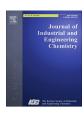


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Review

Applications, treatments, and reuse of plastics from electrical and electronic equipment



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ABSTRACT

Waste of electrical and electronic equipment (WEEE, also known as E-Waste) has emerged as a serious issue for the whole world along with the evolution of modern industry. WEEE plastic contains heavy metals (arsenic, mercury, chromium, cadmium, lead, etc.) and halogen materials (bromine, chlorine, etc.), which are toxic and harmful to the environment, therefore the recycling of WEEE plastic is necessary and critical. Not only the various additives in the polymer but also the contaminations from polychlorinated biphenyl (PCB), batteries, etc, make the recycling process challenging. In this review, the functional application of plastic in electrical and electronic equipment (EEE) including electrical insulation, heat insulation, etc, is firstly introduced. The various components and additives of plastic in EEE are provided. Moreover, the state-of-the-art treatment and recycle methods of the WEEE plastic are summarized and discussed. This review can provide a comprehensive reference for investigation of plastic in e-waste. © 2022 The Korean Society of Industrial and Engineering Chemistry. Published by Elsevier B.V. All rights reserved.

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Introduction

The rapid development of the information technology and electronic industry around the world has facilitated our daily life in the various areas, such as information exchanging, recreation, medical care, education, as well as transportation. The production and usage of electrical and electronic equipment (EEE) are rising intensively in recent decades, while leaving huge amount of WEEE to our living environment. In 2019, the total weight of WEEE yielded from the world was 53.6 million tons (Mt), and it keeps increasing [1–4]. It has been reported that at the end of 2030, the post-consumer e-waste generated all over the word exceeding 74.7 Mt with a per capita generation of 9 kg [4]. Singapore contributes about 60,000 tons of e-waste annually [1,5–11].

WEEE contains large and small household appliances, cooling and freezing, cathode-ray tube television (CRT-TV) and monitors, and others including electrical and electronic equipment for lighting, instrument, toys, entertainment and sports, healthcare, indicating and controlling equipment, automatic dispenser, and so on [11].

In the WEEE, metal and refractory oxides occupy about 40-60 wt.% and 20-40 wt.%, while the plastic accounts for about 15-30 wt.%. Polyethylene, polypropylene, polystyrene, polyesters, and polycarbonates are the typical plastic components used in electrical and electronic equipment [12]. Plastic as a multifunctional material is widely used and in increasing demand within electrical and electronic devices and it occupies a weight content of 20-30% in all the WEEE [5,13-16]. The plastics make the electrical products safer, lightweight, more advanced, and attractive. In fact, the portion of plastic used in the electrical and electronic equipment keeps growing [6]. The functional applications of plastic in electrical and electronic equipment include: (a) electrical insulation, (b) heat insulation, (c) lightweight, (d) freedom of design, (e) durability, (f) energy-efficient, and (g) recyclable. Therefore, plastic makes a crucial contribution to the development of electrical and electronic products [17-20].

It is a complicated process to treat the WEEE because of the increasing amount of e-waste and the various components. European Directive reports 6 categories of WEEE that 75% to 80% must be recovered and 55% to 80% must be recycled depending on the category [European Directive 2012/19/EU]. The hazardous substances include heavy and toxic metals including Hg, Cd, Pb, etc., and flame retardants, such as tetrabromobisphenol-A chemicals, polybrominated diphenyl compounds, etc. In addition, there are some other substances in e-waste, which are potentially harmful but not well-studied [13,21–23]. If these wastes are not managed

properly, the harmful components may cause serious health risks for human and environment. Therefore, various technologies have been investigated to treatment of plastic waste [24–28]. However, till now, the recycle rate of e-waste is still low. The modern society needs sustainable e-waste management, processing, and recycling methods. Many factors, such as import and export trade, law and regulations of different governments, strategies and technologies of hazards and waste post management, as well as procedures for recycle, can affect the e-waste in the worldwide [29–31]. The strategies to treat the e-waste include: [32–34] reducing the disposal, using incineration and landfill, reusing the end of life electrical and electronic devices, recycling, and recovering. In this review, the applications of plastic in WEEE, and treatment and recycling of plastic in WEEE are summarized.

Plastics in electrical and electronic products

There are many advantages of plastic in electrical and electronic equipment. Frist, plastic usually possesses superior electrical and heat insulation, light weight, and freedom of shape and design compared with other materials, such as metals and ceramics. For example, polyvinyl chloride (PVC) is generally used for electric wiring insulation. In addition, plastic has high stability and durability. It is hygienic (as a result easily to be cared and cleaned) and also offers good resistance to oil and even acids. Plastics consume only $\sim 3\text{-}4\%$ of oil in the world and are cost effective to be produced compared with other materials, such as metal or ceramics. At last, most plastic components can be recycled and reused after processing, to save energy and raw materials, when the EEE reaches its end of life.

General guidelines for the use of plastics/polymers in EEE

Electronic circuit boards also known as Printed circuit boards (PCBs) are important internal components of EEE and its accumulation after dismantling of WEEE is huge (3–6 %). The PCBs are mainly composed of epoxy resin and fiberglass. In general, metals, ceramics (mainly SiO₂ and Al₂O₃), and plastics commonly occupy about 40%, 30% and 30% of the total PCBs, respectively [35]. The common plastics in PCBs include polyethylene, polypropylene, polyesters, epoxides, poly(vinylchloride), poly(tetrafluroethane), and Nylon. Electrical insulation, heat stability, and good mechanical strength are the basic guidelines for plastics used in PCBs.

