermal process is covered due to its applications in removing the non-bonded BFRs from the e-waste plastics (Zhang and Zhang, 2012a). On the other hand, incineration, pyrolysis and hydrothermal processes are known to transform the plastics into energy, chemical feedstock and fuels. A detailed assessment of different reactor configurations for the pyrolysis is carried out to give a comprehensive idea about the relevance of pyrolysis in the treatment of contaminated e-waste plastics to produce value-added products.

Previous review articles on this topic mostly focused on the impact of toxins (He et al., 2012; Wu et al., 2015; Malliari and Kalantzi, 2017) present in e-wastes on the environment as well as on the recovery of metals from e-waste (Kinoshita et al., 2003; Oh et al., 2003; Tuncuk et al., 2012; Weerachanchai et al., 2014; Rarotra et al., 2020). Despite plastic being one-fourth of total e-waste, the amount of work on e-waste plastic is still inadequate compared to the metal recovery. Ma et al. (2016a) in their review article emphasize on the recovery of fuels and chemical feed stock from the e-waste plastic containing BFRs by thermal means, such as pyrolysis and gasification. The study covers only the thermal degradation of BFR laden plastics. The influence of operating conditions and the reactor configurations needs to be investigated. Wang and Xu (2014) reviewed the recycling strategies of the non-metallic fraction of e-waste that includes plastics and glass from different types of displays. Non-destructive ways of recovering original plastics were missing in those reviews as one might find different means of separating techniques of toxins from e-waste plastics in the past. The current review focuses on the thermal and chemical routes of recovering plastics and chemical feedstock from contaminated e-waste plastics. Hence, this review article addresses the gaps in the current status quo of the industrial recycling of e-waste plastics and the latest developments in this field. This critical study will furthermore help future researchers and engineers to choose how to process and recycle the mixed stream of plastic wastes containing e-waste plastics including their possible conversion into energy, feedstock chemicals and fuels.

2. Recycling processes associated with e-waste plastics

2.1. Mechanical recycling (primary/secondary) recycling

Primary and secondary recycling deal with the mechanical means of recycling of plastic wastes, such as grinding, washing, separating, drying, re-granulating, extrusion and compounding (Al-Salem et al., 2009). Mechanical processes are utilized to produce recyclates from e-waste plastic which can be re-utilized to produce secondary products, such as dustbins, plastic containers, garden furniture and filaments for 3D printers (Gaikwad et al., 2018). Mechanical processes are basis of any e-waste recycling to dismantle and separate different parts such as PCBs, screens, batteries, wires etc. PCBs, are an essential part of any electronic equipment and represent approximately between 2 and 3% of total e-waste generated worldwide (Vats and Singh, 2014). A typical PCB is constructed with a Cu-clad laminate which consists of glass fibre reinforced with epoxy resin, plastics and metals (Hsu et al., 2019). It was approximated that a PCB contains roughly 40% metals, 30% organics and 30% refractory (fibre glass) (Shuey and Taylor, 2005). PCBs are generally treated for the extraction of the valuable metals, such as Cu, Al, Ag and Au (Oh et al., 2003). The mechanical steps that are carried out to separate the metallic fraction (MF) and the non-metallic fraction (NMF) of PCBs are illustrated in Fig. 4. The MF

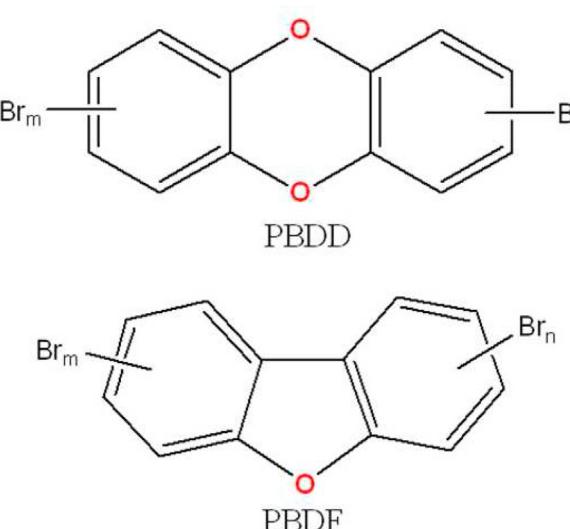


Fig. 3. Molecular structure of PBDD and PBDF.

then undergoes intense chemical treatment for the recovery of valuable chemical/metal components. Separation processes in Fig. 4 include screening (Wilson et al., 1994), shape separation (Furuuchi and Gotoh, 1992; Furuuchi et al., 1993), magnetic separation (Veit et al., 2005), electric conductive base separation (Meier-Staude and Koehnlechner, 2000), density base separation (Schubert, 1991), etc. Wäger et al. (2011) confirmed that mechanically recycled electronic and electrical waste plastics have five times lower environment impact than virgin plastics in the manufacturing of plastic products. Mechanical recycling requires a certain degree of pure plastic to produce secondary items using mechanical means. The level of purity of the e-waste plastic is compromised due to the presence of BFRs and heavy metals and makes them unsuitable for primary and secondary recycling. Past studies suggested that 25–30 wt% of the collected e-waste plastics have BFR as an additive (Brennan et al., 2002; Stenvall et al., 2013; Baldé et al., 2017). The focus should be on removing the toxins from the plastics to make them suitable for primary and secondary recycling.

2.2. Tertiary (chemical and thermal) recycling

Tertiary or chemical recovery of plastic waste is a promising aspect in the recovery of valuable chemicals and fuels (Achilias et al., 2007; Yildirir et al., 2014; Zhou et al., 2014). Its general principle, applicable to all plastics, lies behind the alteration of the polymer's chemical structure by depolymerisation. Major advantages of chemical recycling over mechanical recycling is that it allows processing of heterogeneous and contaminated plastics with limited pre-treatment opportunities, a key point for e-waste plastics' recycling. The former can convert plastics like polyethylene terephthalate (PET) and nylon into their monomer units (feedstock recycling) (Veregue et al., 2018; Choi and Choi, 2019; Jehanno et al., 2019; Vesnin et al., 2020), while the polyolefin type of plastics produce a mixture that can be a precursor to liquid fuel (Coates and Getzler, 2020). Processes, such as dissolution, hydrogenation, gasification, pyrolysis, and catalytic cracking, come under chemical recycling of plastics. More specifically concerning e-wastes, Achilias et al. (2009) carried out chemical recycling of PC, ABS, and PS from such wastes by means of dissolution/re-precipitation and pyrolysis. More than 90% recovery of polymer was reported by the dissolution process where pyrolysis converted the model PC sample into 63% liquid product. Pyrolysis or thermolysis of plastics is a process of

thermal decomposition of plastics under inert or oxygen-starved conditions. These processes are able to convert high molecular weight plastics into low molecular weight hydrocarbons and chemicals (Kaminsky et al., 1995; Adrados et al., 2012; Das and Tiwari, 2018a). The process of plastic waste into oil (fuel) through pyrolysis has been developed and practiced. Processes like Thermofuel™ (Ozmotech Pty Ltd), Smuda process, Royco process (EZ-oil generator), Reentech process, Hitachi process, Chiyoda process (Chiyoda corp. Japan), Blowdec process, Conrad processes are some of the processes available for the commercial utilization (Scheirs and Kaminsky, 2006). These processes are mostly handling polyolefin types of plastics available in MSW. Efforts could be made to produce value-added products form e-waste plastics using similar techniques. The heat recovery processes like incineration and gasification are also considered as tertiary recycling processes, however, these techniques are sometimes separately identified as quaternary recycling processes (Al-Salem et al., 2009).

Regarding PCBs' epoxy resins, they are usually used as fuel in pyrometallurgy plants that recover precious metals. Since they are made of cross-linked polymers with the aim of high chemical, time and temperature stabilities, their up-cycling is a difficult problem. In the field of depolymerisation, Chen et al. (2019) reported an original process enabling the dissolution of anhydride epoxy thanks to a transesterification reaction occurring between the –OH group in ethylene glycol, used as solvent, and the ester bond within the epoxy network. To be efficient, the reaction uses the transesterification catalyst (1,5,7-Triazabicyclo[4.4.0]dec-5-ene). Also, a diffusion solvent (N-Methyl-2-Pyrrolidinone) allows for the swelling of the resin, allowing for both the ethylene glycol and catalyst to penetrate within the cross-linked epoxy network. Reaction time varied from 6 h to 40 min when using reaction temperatures of 100 °C and 180 °C, respectively.

3. Solvothermal processes to remove BFRs from e-waste plastics

High BFR extraction efficiencies can be achieved on e-wastes' plastics with conventional extraction using organic solvents (Suzuki et al., 2002; Zhang and Zhang, 2012b). Toluene, methanol, 1-propanol, isopropanol, acetonitrile, and ethanol are some of the common organic solvents used in the process of solvent extraction (Altwaik et al., 2003; Evangelopoulos et al., 2019). Different extraction processes, such as pressurised liquid extraction (PLE),

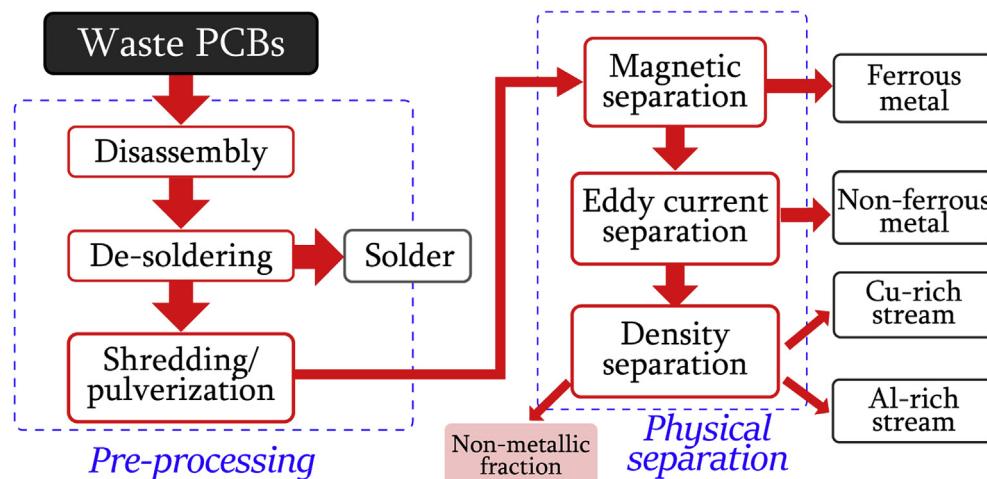


Fig. 4. Schematic representation of the physical separation process of MF and NMF of PCB.

ultrasonic assisted extraction (UAE), microwave assisted extraction (MAE) and simple Soxhlet extraction, have been proposed and investigated for BFRs extraction (Vilaplana et al., 2008).