Considering the high versatility of plastics and polymers, the general guideline of use of plastics and polymers in electronic circuit boards and other electrical and electronic equipment is to

Table 1The typical compositions, features, and applications of various polymers in plastic.

Types	Composition	Features	Applications	Reference
ABS	Combination of different monomers: acrylonitrile, butadiene, and styrene	The styrene offers plastic surface shininess and impervious while the rubbery substance (polybutadiene) maintains toughness.	Telephone handsets, keyboards, monitors, and computer housings.	[43]
PS	Polystyrene	High impact, tough, cheap, and easy to thermoform	Computers, computer monitors, printers, computer housing, etc. The application area of PS includes refrigerator trays and TV cabinets	[44,45]
PC	Polymerized through condensation polymerization from carbonate functional groups-containing thermoplastic polymers.	It has excellent strength, toughness, and optical properties.	Bulletproof windows, lenses for eyeglasses, and electronic devices, telecommunications products, as well as high-stability capacitors.	[44,46]
PVC	Amorphous polymer and is made from vinyl chloride.	Thermally unstable, can degrade and release hazardous hydrogen chloride gas after long term usage, low flammability warning.	Widely used to produce cable and wire insulation as well as cable trucking, etc.	[44]
PE	Formed through polymerization of the ethylene monomer.	Wide range of properties depends on its molecular weight.	Wide range such as film, coating and laminating, sheet extrusion, conduit & pipe	[44,47,48]
PP	Polypropylene	Has a lower percentage of crystallinity than PE but displays better strength and stiffness	Applied in ropes, twine, tape, carpets, upholstery, clothing and camping equipment.	
Other polymers	Such as aikyd resins, polyacetal, polyphenylene suiphide, polymethyl methacrylate, polyester	Various features	Such as circuit breakers, business machine parts, hairdryer grilles, element bases, transformers, coffee machines, toasters, printers, monitors, and televisions etc.	[47–52]

enable the design and manufacture of various polymer products to meet very different application requirements at acceptable cost. Polymers can also be produced in blends or matrix composites, and they can further incorporate with many other types of performance-enhancing additives or reinforcing agents including fibers as well as platy and particulate fillers to further enhance its versatility. According to the material properties of plastics and polymers, they generally can be sorted to three broad areas in EEE:

Electrically insulating and thermal insulation plastic or polymers [36]

The general guideline of polymer for a specific electrically insulating application depends on its detailed requirement. For instance, rigid polymers, such as Acrylonitrile-butadiene-styrene (ABS) copolymers, polycarbonate, polyoxymethylene (POM), and their blends, are selected as covers, enclosures, and housings as well as electrical switches, distribution boards, and power sets. This is because they can function as both mechanical protection and electrical insulation for EEE. These tensile materials also process good impact resistance and high dimensional stability. In contrast, flexible plastics with low elastic modulus, such as PVC and Polyvinylidene fluoride (PVDF), are preferred as electrical insulation materials for wires and cables. Polytetrafluoroethylene (PTFE) is applied in hook-up wiring and coaxial cables in high standard environment, such as aerospace and computer, while polyethylene (PE) is usually preferred in low-cost but similar applications. Polystyrene (PS) is widely used as foam packaging material for shipment of EEE. Moreover, polymers inherently have a low thermal conductivity so they have good thermal insulation, which can provide cool surfaces and enhances the safety.

Intrinsically conducting plastic or polymer [37]

Some special polymers may exhibit similar electrical conductivity with metallic conductors and inorganic semiconductors. The typical intrinsically conducting polymers include polyacetylenes with potential use in organic semiconductors, doped poly(p-phenylene vinylene, PPV), polypyrroles (PPy), polyanilines (PANI), and polythiophenes (PTs). The delocalized electrons in aromatic rings and/or double bonds are the key for its high conductivity. In some special cases, some plastics with poor conductivity can be doped or modified to enhance its electrical conductivity.

Polymers act as adhesives, coatings, potting materials and sealants [38]

Adhesives are widely used in PCBs and most of them are ester type polymers, such as epoxy, acrylate, epoxy acrylate, and urethane acrylate. The board of a typical PCB is made of a multilaminate glass fiber reinforced epoxy thermoset matrix polymer. Polyimide as a special adhesive can tolerate up to 300 °C [35]. Acrylate, epoxy, urethane, and amorphous fluoropolymers are widely used as conformal coatings, which can protect the components of PCB from environmental hazards. Epoxies, silicone, and urethane are the three typical potting materials or sealants to fill an EEE which can make EEE resistant to shock or vibration or isolate the external environment. In all, the general guidelines for the use of plastics and polymers in electronic circuit boards and other electrical and electronic equipment are saving cost based on meeting needs.

Plastic composition in EEE

There are various components in plastic materials for EEE, which enable high overall performances. Acrylonitrile butadiene styrene (ABS), polycarbonate (PC), high impact polystyrene (HIPS), and polypropylene (PP) are the majority of WEEE plastics. They are applied mostly in different size household instruments and cooling devices [39]. According to the plastic composition report [40,41], ABS occupies 30%, and HIPS 25%. The others include PC, PC/ABS blends, and PP accounting for 10%, 9% and 8% respectively. The typical compositions, features, and applications of these plastics are briefly summarized in Table 1.

Physical and chemical properties of the plastics used in EEE

The physical and chemical properties of various plastics and polymers used within the electrical and electronic products EEE are summarized in Table 2. Apart from the polymers different plastics additives are also added to the product to improve both physical and chemical properties of the final product. Some of the additives were addressed in Table 2. Antioxidants, blowing agents, clarifying and nucleating agent, fillers (talc, nanofillers), flame retardants, smoke suppressant, heat stabilizers, impact modifiers,

Table 2Chemical and physical properties of polymers in plastic.

Types	Chemical properties	Physical properties	Reference
ABS	 Corrosion-resistant to most acids, alkalis, and salts, but can be dissolved in organic solvents. Can mix with other polymers (PC, PVC, PA, PBT). 	 white solid, good formability, smooth surface, easy to dye and electroplating Density: 1.04 ~ 1.06 g/cm³, Glass transition tempera- 	[53]
		ture = 100–110 °C	
		• Application temperature range $-25~^{\circ}\text{C}\sim60~^{\circ}\text{C}$.	
PS	• Good chemical inertness to most acids and bases, Insoluble in water but	• Solid or foamed state, density: 0.96–1.05 g/cm ³	[44,45,54]
	easily dissolved by common organic solventsDepolymerization can be achieved by pyrolysis (>430 °C)	• Melting point: $\sim 240~^{\circ}\text{C}$, maximum service temperature 90 $^{\circ}\text{C}$. • Glass transition temperature above 100 $^{\circ}\text{C}$	
PC	Amorphous transparent thermoplastic.	• Density: 1.20–1.22 g/cm ³ .	[44,46]
	 Hydrolyzes slowly above 70 °C under high humidity. 	• High heat resistance ($-40 \sim 140$ °C).	
		 High impact-resistance but low scratch-resistance. 	
		Allows high light transmission.	
PVC	 Chemically resistance to common acids, salts, bases, fats, alcohols, and a few organic solvents (Tetrahydrofuran and acetone can dissolve PVC) Thermoplastic polymer 	 White, brittle solid with density of 1.38 g/cm³, stable < 80 °C. Good hardness and mechanical properties. Flexible if mixed with plasticizers. 	[55]
		 Good insulation properties 	
		• UV resistant	
		Low corona resistance	
PE	Molecules linked linearly, branched, or crosslinked change the properties of the polymers.	 Density: 0.91–0.965 g/cm³, Melting point: 115–135 °C 	[44,47,48]
	of the polymers. Nonpolar, excellent resistance to most solvents	 Merting point. 113–133 °C Low strength, hardness, friction, and rigidity, high ductility, 	
	 Very good stability to acids, bases, and organic solvents. 	and impact strength.	
	Negligible water absorption	Good electrical insulator.	
PP	Chemically resistant to non-oxidizing acids, bases, and organic solvents	• Density: $0.85 \sim 0.95 \text{ g/cm}^3$, melting point: 160 °C ,	[56]
	Stabile to strong oxidants.	Translucent, semirigid and flexible.	
	 Dissolvable in nonpolar solvents such as xylene, tetralin and decalin. PP is chemically less resistant than PE. 	Fatigue and heat resistance	

Table 3 Functions, contents, and typical components of the additives in EEE plastic.

Additives	Functions	Content	Typical components	Reference
Antioxidants	To prevent or slow down the degradation process from oixdation	~1 %	BPA (Bisphenol A), BHA (butylated hydroxyanisole), BHT (2,6-Di-tert-butyl-4-methylpheno), Ionox 100 (6-ditert-butyl-4-(hydroxymethyl)phenol), Irganox 1010 (pentaerythritol (tetrakis)-3,5-di-tert-butyl-4-hydroxycinnamate), and Irganox 1076 (octadecyl 3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate)	[74]
Heat and light stabilizers	To prevent degradation form heat and light to extend the life of plastic	Up to $\sim 5\%$	Typical lead compounds such as lead chloride that is not easy to dissolve. New organic stabilizers without heavy metal such as calcium-zinc.	[72,75,76]
Plasticizers	A softening agent to decrease the plasticity or reduce material's viscosity	Up to 90 % (e.g. in PVC)	Phthalates occupy 92% of plasticizers produced worldwide. In addition, DEHP (bis(2-ethylhexyl)phthalate) represents 51% of the phthalates which is widely applied in films and cables.	[72,77,78]
Colorants	Coloring, dyes is usually brighter and sharper than that from pigments.	~ 1 %	Both dyes (soluble colored organic compounds) and pigments (insoluble solid organic compounds) can be used as colorants, dyes bond strongly to the polymer molecules and make up the textile fiber while pigments are applied in areas such as ceramics and plastics.	[74]

plasticizers, UV and visible light stabilizers, etc. are some of the common types of plastic additives also found in EEE. The usage of the additives has been regulated by the governing bodies, such as REACH (Registration, Evaluation and Authorization of Chemicals) controlled by EU. EU also regulates the usage of Phenols and Phthalates as plastic additives because some of these compounds found carcinogenic [42].

Additives in plastic

The electrical and electronic industry has grown quickly in size in the last decades, with increasing demand for mobile phones, computers, digital organizers, and business-based office equipment. To fulfill the requirements of new products which are lighter, more durable, stronger resistance to fire, heat, sunlight, and chemicals, various organic or inorganic additives, such as flame retardant, stabilizers of light and heat, colorants, fillers, pigments, and plasticizers as shown in Table 3, are also added into virgin polymers before their conversion into plastic products. These additives contain heavy or toxic metals (Sb, Zn, Ti, Ca, Ba, Pb, Cd, Cr, Cu, Co, Sr, Ni, etc.) and halogenated substances, which typically