Chandrasekaran et al. (2018) recovered 89% of pure PC from the mixture of PC/PA (from waste cell phones) by solvent extraction using N-methyl-2-pyrrolidone (NMP). NMP was preferred to dichloromethane (DCM) for selective dissolution of PC and to avoid the use of the chlorinated solvents. The study also emphasized the important of the factors, such as solid matrix, initial BFR content, solvent polarity, solid liquid ratio, temperature and pressure, affecting the BFR removal efficiency and temperature being the most dominant factor in the extraction process. Vilaplana et al. (2008) reported the use of mixed solvent systems (isopropanol/hexane and isopropanol/methanol) and achieved a high recovery of BFRs. Complete recovery of TBBPA and HBCD was reported by the process of MEA, using isopropanol/n-hexane as solvents at 130 °C for 60 min, however, UAE gives recovery rates that are strongly temperature dependant and that increase with temperature. The comparative study of UAE, MAE and PLE shows that MAE gives better BFR extraction than the other two and complete BFR extraction can be achieved using MAE and PLE. The comparison of extraction techniques is shown in Fig. 5 (Vilaplana et al., 2008; Romarís-Hortas et al., 2009). Lateef et al. (2008) carried out extraction of decabromobiphenyl ether (deca-BDE) from HIPS matrix using an ionic liquid (IL) (Papaiconomou et al., 2007; Salminen et al., 2007; Chen et al., 2012; Weerachanchai et al., 2012; Yao et al., 2017). The ILs were prepared by heating stoichiometric amounts of the liquid mixture. Two types of mixtures were selected. The first kind of IL was prepared by reacting methylimidazole with 1-bromobutane, 2-(3-chlorethoxy) ethanol and 3-bromopropionitrile. The second kind of IL was prepared using pyridine as starting material reacting with 1-bromobutane, 2-(2-chloroethoxy) ethanol, 3-bromopropionitrile, 1-bromohexane and 1-bromoocetnae. The IL was able to achieve 92.7% of BDE recovery from the HIPS. New ILs can be designed to improve the extraction efficiency of BFRs from the plastics. However, the cost of IL can be a limiting factor to apply it in recycling (Lee, 2011; Chen et al., 2013,

2017; Li et al., 2014). Zhang and Zhang (2012a) studied three alcohols (methanol, ethanol and isopropanol) as solvent for the extraction of TBBPA from e-waste plastics inside a pressurised vessel reactor, and reported that methanol was the most suitable one under optimum conditions of 90 °C, 2 h of extraction time and liquid to solid ration of 15:1, respectively. It should be noted that at high extraction temperature (-glass transition temperature, T_g) it was observed that plastics underwent marginal swelling, but this did not seem to affect its properties.

Properties of the solvent play critical roles in the solvothermal treatment in the removal of BFRs from e-waste plastics. The solvents must have swelling capacity without dissolving the plastic and at the same time must have the affinity to dissolve the BFRs. Mixed solvents with polar and non-polar character tend to increase the efficiency of the extraction if we are dealing with different types of analytes having contrasting polarity and molecular weight (Vandenburg et al., 1998; Chen et al., 2013). Temperature plays a crucial role in the extraction process. The optimum temperature for the extraction of additives from the plastics should not exceed the T_g (Lateef et al., 2008). High temperature may result excessive swelling and collapse of the polymer structure.

Supercritical carbon dioxide (Sc-CO₂) extraction processes have also been proposed, as early as year 2000 (Gamse et al., 2000), as an advanced sustainable processes for BFR removal (Altwaiq et al., 2003; Wang et al., 2004) and the decontamination and recovery of pure plastics from the contaminated e-waste plastics (Gamse et al., 2000; Nakajima et al., 2002; Ma et al., 2016a; Ben Said et al., 2017; Egerer et al., 2019). The interest in using Sc-CO₂ reside in its intermediate properties between liquid and gas, which enable its fast diffusion within the polymer and its reasonable solubilizing capacity. High extraction efficiencies have only been reported when using organic co-solvents. This significantly reduces the environmental advantage and increase running costs. More research is therefore needed to enable a pure Sc-CO₂ based extraction process allowing for decontamination and reuse of plastics.

Zhang and Zhang (2020) reported a high efficiency Sc-CO₂ based

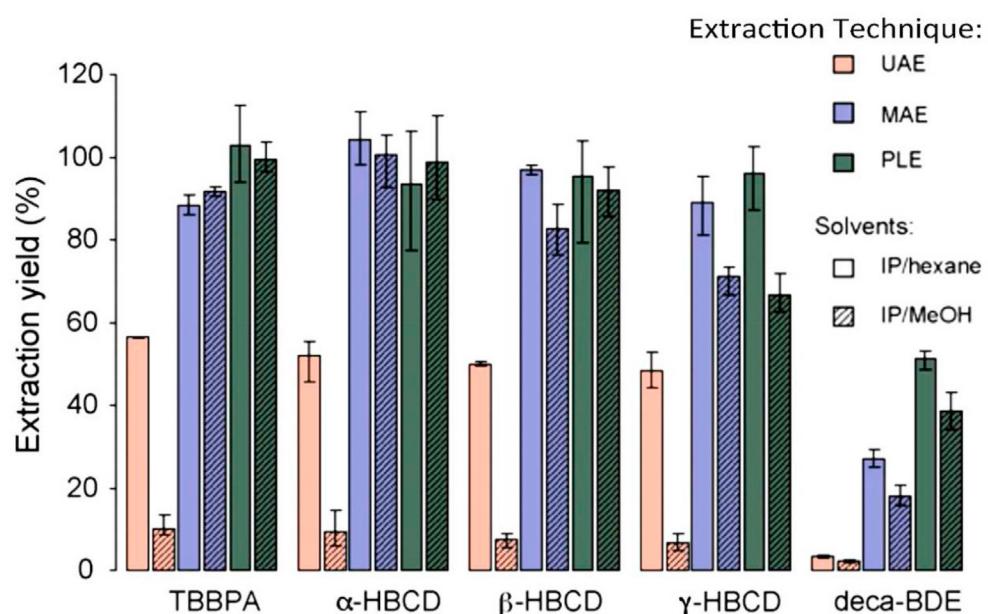


Fig. 5. Comparison of the extraction yield of UAE, MAE, and PLE for the different BFRs from standard HIPS samples [Reprinted from Journal of Chromatography A, Vol 1196–1197, Vilaplana, F., Karlsson, P., Ribes-Greus, A., Ivarsson, P., Karlsson, S., Analysis of brominated flame retardants in styrenic polymers: Comparison of the extraction efficiency of ultrasonication, microwave-assisted extraction and pressurised liquid extraction, Pages No. 139–146, Copyright (2008), with permission from.

process for dehalogenation of e-waste plastics into halogen-free products without the need of any catalysts or additives. When temperature is in the range of 375–550 °C, dehalogenation of up to 99.51% could be achieved producing halogen-free products (solid carbon materials as well as organic chemicals). This study performed at the laboratory scale will still need further investigations to prove its industrial potential and economic viability.

These results indicate that a significant amount of work is required for the development and optimization of large-scale extraction processes adapted to additive BFRs found at high concentrations in e-waste plastics. In cases of reactive BFRs, which cannot be lixiviated out, a thermochemical treatment is usually required. Br or its derivatives must be separated during the pyrolysis process from the oil/gas products, ideally into solid residues, by using suitable catalysts. The following sections will discuss the thermochemical treatment of plastic waste and role of high temperature processes, such as incineration, hydrothermal treatment and pyrolysis in the recycling of e-waste plastics.

4. Thermochemical degradation of e-waste plastics

Thermochemical approaches of recycling e-waste plastics entail processes that involve breaking of the chemical bonds, such as incineration, hydrothermal treatment (hydrolysis) and pyrolysis. Incineration involves burning the waste (oxidation) and pyrolysis is here referred as a process of degradation by heating the material under an oxygen-starved environment (Das and Tiwari, 2018b). The hydrothermal treatment uses supercritical/subcritical water or liquid solvents as possible reaction media for the plastic degradation (Watanabe et al., 1998; Weerachanchai and Lee, 2014; Zhao et al., 2018b; Wang et al., 2019).

4.1. Incineration

Incineration of solid waste are usually carried out to reduce the amount of waste to landfill as well as whenever possible to recover some of its chemical energy, to convert it into electricity (Liu and Liu, 2005). A major advantage of incineration of plastics is the solid volume reduction by 90–99% (Arvanitoyannis, 2013). Incineration can reduce volume of the waste and at the same time produce energy, however, high installation cost and toxic emission during burning limits the process into widespread utilization. Stewart and Lemieux (2003) tested the emissions from a two-stage control incineration of e-waste that consisted of computer keyboards, motherboards (without batteries) and their casings. Apart from the common flue gas components (5–10% CO₂, 50–400 ppm of CO), a significant amount of volatile and semi-volatile organic components (VOCs and SVOCs), dioxins and halogens were detected. Among the organics, bromobenzene (830–920 µg/dscm), tri-bromomethane (40–70 µg/dscm), bromomethane (20–190 µg/dscm), dibromomethane (0–20 µg/dscm) and benzene (25–35 µg/dscm) were significant. Whereas, small amounts of naphthalene (1.2–3.3 µg/dscm), acetophenone (1–2 µg/dscm), chlorobenzene (0.4–1.4 µg/dscm) and dibenzofuran (0.3–0.8 µg/dscm) were also found during the flue gas analysis. Brominated hydrocarbons were the results of brominated flame retardants in the computer components. Both Cl and Br were found in their ionised form (HCl and HBr) while, both Cl₂ and Br₂ were found in the flue gas with concentration of 0.3 and 17.8 mg/dscm, respectively. Vehlow et al. (2000) mixed e-waste plastics with household MSW to find out the toxic emission in the different exit streams from an incineration plant. Increasing the e-waste plastic fraction in the feedstock increases organic compounds related to flame retardants, such as PBDE, PBB, TBBPA, as well as active elements like Br, Cl or P. A well-controlled incineration chamber can produce heat energy

efficiently, which could produce electricity. However, the toxic emission to the environment must be restricted.