come out in the pyrolysis residue of plastics, thus affecting their further utilization [44,57-61]. In this part, different types of additives will be introduced and summarized. Among them, flame retardants (FR) are important additives for plastics that have more than 200 different types. The flame retardants can be added as high as 15% in plastic [22,62]. Flame retardants can avoid or stop the fire process by (1) disrupt the combustion stage, (2) physical insulation of the fuel sources from material parts, (3) emitting water or inter gases, such as N2 to dilute flammable gases or materials from oxygen. Major halogenated flame retardants are consumed in electrical industry and the flame retardants market is also growing rapidly in Asia region [63]. Among the various flame retardants, about 80 different types of brominated flame retardants (BFRs) are widely used in EEE [64]. Chlorinated FRs are much less used than brominated ones, but they can be combined with magnesium hydroxide in polyolefin wire and cable [65]. At present, flame retardants, such as phosphorus or nitrogen containing ones without halogen or inorganic minerals, are more attractive and intensively studied due to their low toxicity and flexible regulatory issues than halogen ones. However, phosphorus flame retardant is generally more expensive and has some problems in processing

and performance. Even though phosphorus FRs have been offered by several FR manufacturers, which now occupy 25% of the global FR market [5,66,67]. Another widely applied, less toxic, and much cheaper flame retardant (FR) is aluminum trihydrate or it derived inorganic hydrates. However, it can be only used for polymers at low temperatures because aluminum trihydrate is unstable above 190 °C [63,68,69]. For high temperature thermoplastic applications, inorganic hydrate magnesium hydroxide (Mg(OH)2) can be used because it is more thermally stable than aluminum trihydrate. In addition, due to the white or light color of these inorganic materials and the low toxicity and cheap prices, thus they can be used directly or mixed with other RFs for various EEE [70]. However, high concentrations of Mg(OH)2 need to be applied to achieve good FR performance (similar with aluminum trihydrate), which may enhance density and brittleness of materials.

There are some other additives, such as mold release agents. foaming agent, mineral filling, coupling agents, which are added in the plastic production processes. Foaming agent, such as surfactant or a blowing agent, can facilitate the generation of foams. For example, triazine polymer is used in intumescent flame retardants, designated as charring-foaming agent (CFA) to improve thermal stabilities [71]. In addition, minerals are generally filled in plastics and applied to the wire and cable [72]. Fillers can occupy about 40% of plastic and calcium carbonate is most widely used as a filler [73]. Coupling agents typically are silane-based materials which are used to combine different materials together by chemical bond in demanding conditions. Toughness enhancers as a plastic reinforcing agent can account for 10% of plastic. They contain two different materials for chemical and/or physical effects. In all, these additives are conducive to increasing the mechanical strength, apparent quality, and processing performance of the material products.

Management and treatment of the e-waste plastic from WEEE

Considering the hazardous materials contained in the WEEE, both government and public need to solve the problems. WEEE is composed of various materials (such as metals, plastic, and refractory oxides) and components with different properties. The problems become more serious when some of the components in the waste are highly toxic. Therefore, safe handling precures and recycling methods are needed to protect environments and human from contamination and detrimental effects [41,79]. Before the treatment of plastic in WEEE, it first need to be separated or dismounted from the WEEE. The recycling of WEEE plastic can provide valuable materials and polymers [80]. As discussed in section 2.2, plastics materials are composed of various metals and halogen-rich additives [81]. Moreover, the toxic components in the plastics not only come from the additives but also from the contaminations, which are generated during use and/or by storing together with waste printed circuit boards or batteries. The additives have two effects: they can benefit the properties of products but bring problems together to the environment and human [17]. Currently, the main strategies for the treatment of e-waste can be considered by following four directions, including direct landfill of waste without process, energy recovery by incineration method, mechanical recycling by adding virgin monomer or polymer, and chemical recycling to produce valuable products [5,80,82-84]. These methods and processes will be summarized and discussed in detail in the following sections.

Landfill

Landfill is a direct procedure with low direct cost, however, landfill of electronic wastes can stay for decades, centuries or even

millennia, which increases the toxic components in the environment and leads to land sacrifice and pollution, as well as surface and groundwater contamination as a result. When electronic wastes come in contact with water in the ground, leaching occurs and releases potentially hazardous substances [85,86]. The problem of landfilling is more common in developing countries and the constructions of controlled landfilling are not adequate. As reported, in developing countries, open dumps account for 12.5%, 48.8%, and 32.4%, respectively for low-income, lower-middleincome, and middle-income sections. In addition, landfills occupy 58.51%, 11%, and 59%, respectively for the three sections [87]. More legal provisions are available to control the situation all over the world, for example, the European Commission has enhanced the ultimate target of zero landfill by the year 2025. Therefore, landfill is the last undesirable resort because of the drawbacks of the disposing, such as high environmental cost, the co-production of explosive gases, and plastics' low biodegradability [88].

Sorting and separation

WEEE is a complicate mixture, which needs to be sorted and separated firstly before the recovery or recycling. The treatment techniques include sorting, size reduction and mechanical recycling, are commonly applied to concentrate the substances and generate materials, such as metals, ceramic materials, and flame retarded plastics. The recycling process of WEEE to recover materials includes several sections including preprocessing, recovery, and disposal. The process can be carried out manually, automatically, or semi automatically (the combination of manual and automatic techniques). The first stage of preprocessing is sorting, followed by selective disassemble (for separating hazardous and valuable components). Next stage is upgrading which applies mechanical or physical treatment and/or metallurgical treatment to generate materials for end refining step [89]. The sorting process with the use of mild size reduction, density separation and sensor is able to separate the plastics from back covers of LCDs [90]. The mostly employed sorting and separation method in the world may be the manual separation utilizing manpower to separate the plastics. The other techniques are based either on physical properties or on chemical compositions of plastics.

Physical-based sorting and separation technology

The mostly applied technique based on physical properties is to separate the plastic by density. Float-sink segregation is an example using density difference to separate plastics. As the density of components in plastic are different in wide ranges, thus, the separation by density difference is complicated. In addition, additives, such as fillers and foams, can further affect the separation process by adjusting the density.

The physical parameter of plastic triboelectric effect can be also used to separate the positively and negatively tribo-charged granular plastic mixtures which possess similar properties, such as density or electric conductivities [91,92]. In addition, the magnetic separation is applied for detecting ferrous components, while electrostatic separation is the solution for nonferrous parts of WEEE plastic [93]. Recently, Robert. Et al applied the electrostatic separation technique with a metallic belt-type electrode to separate the End of Life Vehicles (ELVs) [94]. In addition, to further advance the recycling of plastics from waste electrical and electronic, two stage electrostatic separators were invented and shown enhanced separation efficiency [95].

Chemical-based sorting and separation technology

Sorting and separation can be also conducted based on different chemical properties of plastic. For example, the difference on the colors or transparency of the plastics brings the optical separation

Table 4Terminology used in different types of plastic recycling and recovery [117].

ASTM D5033 definitions	Equivalent ISO 15720 definitions	Other equivalent terms
Primary recycling Secondary recycling	Mechanical recycling Mechanical recycling	Closed-loop recycling Downgrading
Tertiary recycling Quaternary recycling	Chemical recycling Energy recovery	Feedstock recycling Valorization

to be possible and optical sensor can be applied to separate plastics. Near infrared (NIR) and Raman spectroscopic methods are also investigated for separation, but they are more expensive [96–99]. Recently, sliding spark spectroscopy and X-ray Fluorescence are suitable to detect metals and halogens in polymers directly [100–106]. During the process, the addition of tracers (rare earth oxides) can benefit the promotion of sensitivity [107]. In addition, laser induced breakdown spectroscopy (LIBS) is a spectroscopic technique which is based on the analysis of spectra from the laser-induced plasma, and it is a relatively new analytical method. LIBS has powerful ability to provide the fast identification of various plastics (HDPE, LPDE, PP, PS, PET and PVC based on carbon and hydrogen (C/H) line intensity ratio) [106,108–112].

Currently, the sorting process is inefficient and complicated. Developing more efficient methodology is required. Artificial intelligence (for example, machine learning) may apply for robotics and separation, thus help on this technology evolution.

Treatment and recycling

Development of environmentally sustainable approaches of WEEE recycling is urgent not only from the aspect of waste management but from the consideration of energy saving, environmental protection, as well as economic and political [7,113,114]. In addition, recycling polymer from WEEE can save up to 80% energy compared with the virgin material production [29]. For example, according to a cost model calculation, it is showed that the energy for recycling 1 kg of ABS polymer is 71.96 MJ, while 95 MJ is needed to produce a virgin ABS polymer [115]. In the recycling process, CO2 emissions can be reduced by 2.5 Mt annually for every 9.5 Mt of WEEE recycled [116]. Furthermore, sustainable recycling of plastic provides the feedstock to replace the virgin polymer as well as valuable chemicals or fuels [29]. After plastic is separated from equipment, it can be recycled by several different methods as listed in Table 4, which will be summarized and discussed in detail in the following sections.

Recycling methods

Primary recycling is the most popular and direct process considering their simplicity and cheap price. This process is to reuse the end of life of product by the original structure but the recycle efficiency is limited by the cycle's times. Mechanical recycling (or secondary recycling) consists of reusing a plastic material in similar products which supply the high values of waste plastics. However, loss in mechanical performance of waste plastics will narrow the applications [118,119]. For instance, mechanical recycling of thermoplastic polymeric components (HDPE and LDPE) has been developed, while the recycle of ABS/PC thermoplastics has not been developed. In addition, it is found that mixed recycled polymers typically show weak mechanical performances, thus may limit their application in demanding conditions [15,120]. Waste plastics can be mixed in a molten state in a twin-screw extruder with virgin polymers, fillers, fibers, and various additives. The tertiary recycling, also known as chemical recycling, can convert plastics into monomer or mixtures of chemicals or gases by (a) thermal

cracking or pyrolysis, (c) hydrothermal treatment or supercritical fluid treatment, and (d) gasification. Energy recovery by thermal recycling or valorization is used to recover the heat of combustion. At the meantime, the volume of organic materials is reduced in the process which involves the recovery of energy by incineration. However, toxic substances from the combustion make the process ecologically unacceptable for most plastic wastes [5,40,121].

Thermal cracking or pyrolysis

During feedstock recycling (chemical recycling), the nature, amount, and behavior of components may change and thus affect the reaction products. In addition, it is difficult to predict the breakdown mechanism, this is more serious in waste plastics with unknown components [114,122–125]. The main gaseous and liquid products of the process are hydrocarbons, which can be used as feeds for producing monomers or as power station fuels. In the traditional chemical industry process, raw materials account as high as 60% of the production costs, thus feedstock recycling provides an alternative way to provide cheaper raw materials to produce valuable products. Great efforts have been made to develop economically feasible pyrolysis techniques and the chemical recycling is regarded as an important process to treat the end-of life electrical and electronic devices [126–129].