4.2. Hydrothermal treatment

Hydrothermal treatment can be used for the removal of Sb and Br (BFR) additives from e-waste plastic. It has a huge potential to treat contaminated plastics and convert them into valuable products (Zhao et al., 2018a, 2018b; Zhan et al., 2020). Hydrothermal treatment generally carried out inside a close autoclave reactor. High temperature water (sub/supercritical water) acts as reaction medium and solvent for the dissolution of small organic compounds like BFRs (Hunter et al., 2004). The native hydronium and hydroxyl ion concentration also increase by factor of 25 over a temperature range between 20 and 250 °C (Marshall and Franc, 1981). Alkaline hydrogen sulphide is found effective for hydrothermal treatment in removing decaBDE and Sb₂O₃ embedded e-waste plastics. Zhan et al. (2020) simultaneously extracted Sb and Br with the efficiency of 85.6% and 90.13%, respectively, using alkaline sulphide (Na₂S and NaOH) systems under hydrothermal condition from BFR and Sb₂O₃ embedded HIPS that were used mainly for computer casing. A temperature of 220 °C, 2 h residence time, 50 g/L Na₂S and 20 g/L NaOH were found optimum for the hydrothermal system to get maximum removal efficiency of Br and Sb. It was observed that both have significant contribution in removing Br and Sb, however, Na₂S takes a pivotal role, whereas NaOH is prominent in removing Br. First, Sodium sulphide hydrolyses in water (HS^- and OH^-) and stabilizes SbS_3^{3-} ($2SbS_3^{3-} \rightleftharpoons Sb_2S_3 + 3S^{2-}$) in the equilibrium state. On the other hand, NaOH is fully responsible for the Br removal. Hydrothermal treatment with water at 280 °C enabled the removal of 90% of Br from the HIPS–Br, however, strong mass loss of plastic limits the process (Brebu et al., 2006). On the other hand, alkaline hydrothermal treatment with KOH is able to remove Br as KBr with less degradation of the plastic. There is no effect on the leaching of antimony oxide, which was reported to be due to KOH presence. Yin et al. (2011) applied hydrothermal technology to decompose brominated epoxy resin (BER) in waste printed circuit boards (WPCBs) with sub/supercritical water. The non-metallic fraction of PCBs can be recycled using the hydrothermal treatment. In the temperature range of 200–400 °C and pressure range of 2–25 MPa, the supercritical water can decompose the BER. At low-temperature stage, the BER undergoes decomposition to form bisphenol A, bromophenol, isopropyl phenol monomer etc (Fig. 6). On the other hand, at high temperature (pyrolysis degradation) further, decomposition results phenol, o-cresol, p-cresol and other small organic compounds without bromine (Fig. 7). At a sufficient energy level, the resin molecules breakdown at weak links to produce free radicals. Free radicals then initiate further decomposition of the resin by chain initiation, growth and termination.

The hydrothermal treatment operates at lower temperatures than the pyrolysis for the decomposition of BFR laden plastics. The sub/supercritical water is capable enough to decompose the polymer matrix to produce low molecular weight hydrocarbon products. High installation cost and safety involving the handling of a high-pressure autoclave can sometimes prevent the use of hydrothermal treatments for the recycling of e-waste plastics.

4.3. Pyrolysis

Pyrolysis is a process that turns organic material into gas, oil and char through the degradation processes induced by the application of heat in the absence of oxygen (Das and Tiwari, 2018b). Pyrolysis end products can be valuable petroleum-like feedstock, which can

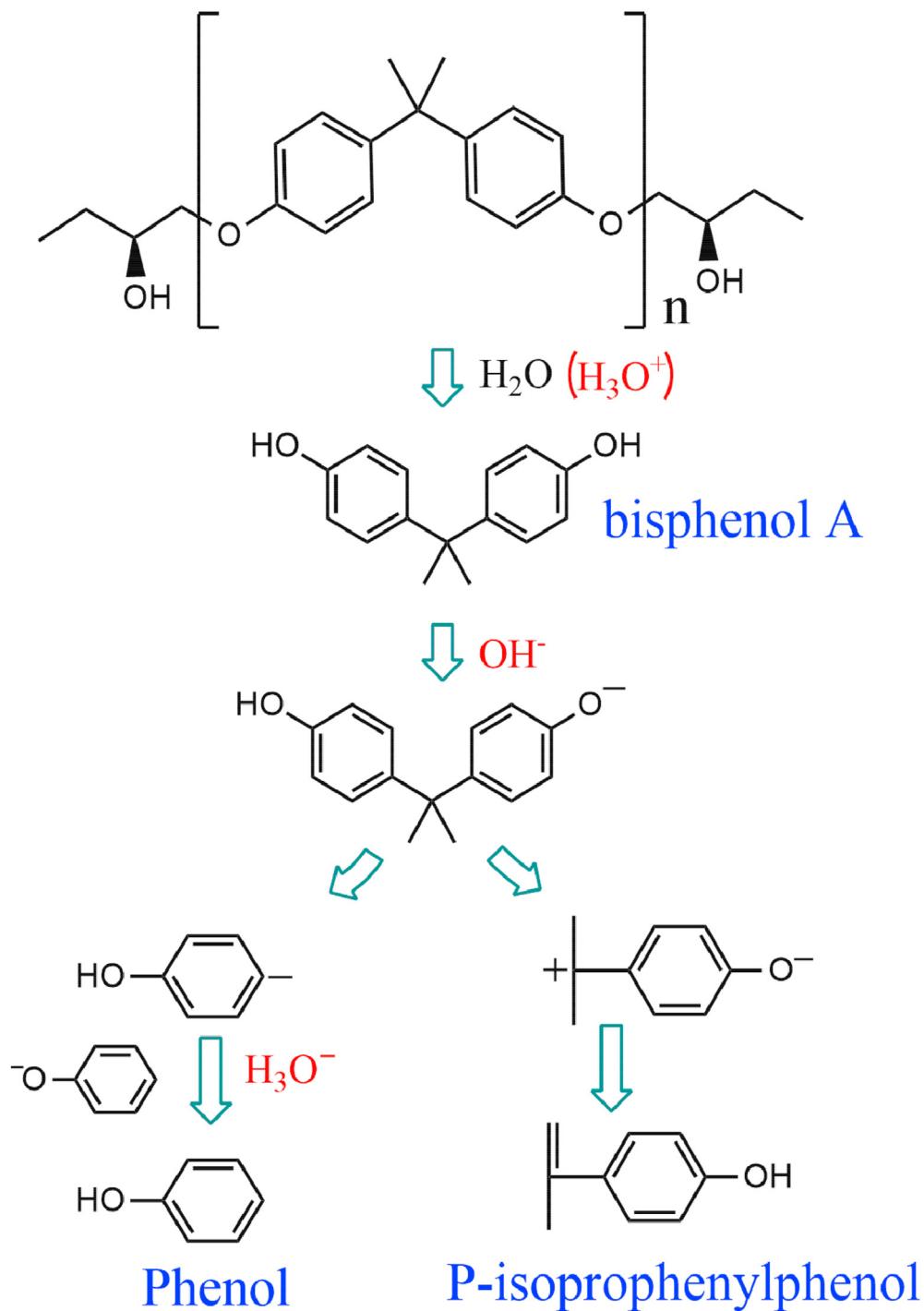


Fig. 6. Low-temperature hydrolysis degradation pathway of BER (Hunter and Savage, 2004; Yin et al., 2011).

be treated in oil refineries for the synthesis of various chemicals and fuels. Under pyrolysis, macromolecular structures of polymers present in plastic wastes are broken down into smaller molecules and produce a wide range of hydrocarbons. The pyrolysis technique has been widely investigated to recover valuable chemicals and fuels from e-waste plastics (Hall and Williams, 2006b; Antonakou et al., 2014b; Muhammad et al., 2015). Pyrolysis of e-waste plastic often produces wax (Ma et al., 2016b) and the decomposition of chlorinated or brominated plastics may end-up producing HCl or HBr (Shen et al., 2018b) at high temperature, which could lead to

the choking and corrosion of downstream pipes. However, the process of pyrolysis of e-waste plastics can compete with landfill disposal and incineration in terms of economy and sustainability. Pyrolysis treatment has been studied widely for the treatment of e-waste plastics and comes out successfully as a viable process of degradation and to fix the Br into the less valuable product streams (Ma et al., 2016b). Conventional pyrolytic degradation (under 500 °C) of Br-ABS can result in gas, liquid and residue stream. It was reported that the brominated compounds in general were found in both liquid and the residue streams (Bhaskar et al., 2003). However,