Thermal cracking or pyrolysis is commonly applied to recycle heterogeneous WEEE plastics containing contaminates and additives and to produce valuable condensable liquid products for liquid fuel production or for feedstock recycling [129–132]. The products from pyrolysis are mainly divided into three parts: liquid, gas, and solid. Liquid products are mostly a complex mixture of valuable organic chemicals, including toluene, styrene, ethylbenzene, aromatic, aliphatic, xylene, single-ring hydrocarbons, etc. [133–139]. Gas products include some light hydrocarbons (C1-C4), CO2, CO and H2, etc. [127,132–134]. The residue of the process comprises solids which are mainly carbon, char, and some metals, such as Cu, Zn, Al, etc. [133,134,139]. Different pyrolysis conditions (operating temperature, heating rate, reaction time, reactor type, and operating pressure) generate different products and product distributions, including the chemical compositions of virgin polymer [114].

The lab-scale setups for pyrolysis in the references are shown in Fig. 1. Both fixed bed and fluidized bed are applied for the pyrolysis, and microwave is beneficial to produce liquid products. Compared with conventional thermal treatment, the catalytic cracking process has advantages: lower reaction temperature and shorter residence times (lower energy consumption), higher yield of potential fuels with high quality, lower formation of the undesired products [123,140]. The catalytic pyrolysis process applies catalysts (Ni/Mn/Al, zeolite ZSM-5, Y-zeolite, biochar, electronic waste char (EWC), Fe/YZ, Fe/ZSM-5, Fe/BC and Fe/EWC, HUSY, FA, FAMA, FAMB, etc. to target value added products using less energy [127,135,137-139,141]. Catalysts applied in the process include homogeneous (such as aluminum trichloride) and heterogeneous types (such as Zeolites: HZSM-5, H-Beta, and HY) [142-145]. Novel catalysts were further developed for this process, such as (ZSM-5), hierarchical zeolites [146,147], nanocrystalline mesostructured catalysts (MCM-41, FSM-16, Al-SBA-15, AlUTD-1), etc. [140,148–151]. The rapid development of recycling technique may accelerate the conversion of waste plastic to pure virgin polymer resources without toxic components, and also the production of chemicals and fuels with economic and environmental benefits [15,152,153]. The current catalysts still have many problems, such as low selectivity to specific products and low stability due to carbon deposition or coking. Therefore, more research work is needed to develop more efficient catalysts for the pyrolysis of plastic waste.

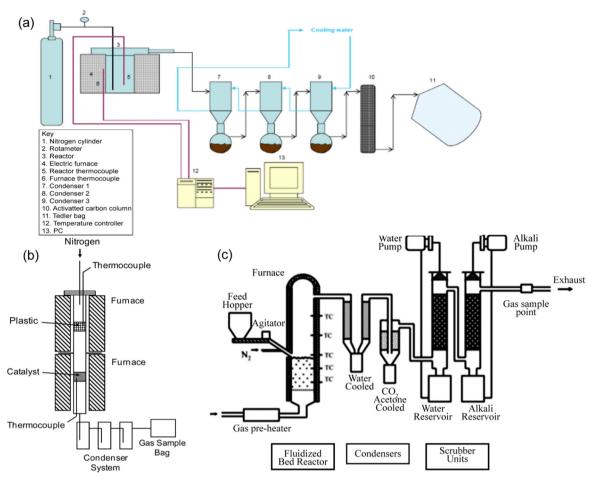
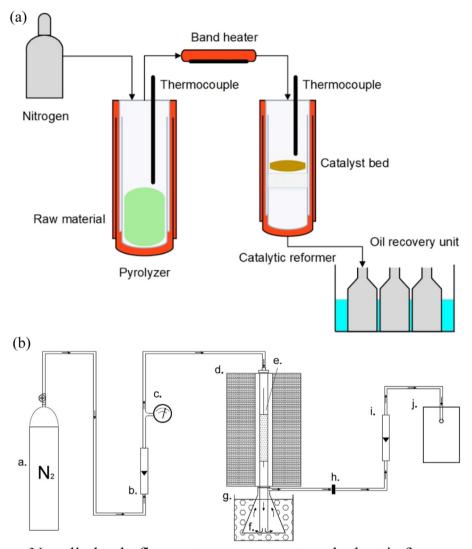


Fig. 1. (a) Pyrolysis reactor for recycling plastic rich waste [133]. (b) Two-stage pyrolysis – catalysis batch reactor system [132]. Fluidized bed pyrolysis reactor. (Figures were reused with the permission from the respective journals).

In this laboratory-scale system shown in Fig. 1(a), plastic waste is placed in a 3.5-litre semi-batch pyrolysis reactor heated by an electric furnace. Thermocouples in the furnace and reactor were used to monitor and control the temperature. The products are passed through three-stage condensers to separate the solid, liquid, and gaseous products. An activated carbon column is used to adsorb harmful organic waste products and a sampling bag. Typical conditions are 500 °C, 30 minutes and 1 bar. The limitation of this process is that the solid carbon product remains in the reactor. In addition, it is difficult to scale it up for continuous operation because it is a batch reactor [154,133]. In Fig. 1(b), a two-stage vertical pyrolysis reactor is used in which the plastics change phase in the first stage and pass through the catalyst bed in the second stage. Typically, 2 kg of plastic and 2 g of catalyst are used, and the reaction is carried out at 500 °C for 30 min. The pyrolysis products are condensed in three-stage condensers and the gas is collected in a gas sampling bag [132]. The two-stage reactor allows the catalytic pyrolysis of the plastic to target specific product as end-product. The catalyst and its regeneration are the challenging issues for the scaling up. In Fig. 1(c), preheated N₂ gas was used as the fluidization medium and injected into the externally heated tubular reactor from below. The fluidization bed consists of sand $(300-425 \mu m)$ and the plastic sample was added to the bed using a screw conveyor. Halfway into the reactor column, an N2 gas was purged to prevent backflow from the bed. The gaseous product from the bed exited the reactor and passed through the condenser system. Uncondensed gases then passed through the gas purification system, which consisted of water and alkali scrubbers. The fluidized bed system is suitable for catalytic pyrolysis, where the sand bed can be replaced by a catalyst bed and a catalyst recirculation system can be integrated into the reactor, which can be used to regenerate the catalyst. However, fluidized bed pyrolysis is more suitable for fast/flash pyrolysis systems where the goal is to gasify the plastics, while this type of system is not recommended for plastic liquefaction or low temperature slow pyrolysis [155,156].

Areeprasert and Khaobang [136] applied a two-stage pyrolysis system for reforming the pyrolysis products from PC /ABS and non-metallic components of PCBs using biochar as a catalyst (Fig. 2(a)). In the first stage, the plastics convert into gas phase and pass through the catalyst bed for reforming in the second stage. Catalytic reforming can improve the properties of the liquid fuel and also have the potential to produce H₂ gas. The process is limited by the efficiency and cost of the catalyst, and a two-stage system makes it more complicated to maintain process parameters during scale-up. Santella, Cafiero [137] also used a tubular reactor with a catalyst bed for the conversion of the mixed e-waste plastic stream into valuable liquid and gaseous products at high temperature (600-800 °C). The process is a batch system where the products generated from the catalyst bed pass through the bottom opening into a cold trap and are condensed (Fig. 2(b)). Uncondensed gases were collected in a gas sample bag. N₂ purging helps to inert the reactor environment and transport pyrolysis products out of the reactor. Catalyst loading and replacement could be an issue with this type of system when scaling up occurs. Pyrolysis systems are commercially available in a variety of sizes for both batch and continuous operation. The pyrolysis process consumes energy to convert the plastics into a gaseous or liquid form. The liquid products from plastic pyrolysis must be further processed to be



a. N_2 cylinder, b. flowmeter, c. manometer, d. electric furnace, e. reactor, f. flask for condensed products collection, g. cold trap, h. filter, i. flowmeter, j. gas sample bag.

Fig. 2. (a) Reactor of pyrolysis and catalytic reforming [136]. (b) Thermal and catalytic pyrolysis reactor [137]. (Figures were reused with the permission from the respective journals).

used as fuel for transportation or as monomer to produce new polymers. In comparison, pyrolysis of plastics is more sustainable than incineration and landfilling. The idea of treating contaminated e-waste is still subject to product decontamination technology and process economics, and research to date on this topic is still limited.

Pyrolysis is the mostly applied chemical recycling technology to recycle the heterogeneous WEEE plastics containing contaminated and additives and has been studied widely in recent years. In pyrolysis, product selectivity still needs to be optimized with product values. Due to the high cost of pyrolysis equipment and the high energy consumption, researchers should consider the overall benefits to design the pyrolysis process and dehalogenation to obtain maximum products values. Further research may focus on the scale up and optimize the operational conditions to realize large-scale applications.

A few additives which are considered as carcinogenic and is required to be maintain the concentration below the RoHS limit

[157]. These additives are PBB, PBDE, Cd, Cr6+, Pb and Hg [158,159]. Therefore, effective chemical recycling is necessary to reduce the toxicity contents in the disposal waste. In the chemical recycling process, three important aspects: Dehalogenation, brominated flame retardants, and heavy metal extraction, will be summarized and discussed in the following sections.

Dehalogenation of plastic during recycling

Pyrolysis can provide liquid oil products. Halogenated organic compounds often remain in the products. Therefore, dehalogenation is necessary for the recycling of WEEE plastics by pyrolysis.

Dehalogenation of WEEE plastics can be done by various methods: [5]

a) The first method consists in carrying out pyrolysis in two stages to remove halogens before the decomposition of plastics [129]. For example, the two-stage pyrolysis method is

- employed to remove HCl and HBr at lower temperatures as the first step, followed by the decomposition of polymer matrix at higher temperatures [132].
- b) The second method refers to simultaneous dehalogenation and degrading the polymer. Hydrothermal treatment using supercritical water is shown as a promising high efficient process which prevents corrosive gas formation by dissolving halogenated components into their corresponding acids [5]. In addition, catalytic cracking is another strategy to remove halogens during the pyrolysis process. Various catalysts (Y-Zeolite and ZSM-5, 4A, 13X, Al-MCM-41 and NaY, FeOOH, Fe(Fe3O4)-C and Ca(CaCO3)-C, etc) have been used in the pyrolysis to improve the removal performance of brominated compounds [160-164]. A series of catalysts were verified to be effective for the dehalogenation. Ni/AC. Ni/c-Al2O3. Ni/SiO2 and Raney Ni are able to completely eliminate chlorine in chlorinated benzene [165]. FeCl2.4H2O, FeCl3, NiCl2, CoCl2, CuBr and some iron complexes with benzimidazole ligand are favorable for hydrodebromination [166]. Moreover, Pd-Fe/C displayed high dehalogenation activity under certain conditions [167]. During the process, NaOH is beneficial for the catalysis performance and can fix Br to the solid char. The formed HBr may react with NaOH/KOH to generate the stabilized NaBr/ KBr [168].
- c) The third method is a pre-treatment, such as pollutant extraction methods (halogens and heavy metals) before chemical recycling. These extraction methods will be detailed in the following sections.
- d) The last method consists in carrying out pyrolysis and then in treating the gas phase or the liquid phase by catalytic hydrohalogenation to transform the halogenated compounds into easily recoverable inorganic compounds, which means pyrolyzing plastics first, followed by upgrading the products.