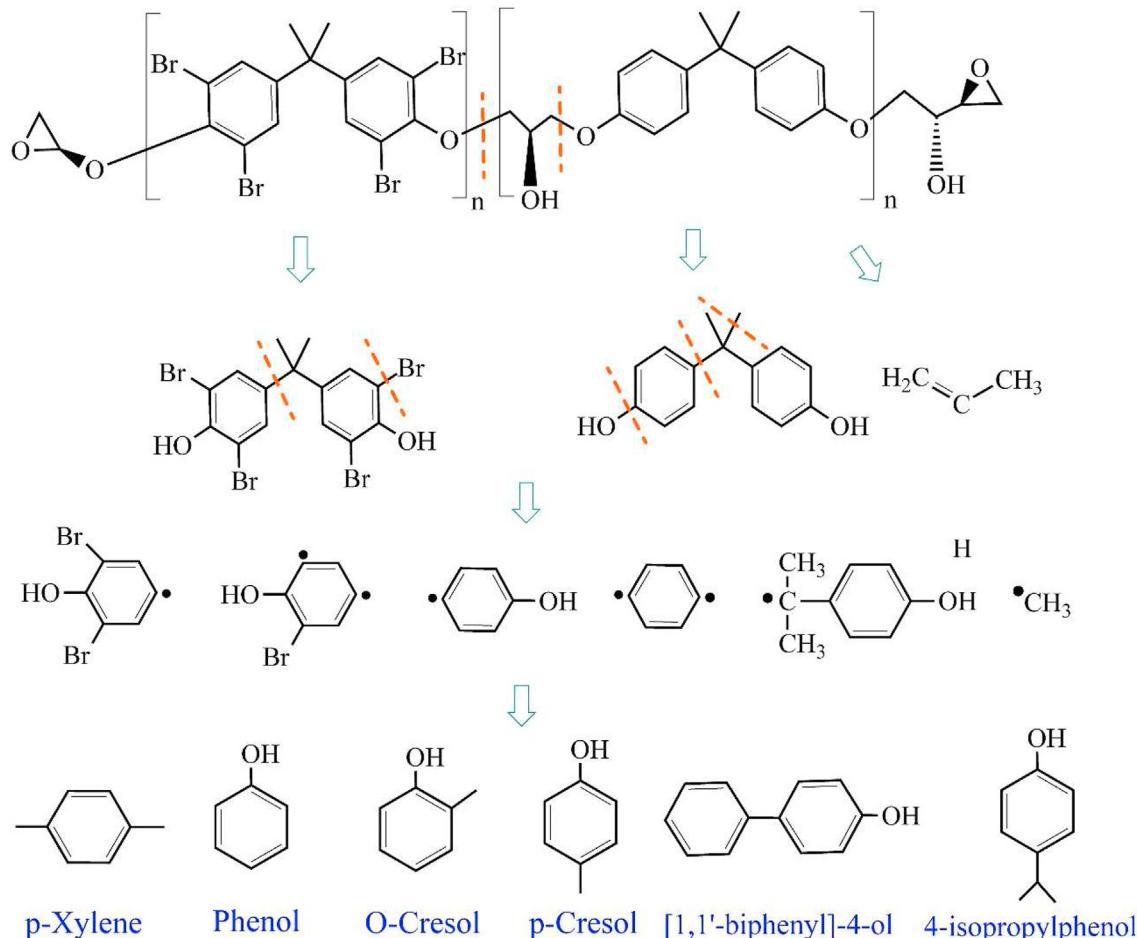


Fig. 7. High temperature pyrolysis degradation pathways of BER (Hunter and Savage, 2004; Yin et al., 2011).

at high temperature (>500 °C), the Br also released as HBr in the gaseous stream (Zhao et al., 2018c). Catalytic pyrolysis of e-waste plastics could restrict the evaluation of brominated compounds. However, the catalyst efficiency may get reduced due to the Br absorption and coke formation during pyrolysis (Hall and Williams, 2008; Ma et al., 2017).

Degradation temperatures of e-waste plastics (ABS, HIPS and PC) have been evaluated using thermogravimetric analysis (TGA) (Vouvoudi et al., 2017). TGA coupled with Fourier transform infrared spectroscopy (TG-FTIR) as well as pyrolysis coupled with GCMS (Py-GC/MS) has been used to determine real-time compositions of evolved gases during e-waste plastic degradation under an increasing temperature program (Evangelopoulos et al., 2015; Liu et al., 2016). End products of pyrolysis directly depend on the molecular structure of plastic feed and its additives, including the contaminants. Oxygenated hydrocarbons may appear in the pyrolysis product stream if the feed contains oxygen-bearing polymers, such as PET, PC, polylactic acid (PLA), PMMA (Anuar Sharuddin et al., 2016). Halogen laded plastics, such as PVC, are bound to produce inorganic hydrogen halides (HCl or HBr) due to their decomposition at high temperature (Anuar Sharuddin et al., 2016). Hence, the presence of BFR in the e-waste plastics produces halogenated organic compounds during pyrolytic degradation. Brominated organic compounds are the precursor to the toxic PBDD/Fs during utilization as fuel. It is therefore recommended to remove the BFRs from the e-waste plastics before pyrolysis but in most cases, it is not feasible due to economical or technical

limitation as well as sometimes because the Br element is covalently attached to the polymer chain, such as in brominated polystyrene.

Major gaseous and liquid components reported in the pyrolysis of e-waste plastics consisting of mostly ABS, HIPS, PC and epoxy resins from PCBs are listed in [Table 3](#) and [Table 4](#), respectively. The gaseous component consists of hydrocarbons between the range of C₁–C₄. ([Hall and Williams, 2008](#)). However, CO and CO₂ may also get produced due to the presence of oxygen in the polymer structure (PC, PMMA). [Antonakou et al. \(2014b\)](#) found out that the presence of PC in the feed significantly increases the gaseous products and the emission of CO and CO₂ gases. At high-temperature pyrolysis (>500 °C), the brominated organic components may reduce to HBr and come out as gaseous product. Light brominated organic components, such as bromomethane, can also be found in the gaseous stream.

Different organic components were identified in the chromatographic analysis of the pyrolysis liquid of e-waste plastics, such as ABS, HIPS and PP. The presence of BFR generates additional brominated components listed in Table 4. Pyrolysis of polycarbonate (PC) produces phenolic components (Antonakou et al., 2014a) and pyrolysis of ABS and PS results in aromatic hydrocarbons, like styrene, ethylbenzene (Rutkowski and Levin, 1986; Chaukura et al., 2016). At high temperature (~900 °C), aromatic components like benzene, toluene, xylene may appear in the liquid product of PC pyrolysis (Day et al., 1999).

PCBs have plastics as well as organic epoxy in addition to its

Table 3

Gaseous components from the pyrolysis of e-waste plastics.

ABS/HIPS (Hall and Williams, 2006a; Jung et al., 2012)	PC (Antonakou et al., 2014)	Non-metallic fraction of PCB (Sun et al., 2011; Guo et al., 2014)
Hydrogen	Hydrogen	Hydrogen
Methane	Carbon dioxide	Carbon dioxide
Ethane	Carbon monoxide	Carbon monoxide
Ethene	Methane	Methane
Propane	HBr ^a	Bromomethane
Propene		Bromoethane
Butane		2-methyl propene
Butene		Propene
Bromomethane ^a		Butene
Butadiene		Methylfuran
HBr ^a		Ethene
		HBr

^a For BFR laden plastics.**Table 4**

Composition of pyrolysis liquid products of e-waste plastics.

ABS (Day et al., 1999; Ma et al., 2016)	HIPS (Hall and Williams, 2006b; Ma et al., 2017)	PC (Day et al., 1999)	Non-metallic fraction of waste PCB (Vasile et al., 2008; Sun et al., 2011)
Styrene	Benzene	1-phenoxy-2-propanone	ethanol
α -Methylstyrene	Toluene	Phenol	Acetone
Iso-propylbenzene	Ethylbenzene	Benzofuran, 2-methyl	tetrahydrofuran
4-Vinyl-1-cyclohexane	Xylene	4-bromophenol	benzofuran
Benzaldehyde	Styrene	3-(1-methylethyl)phenol	Phenol, 2-methyl-
Ethylbenzene	Cumene	1-phenoxy-2-propanone	Phenol
Ethylmethylbenzene	Propylbenzene	2-(2-propenyl)phenol	Phenol, 3,5-dimethyl-
n-Propylbenzene	2-Ethyltoluene	3-(1-methylethyl)phenol	Phenol, 4-methyl-
β -Methylstyrene	Naphthalene	2-methyl-benzofuran	Phenol, 2-(1-methylethyl)-
Acrylonitrile	Diphenylmethane	1,3-bis(1-methylethyl)-benzene	Benzene, 1,10-(1,3-propanediyl)bis-
Phenylcyclohexane	Anthracene	2,3-dihydro-2-methyl-benzofuran	Naphthalene, 1,2,3,4-tetrahydro-2-phenyl-
Di-methylbenzene	1,2-Diphenylethane	1-methyl-2-phenoxy-benzene	1,2-Diphenylcyclopropane
3-phenyl-1-propene	2,2-Diphenylpropane	2,3-dihydro-2-methyl-benxofuran	1,4-Diphenyl-2-butene
Toluene	1,3-Diphenylpropane	1-naphthol,6,7-dimethyl-	2,6-Diphenyl-1,6-heptadiene
Xylene	Phenylnaphthalene	1-naphthol,6,7-dimethyl-	isobutylphenol
Cumene	Diphenylbenzene	Styrene	Benzene, 1,10-(3-methyl-1-propene-1,3-diyl)bis-
Acetonitrile	Triphenylbenzene	Diphenyl carbonate	1,4-Diphenyl-2-butene
Propanenitrile	α -Methylstyrene	Phenol, 4-(1-methyl-1-phenylethyl)-	Xylenols
2-Propanenitrile	1,2-Diphenylcyclopropane	Phenylene oxide, 4-(1-methylvinyl)-	Levoglucosanone
2-methyl-2-propenonitrile	(1-Phenyl-1-butenyl)benzene	Anthrone	Trimethylphenol
Benzenebutanenitrile	1,3-Diphenyl-1-butene	Phenol, 4,4-(1-methylethylidene) bis-	Isopropylphenol
1-3-Diphenylpropane	Isoquinoline	Benzene (at 900 °C)	Pentylphenol
1,4-diphenylbutane	Cumene	Toluene (at 900 °C)	4-methylbenzonitrile
1,3-diphenyl-1-butene	Br-compounds (Br-HIPS)^a	p-xylene (at 900 °C)	1,2,3,4-tetramethyl-5-isopropenyl phenol
1-phenylnaphthalene	1-(Bromoethyl)benzene	Br-compounds (Br-PC)^a	Br-compounds
1,3-diphenylbenzene	1,3-Diphenylpropane	Bromomethane	Bromophenol
1,2,4-triphenylbenzene	2-Bromo-4'-methylacetophenone	1-(3-bromo-4-hydroxyphenyl)-ethanone	Dibromophenol
1,3,5-triphenylbenzene		1-(3-bromo-4-hydroxyphenyl)-Et	Dibromobisphenol A
Br-compounds (for Br-ABS)^a		Benzene,2-bromo-4-methyl-1-(1-methylethyl)O-	2-(3-Bromophenyl)-2-(3,5-dibromo-4-hydroxyphenyl)propane
Bromomethane			Tribromobisphenol A
1-bromobutane			Tetrabromobisphenol A(TBBPA)
2-bromophenol			
Phenol			
2,3,5-tribromophenol			
4-bromophenol			
2,4-dibromophenol			
2,6-dibromophenol			
bromobenzene			
1-(bromoethyl)benzene			

^a For BFR laden plastics.

metallic fraction. Evangelopoulos et al. (2015) investigated the degradation behaviour of PCBs in a temperature range of 400–900 °C by TGA and Py-GC/MS. It was observed that PCBs' decomposition starts at ~120 °C and around 20–25% of volatile matters was evolved at different temperature programs. Epoxy resins present in the PCBs decomposed along with other non-

metallic components. The py-GC/MS detected a wide range of organic compounds that entailed brominated components (e.g. bromomethane, bromophenol), hydrocarbons, phenolic compounds and furans (e.g. benzofuran, 2methyl-benzofuran). Brominated molecules were produced due to the presence of significant amount of BFRs in the PCBs. PCB pyrolysis is an energy intensive

process since their thermal decomposition is endothermic. [Guo et al. \(2014\)](#) were able to recover 10.17 wt% gas, 18.23% of oil/tar and 71.60% of solid residue by doing the low temperature (~500 °C) pyrolysis of FR4 (epoxy composite) type PCBs. The estimated endothermic value of PCB pyrolysis is about 19.69 MJ/kg. The calorific values of gas and tar samples are 2.38 MJ/kg and 4.5 MJ/kg respectively, which amounted around 35% of the total energy consumption for the pyrolysis.

The pyrolysis is a viable process that thermally degrade the e-waste plastics into valuable chemicals and organic feedstock. However, the presence of BFR in the plastic matrix may reduce the feasibility of the process that urges one to look towards a simultaneous degradation and dehalogenation process with a suitable catalyst. The subsequent section covers the process thermal dehalogenation/Br fixation from e-waste plastics using catalytic pyrolysis.

4.3.1. Fixation of Br content during catalytic pyrolysis of e-waste plastics

Metal oxides can be considered as halogen absorbers or fixers. The metal oxide assisted decomposition of BFR laden e-waste plastics most likely starts with the fission of aromatic C–Br covalent bonds over metal cations. The added metal oxide reduces the decomposition temperature of BFRs (TBBPA). TBBPA and electric arc furnace dust (Fe_2O_3 , CaO and ZnO) exhibit exothermic reaction heating under pyrolytic conditions (>1000 °C) and evaporate metal halides ([Al-Harahsheh et al., 2018](#)). Addition of calcium hydroxide suppresses the formation of metal oxide ([Kumagai et al., 2017](#)). Metal oxide has the ability to remove Br from either BFRs or any brominated molecules. In the process of dehydrohalogenation, the iron and calcium-based catalysts transformed into their corresponding halides (HBr/HCl) ([Sakata et al., 2003](#)). Around 400 °C, HBr starts to appear during the thermal degradation of BFRs of which 90% can be absorbed when iron oxide is present and interacts with TBBPA pyrolysates ([Altarawneh et al., 2016](#)). Possible reactions' schemes of the debromination process of the pyrolytic components, which include bromobenzene, bromoalkane and bromophenols in the presence of Fe_2O_3 , are shown in Fig. 8 ([Altarawneh et al., 2019](#)). Both aromatic and aliphatic C–Br bonds rupture at a high energy level and Br atoms absorbed on the metal's surface. Apart from HBr scavengers, metal oxides were found promoting the formation of PBDD/Fs during the process of Br-fixation. Bromophenol, which is a common product of BFR laded e-waste plastic pyrolysis, may convert into bromophenoxy and OH-phenyl radicals which are precursors to the formation of PBDD/Fs (Fig. 8(c)) ([Altarawneh et al., 2016](#)).

Zeolites (ZSM5, Y-zeolites) are moderately effective in reducing the organobromine compounds but are less effective in removing antimony oxide from e-waste plastics flame retarded with BFRs ([Hall and Williams, 2008](#)). Zeolites also reduce the mass of oil produced during pyrolysis. [Areeprasert and Khaobang \(2018\)](#) reported that the iron oxide loaded zeolite as a catalyst for pyrolysis is cheap and a viable alternative of disposing BFR laden e-waste plastics into valuable products. [Chen et al. \(2020\)](#) use Fe–Ni/MCM-41 catalyst to recover the Br free oil fraction from the pyrolysis of BFR loaded e-waste plastics. Presence of Fe in the catalyst significantly reduces the HBr in the oil fraction whereas Ni helps in the reduction of SbBr_3 in the oil. [Ma et al. \(2016b\)](#) observed a 50% reduction of Br content in the oil product in presence of zeolite catalysts compared with the non-catalytic thermal pyrolysis. Table 5 compares the reduction of Br content in the oil fraction using different catalysts when compared with non-catalytic pyrolysis.

Fig. 9 gives a complete impression of the review article, where one can understand the flow of e-waste plastics from e-waste

stream to its different product streams going through various processes that include separation technique, thermal and hydro-thermal degradation, and incineration. The products that can be recovered by either physical/thermochemical processes are shown in Fig. 9. The non-bonded BFRs can be removed by solvothermal process, whereas thermal treatments like combustion, pyrolysis, and hydrolysis can produce value-added chemicals and fuels, at the same time can restrict the emission of toxins such as brominated organic compounds, and often transfer them into secondary by-product streams like gas or residue solids. Nevertheless, the processes have their advantages and disadvantages in terms of implementation and the decision to choose a particular process depend on the economic and environmental considerations. Fig. 10 shows the pros and cons associated with the four major processes discussed in this review article.

4.3.2. Different reactor configurations for e-waste plastic pyrolysis

To develop a full-scale pyrolysis plant, it is crucial to take into account the pros and cons of the various possible reactor configurations. Although studies on e-waste plastics are limited, pyrolysis plants used in the case of other plastics give us clues on the industrial feasibility of the former. Micro-scale pyrolysis (TGA/DSC) studies revealed the degradation characteristics of different Br rich e-waste plastics. TGA-FTIR-MS ([Yu et al., 2019](#)) and Py-GCMS studies have been conducted to confirm the degradation ranges of polymers as well as the product evolution patterns. TGA decomposition profiles of TBBPA (BFR), ABS and Br-ABS (TBBPA) are shown in Fig. 11. A two-stage decomposition profile was observed for BFR laden ABS in the derivative thermogravimetry (DTG) plot (Fig. 11(c)). The decomposition step of the first DTG peak may represent either: (i) the partial degradation of the ABS copolymer ([Czégény et al., 2012](#)) or; (ii) the decomposition of the TBBPA (11 (a)), which achieve its peak degradation temperature at 331 °C, which is close to the first DTG peak temperature (318 °C) of the Br-ABS ([Ma et al., 2019](#)). The chemical interaction between different species was observed during thermal degradation of e-waste ([Czégény et al., 2012](#)). TGA analysis confirmed the epoxy resin as the principal polymer component present in the PCB. Decomposition of PCB occurs in the temperature range between 180 and 350 °C. TG-FTIR degradation study by [Zhu et al. \(2008a\)](#) on PVC pyrolysis revealed that Ca-based additive improves the HCl removal efficiency with increasing the Ca/Cl molar ratio. TG-FTIR also suggested the combustion (O_2 environment) rate is higher than the rate of pyrolysis (He environment) of BFR laded TV shell made of ABS. TG with GCMS confirmed the release of brominated compounds including HBr with the increase of temperature (up to 600 °C) ([Yu et al., 2019](#)).

TGA based degradation studies help in settling the pyrolytic conditions for macro-scale operations. Macro-scale reactors such as batch, semi-batch, fluidized bed, screw kiln reactors have been used extensively for pyrolysis.

For lab-scale application of the batch/semi-batch type, a fixed bed pyrolysis system is effective as it is easy to control and maintain. A fixed bed reactor can be a horizontal tube reactor ([Shen et al., 2018a; Gao and Xu, 2019](#)) or vertical tube reactor with two stages ([Ma and Kamo, 2019](#)). The second stage contains the catalyst bed necessary for catalytic up-gradation of the pyrolytic products. Two-stage reactors are also utilized for the catalytic pyrolysis and Br removal of the non-metallic fractions of waste PCBs. [Ma and Kamo \(2018\)](#) applied low-cost Fe and Ni catalyst pyrolysis of waste PCBs in a two-stage fixed bed reactor and reported that Fe particles have excellent Br removal efficiencies from the pyrolysis products. A schematic representation of the two-stage fixed bed pyrolysis setup is shown in Fig. 12. It is simple in terms of operation where, in the

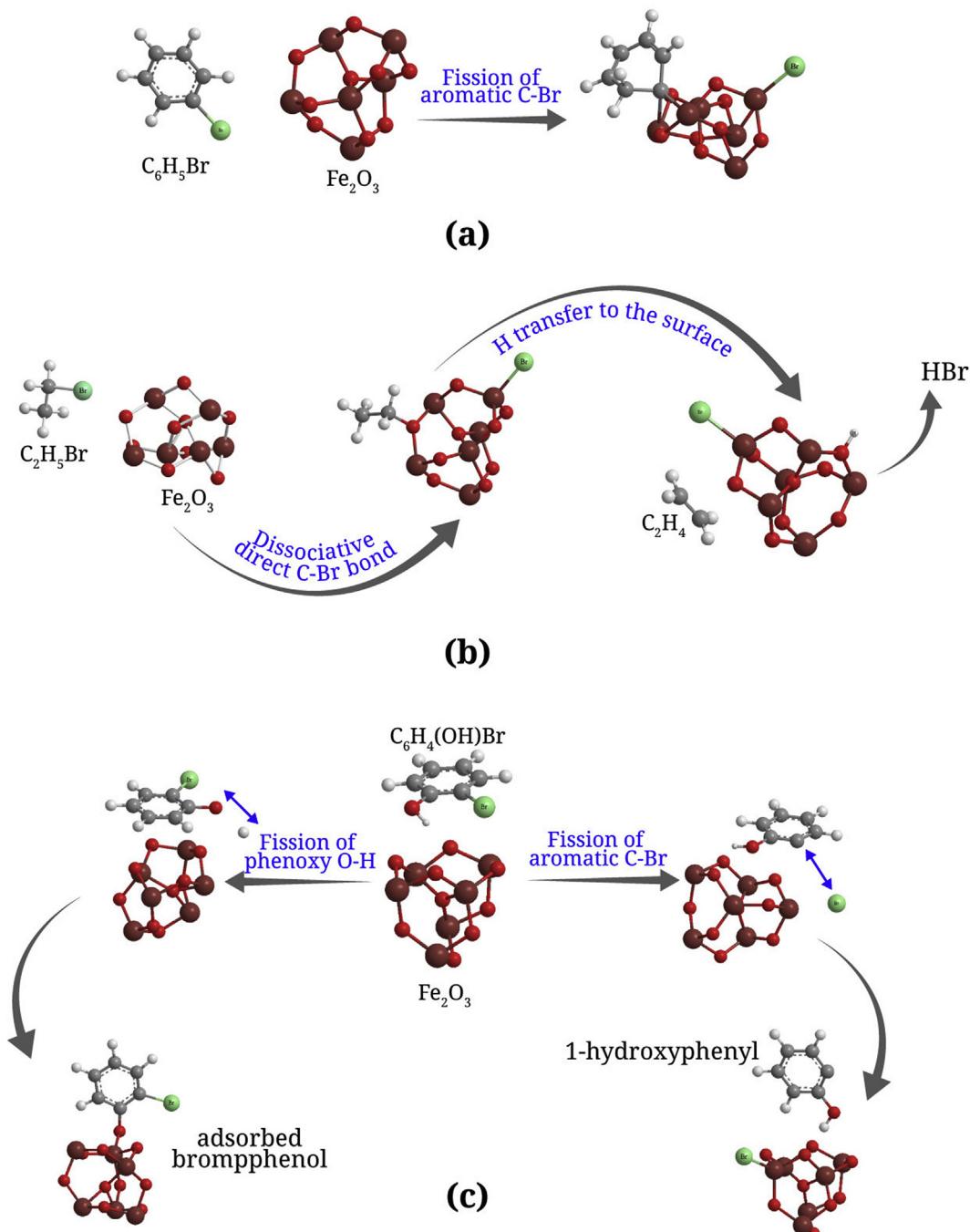


Fig. 8. Proposed mechanism of C–Br fission from the intermediate brominated compounds during pyrolysis with metal oxide (Altarawneh et al., 2016).

first stage, the plastic degrades into smaller molecules and then passes through a temperature-controlled catalyst bed which could enhance the product quality (Williams and Chishti, 2000; Ratnasari et al., 2017). Ratnasari et al. (2017) performed pyrolysis experiments in a two-stage pyrolysis unit where in the second stage, a multi-layered structured mesoporous (MCM41) catalyst stacked on top of a micro-porous catalyst (ZSM5) layer to ensure more gasoline like hydrocarbon production from waste plastics. It's a batch operation that can be upgraded to a continuous operation by using a continuous screw feeding system as in an auger reactor.

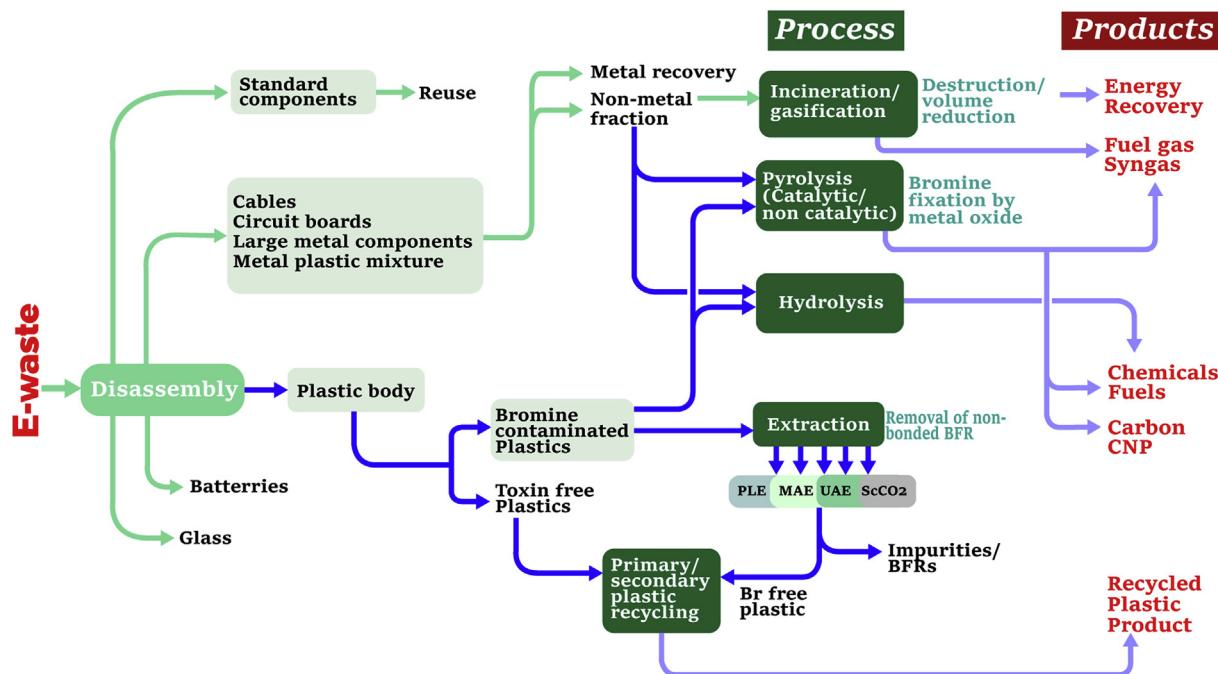
Fluidized bed reactors (FBR) are used for fast/flash pyrolysis conditions. FBRs are efficient in terms of heat and mass transfer and

also keep the process continuous for both feed and product streams. It can be employed both in the laboratory and industrial-scale operations. Most catalytic pyrolysis operations are best suited for FBR as the moving bed of the catalyst can be recycled back to the reactor after regeneration (Mauleon et al., 1990). Hall and Williams (2006a) performed pyrolysis of old computer monitor casing composed of ABS and PVC in a fast-fluidized bed pyrolysis setup at 500 °C. To assist the fluidization, the feedstock was mixed with sand particles (300–425 µm). A schematic representation of the setup is displayed in Fig. 13. FBRs were also utilized in the thermal debromination of Br-ABS by Ca-based additives, such as calcium oxide, calcium hydroxide, and oyster shells (Jung et al., 2012). The

Table 5

Effect of catalyst in the reduction of Br from the pyrolysis liquid product compare to the non-catalytic pyrolysis liquid product.

Catalyst	Material	% reduction of Br from the non-catalytic pyrolysis Reference
Y-Zeolite	Waste ABS + PC (80:20) with BFR 75% PCB with BFR 8.1%	Areeprasert and Khaobang (2018)
ZSM-5	Waste ABS + PC (80:20) with BFR 75% PCB with BFR 7.7%	
Bio-char (BC)	Waste ABS + PC (80:20) with BFR 60% PCB with BFR 54%	
Electronic waste char (EWC)	Waste ABS + PC (80:20) with BFR 81% PCB with BFR 58.6%	
Iron oxide loaded Y-Zeolite (Fe/YZ)	Waste ABS + PC (80:20) with BFR 81% PCB with BFR 19%	
Iron oxide loaded ZSM-5 (Fe/ZSM-5)	Waste ABS + PC (80:20) with BFR 89.1% PCB with BFR 46.3%	
Iron oxide loaded biochar (Fe/BC)	Waste ABS + PC (80:20) with BFR 90.5% PCB with BFR 58.6%	
Iron oxide loaded electronic waste char (Fe/EWC)	Waste ABS + PC (80:20) with BFR 98.1% PCB with BFR 70.9%	
Fe–Ni/MCM-41	Waste ABS with TBPPA + Sb ₂ O ₃ ~60%	Chen et al. (2020)
Zeolite catalyst (HY)	Br-ABS ~42%	Ma et al. (2016)
Zeolite catalyst (H β)	~50%	
Zeolite catalyst (HZSM5)	~39%	
MCM-41 (All silica)	~11%	
Al ₂ O ₃	~40%	

**Fig. 9.** Simplified flow chart of e-waste plastics from e-waste stream to product stream.

Ca-based absorbents can neutralize the hydrogen halide formed due to the pyrolysis at 400 °C of halogen-containing e-waste plastics. Process economy and the complications involved in FBRs may reduce its feasibility towards a wide-scale application.

As reported recently, low temperature pyrolysis of carbonaceous material, such as plastics, with long holding times undergoes consecutive degradation to produce lighter hydrocarbon liquid fractions (Das and Tiwari, 2018b; Singh et al., 2019). While on the contrary, at high temperature, ultra-pyrolysis (flash/fast pyrolysis) tend to produce more gaseous products (Alvarez et al., 2015; Lopez et al., 2017). Slow pyrolysis experiments were conducted mostly in batch or semi-batch reactors. For continuous production, screw kiln reactors/auger reactors can be used for low to moderate

temperature pyrolysis. Auger reactors use a screw to convey the feedstock down the length of the externally heated tube (Campuzano et al., 2019). The temperature of the reactor can be programmed to vary along the length of the reactor tube. A typical auger reactor used for the pyrolysis of biomass is shown in Fig. 14 (Campuzano et al., 2019). The similar type of reactor can also be used for plastic waste including e-waste plastics (Solar et al., 2016). These type of reactors are capable of managing the speed of the pyrolysis (fast, slow) by changing the speed of the rotor.

A two-stage reactor can be designed with both FBR (Fig. 13) and auger reactor (Fig. 14) for the pyrolysis of e-waste plastics. A schematic representation of the smme is shown in Fig. 15. A similar reactor configuration is also discussed by Park et al. (2019) in the

Contaminated e-waste plastics

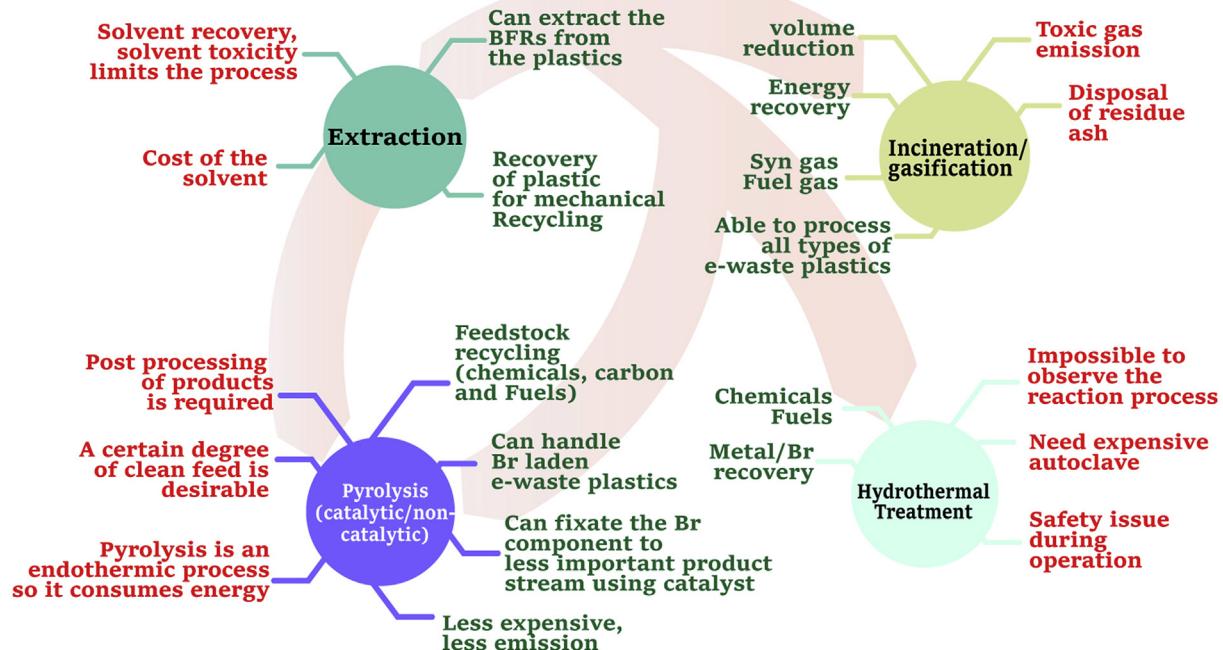


Fig. 10. The advantages (green text) and disadvantages (red text) of extraction, incineration (combustion/gasification), hydrothermal treatment and pyrolysis in the process of recycle of contaminated e-waste plastics. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

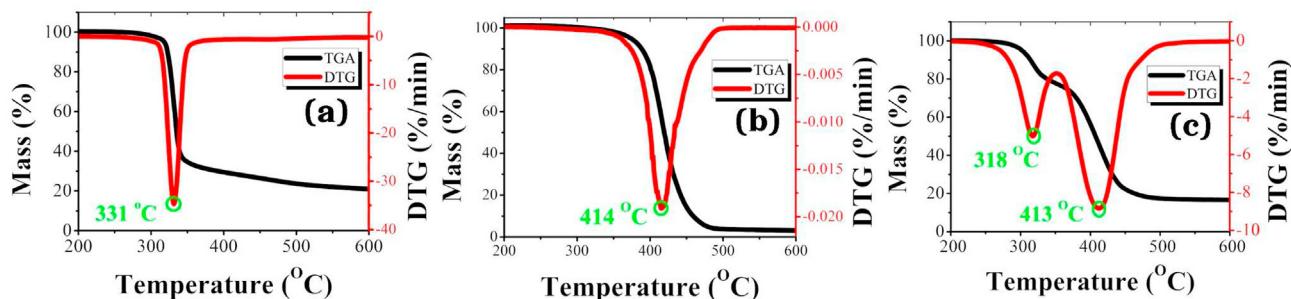


Fig. 11. TGA and DTG profiles of (a) TBBPA, (b) ABS and (c) Br-ABS (TBBPA).

application of two-stage pyrolysis of waste polyethylene. This reactor configuration would be effective in case of pyrolysis of e-waste plastics as the distribution of the heat is efficient and the material enters the fluidized bed reactor at its activated state and the exhaust gases can be reutilized as fuel gas in the heater. The post pyrolysis treatments of the evolved gases are essential so that toxins, like heavy metals, carbon particles, and inorganic acid, can be trapped before releasing into environment. The cyclone separator can separate solid particles (including catalyst) from the exit stream. The CaO filter is effective in absorbing CO, CO₂ and O₂ from the product stream of the pyrolysis. Multiple condensers ensure a complete condensation. The uncondensed gases pass through the glass wool trap and deionized water trap to restrict any particulate matter (carbon) and inorganic substances (HBr, HCl).

Pyrolysis by microwave heating of e-waste plastics gives an extra edge over conventional heating as it enables core volumetric

internal heating (Huang and Lo, 2020). Furthermore microwave heating is rapid, efficient, selective, non-contact, easy to control and safe (Jones et al., 2002). Haque (1999) pointed out that since energy is being converted from electrical to thermal energy in the microwave process, the whole process of heating is faster than a heat transfer from a hot source. One disadvantage of microwave heating is that the formation of hot spots during heating caused by localized microwave fields due to differences in dielectric properties of the various materials and the large wavelength of the radiation (Undri et al., 2013). Suriapparao et al. (2018) reported >90% selectivity of phenol concentration in the tar sample by increasing the fraction (80% Graphite 20% PCB) of graphite susceptors mixed with PCB particles during microwave-assisted pyrolysis (Suriapparao et al., 2018). The H₂ fraction in the pyrolysis gas also increases with the use of a carbonaceous susceptor. Activated carbon as a microwave absorbent can significantly improve the efficiency of microwave

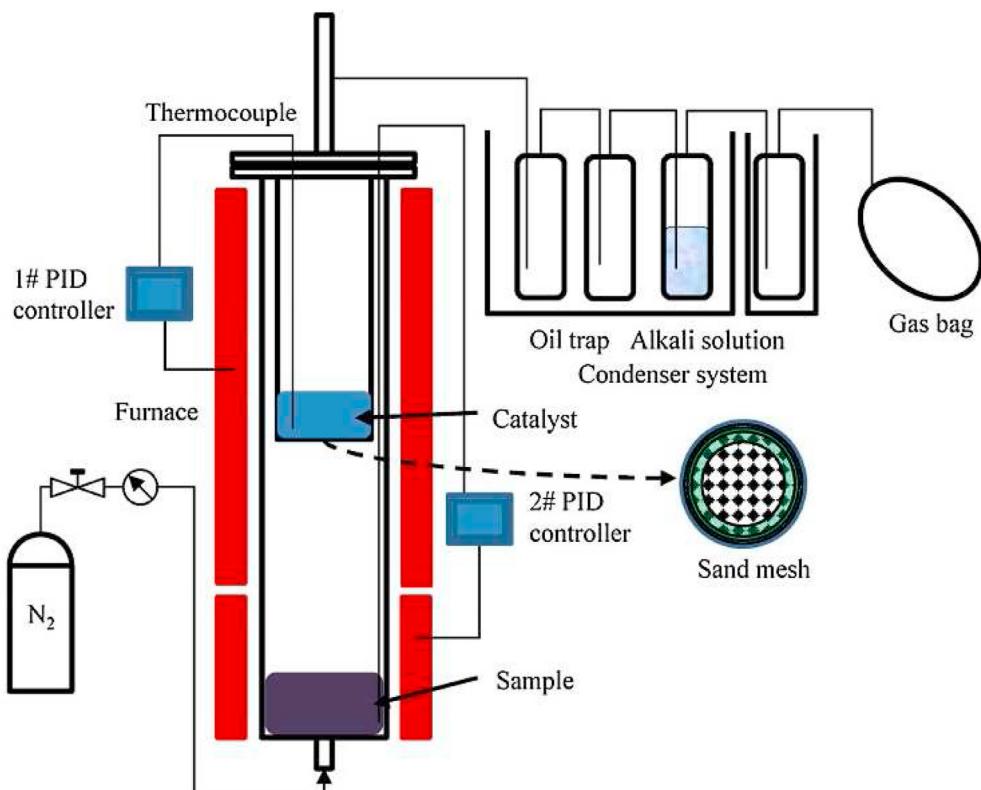


Fig. 12. Schematic representation of the two-stage fixed-bed reactor system(Ma and Kamo, 2018). [Reprinted from the article: C. Ma and T. Kamo, *Journal of Analytical and Applied Pyrolysis*, 134, (2018) 614, with permission from Elsevier].

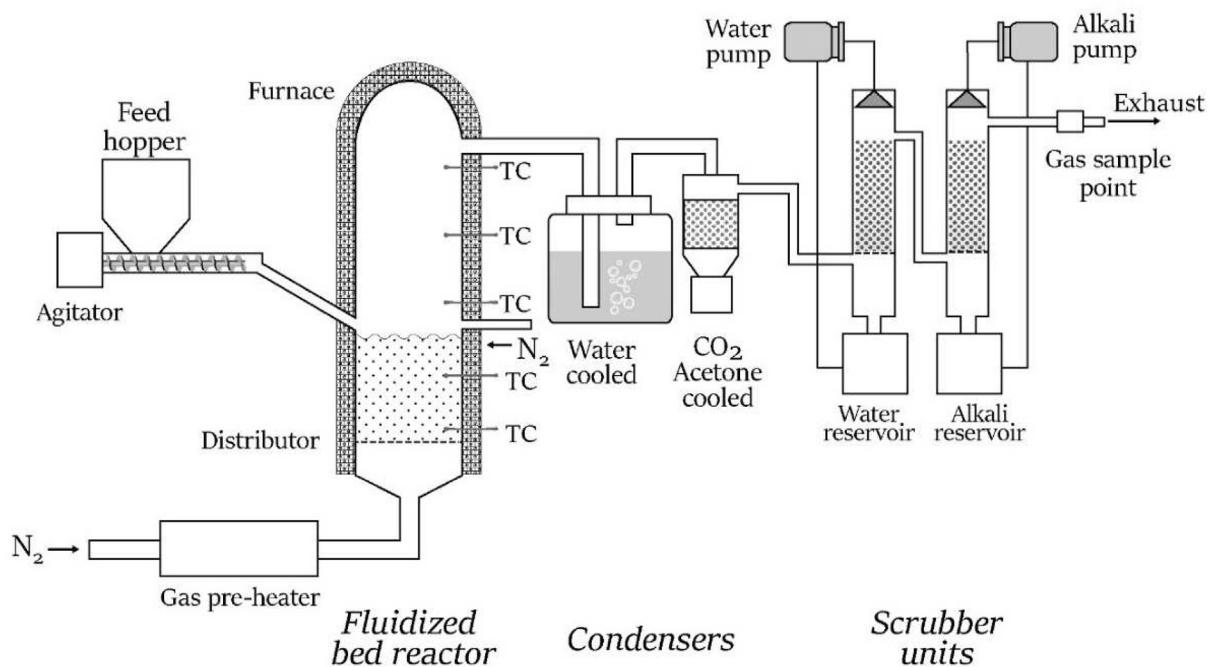


Fig. 13. Schematic representation of the fluidized bed pyrolysis reactor. (Hall and Williams, 2006).

heating, reduce the pyrolysis time, and increase the rate of degradation (Sun et al., 2011a, 2011b). A typical lab-scale microwave-assisted pyrolysis setup is shown in Fig. 16 (Sun et al., 2011a), where the outlet is connected with the condenser and gas washing

system. Although the microwave pyrolysis reactors are mostly operated in lab-scale operation, utilizing multiple microwave systems could help in scaling up.

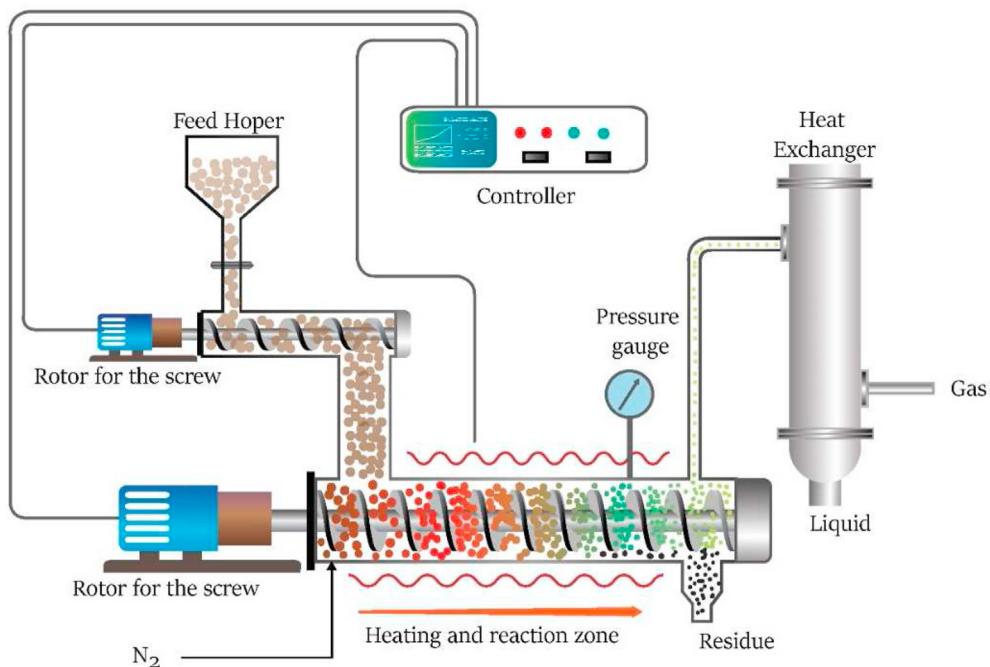


Fig. 14. Auger reactor for pyrolysis of biomass (Campuzano et al., 2019).

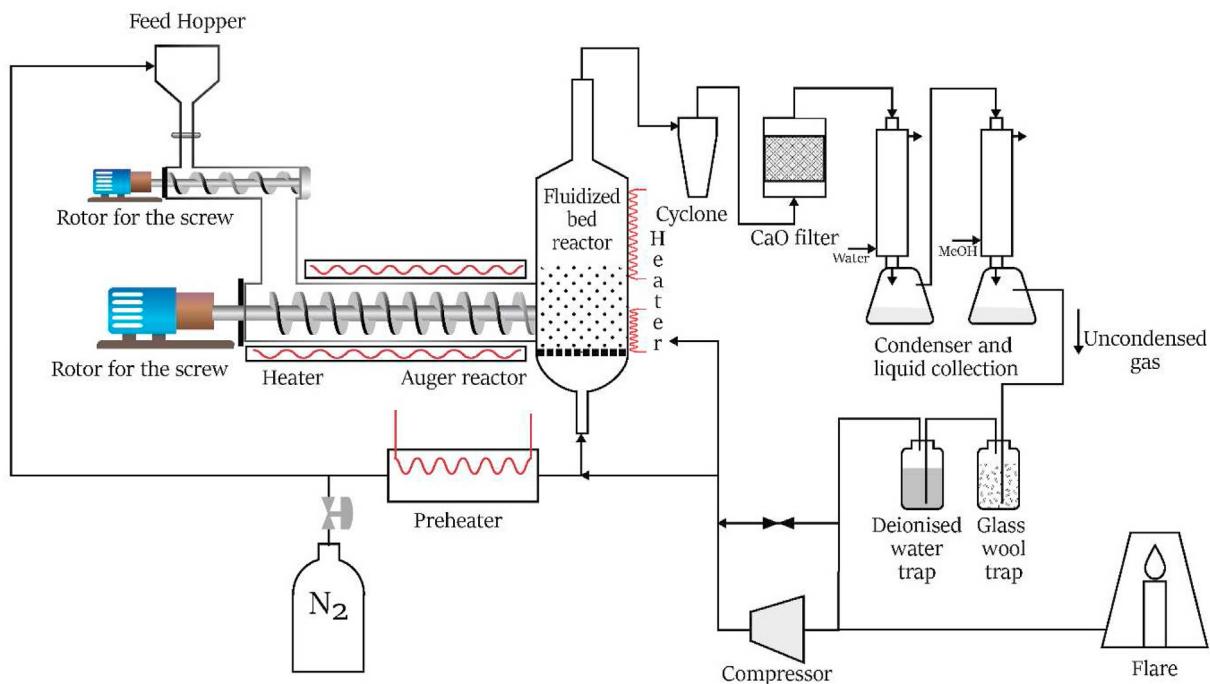


Fig. 15. The schematic representation of the two-stage continuous pyrolysis process for industrial application.

5. Conclusion and outlooks

The recovery of toxin-free plastics or chemical feedstock from the contaminated e-waste plastics is an interesting field of waste utilization and conversion. The solvothermal process is an effective way to recover BFR free plastics which can be exploited as a source recycle plastic products. Extraction processes of BFRs from the e-waste plastics have been studied extensively with different

solvents, such as methanol, propanol, hexane, etc., with good extraction efficiencies. However, it is advisable to carry out the extractions of toxins in a sustainable approach by using green technology (solutions), such as supercritical CO₂, bio solvents and ionic liquids. However, non-bonded BFRs are not susceptible to the process of extraction or leaching and require bond scission from the original polymer using thermal means. The incineration process can destroy the e-waste plastics in the process of generating

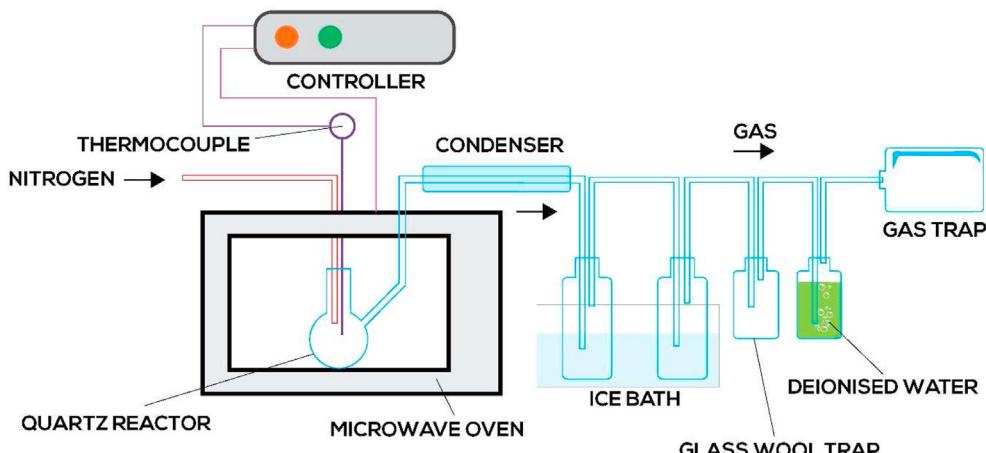


Fig. 16. Schematic representation of any laboratory microwave pyrolysis setup (Sun et al., 2011).

energy. The hydrothermal treatment has appeared to be one feasible option for removing Br from the e-waste plastics. An alkaline sulphide system can simultaneously remove Br as well as Sb from the product stream.

Pyrolysis of e-waste plastics has been performed particularly with a various catalyst based on different metals, such as Ni, Fe, Ca, Mg, etc, which might help in reducing the brominated organic hydrocarbon content in pyrolysis products. Pyrolysis is one of the most recommended technique in the treatment of e-waste plastics to convert them into value-added products, however, complete detoxification of e-waste plastics, the economic feasibility of all the techniques remains a limiting factor for the future implementation. It is pragmatic to convert e-waste plastics into clean recyclates to produce back the original product but due to the depletion of plastic over prolong use, it is less likely to produce high-quality recycled products. Furthermore, the presence of contaminants in e-waste plastics makes it difficult to recycle them directly. Proper thermochemical treatment can produce valuable recycle items that include energy, fuels, chemicals and carbon particle, while at the same time reducing the toxic emission.

6. Future scope

The review article discusses the various thermochemical routes to convert e-waste plastic into reusable toxin-free plastic as well as chemicals and fuels. The issue of non-bonded BFR in the plastic can be resolved by solvent extraction. The supercritical CO₂ extraction and hydrothermal treatment appear promising in the recovery of BFR free plastic. Hydrothermal treatment can also be suitable for the removal of antimony form the plastics especially non-metallic fraction of PCBs. Various process configuration used for pyrolysis in the feedstock recycling of plastics can be well suited for e-waste plastic recycling. Simultaneous de-halogenation and degradation reaction with Fe/Ni-base zeolite seem promising in the treatment of plastics with BFRs. The future challenges and recommendations for the subject are as follows:

- 1) It is far-fetched to remove chemically bonded Br from the polymer matrix (e.g. brominated polystyrene) by the solvent extraction. The future investigators need to explore other conceivable strategies to isolate Br from the polymer either by chemical or thermal means.
- 2) Sustainable and eco-friendly techniques, such as ScCO₂, to separate BFR have shown promising results and these techniques should be further explored more to improve its

feasibility. Bio-solvent, ionic liquids can replace conventional solvents and regeneration of solvent can significantly improve the applicability of the BFR extraction process.

- 3) Novel pyrolysis techniques, like hydrothermal pyrolysis, low-temperature slow pyrolysis, and efficient catalytic pyrolysis, ought to investigate more in-depth to recover value-added toxin-free fuels and chemicals from e-waste plastics in a more sustainable and economical way.
- 4) Apart from BFR, the e-waste plastics often found contaminated with the exceeding permissible level of heavy metals (Pb, Hg, Co, Ba, etc.). These sorts of plastics need special attention and require further investigation to develop sustainable strategies of decontamination. Efforts should be made in eliminating the toxin emissions (POPs, dioxins, PAHs) during the thermal treatment of e-waste.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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