Brominated flame retardants extraction

From the section 2.2.5, BFR plays a critical role in plastic and it helps to improve the fire resistance of the electrical and electronic equipment. However, it is harmful for the environment and shows negative endocrine, reproductive and behavior effects to human body [83,169-171]. The high content in the WEEE plastic and increasing amount have made it a big concern for environmental and human health [172]. The new European Restriction of Hazardous Substances Directive (RoHS 2011/65/EU) has set the limitation of BFR concentration below 0.1% [5,173–175]. Researchers all over the world are struggling to develop effective solutions for recycling, while policymakers globally are trying to make policies for controlling the WEEE generation [22]. EC has recently launched a strategy of using recycled e-plastics to support sustainability, namely it is a more 'circular' approach for plastics and considers all the impacts from environmental, economic and social [176]. 60% of the total BFR volumes is TBBPA which is commonly found in (a) large and small household appliances, consumer electronics, food packaging, and household waste plastics [64,177 178], (b) environmental media (sludge and sediments) [70,179,180] and (c) biota samples (internal organs of marine animals and fish) [180,181].

The first step before extracting brominated species consists of the development of efficient analytical method to determine the amount of Br in the plastic before and after chemical recycling. There are some techniques, which can detect the bromine flame retardant directly with a rapid speed without degradation of the waste plastic. Handheld XRF technique is feasible and preferred for fast test of large volumes of WEEE plastics for on-site measure-

ments. Polybrominated biphenyls (PBBs) and polybrominated diphenyl ethers (PBDEs) from the black TV and grey PC plastic were tested directly after disassembling. The bromine content of over 50,000 ppm was found in 7% of TV plastic parts and 39% plastic parts of PC, which were analyzed by GC-MS [66]. This technique was further improved by combined with Micro-Raman spectroscopy, which provides the possibility of this technique to the recycling industry [182]. More recently, a novel control technology of liquid extraction surface analysis mass spectrometry (LESA-MS) was developed for direct sampling and analysis of the e-waste plastic. It is capable to detect PBDEs and TBBPA. Furthermore, the advantages of LESA-MS include fast test (less than 1 min per sample), simple operation (automated), and robust results. For example, it does not require pre-treatment, separative techniques, and dissolution of the solid sample [183]. At last, high performance liquid chromatography (HPLC) with either ultraviolet (UV) or mass spectrometry (MS) has been proven to be a robust method for the quantification of BFRs in WEEE plastic [179,184–189].

There are various extraction processes at laboratory scale: supercritical-fluid extraction (SFE), pressurized liquid extraction (PLE), ultrasonic-assisted extraction (UAE), and microwave-assisted extraction (MAE). Soxhlet extraction is the most common extraction method, which is used to extract additives from polymers. However, its long extraction times and high solvent volume consumption limit its application and development for an industrial process of extraction of BFR from plastic matrices [190,191]. Table 5 lists the different extraction methods for the BFR removal process including microwave extraction, ultrasonic extraction, solvent extraction, solvothermal technique, pressurized liquid extraction, supercritical-fluid extraction, and ionic liquid. The target FR types focus on TBBPA, PBB, PBDEs and Deca-BDE.

- (a) **Solvent extraction** is used to remove TBBPA in WEEE plastics prior to pyrolysis. The extraction process using isopropanol and toluene can reduce 36.6% of the total bromine in plastic [62,192–194]. From all the investigations, it is clear that toluene is more efficient for extracting TBBPA than isopropanol [195]. After the extraction, the solvent can be purified, recycled, and reused for other applications. As a result, the recycling efficiency by closing the loop of materials can be improved [196].
- (b) Hydrothermal treatment

This process is considered as one of the optional technologies to recycle BFR laden plastics and it can also remove bromine from oil. For instance, Kazutoshi et al. investigated the degradation of Deca-BDE using hydrothermal method. It was decomposed by more than 99% after 10 min at 300 oC with toluene [197]. This method is also able to remove TBBPA by 79.9% at 160 oC for 8 hours when adding iron powders into the system [198].

(c) Microwave-assisted extraction (MAE)

It employs microwave energy to accelerate the solid-liquid extraction by heating the solid sample and solvent thoroughly. Microwave processing is expected to be more efficient than conventional heating because of the merit of volumetric heating [204]. This process has been successfully applied to extract a wide range of additives (antioxidants, stabilizers, and plasticizers) with higher efficiency, lower solvent consumption, and shorter extraction time [205–208]. Furthermore, MAE process has also been applied to extract the brominated flame retardant from a series of WEEE plastics including ABS PP, PC, PC/ABS, HIPS, etc., which were collected and dismantled from microwave ovens, irons, vacuum cleaners, DVD/CD players, computer housing, and the blended plastic [188,189,199]. The microwave extraction efficiency is influ-

Table 5Extraction methods applied in the BFR removal process.

Method	FR type/Removal efficiency	Polymer sample	Detection technique	Optimized condition	Ref.
Microwave extraction	TBBPA/100% Deca-BDE	HIPS	HPLC-UV	Isopropanol/n-hexane, 130 °C	[188]
	TBBPA	ABS	HPLC/MS	Isopropanol/n-hexane 1:1, v/v	[189]
	PBB and PBDEs	WEEE plastic	GC/MS	Hexane/H ₂ O = 5:2, 100 °C, 10 min	[199]
Ultrasonic extraction	TBBPA	WEEE plastic	HPLC-DAD	Isopropanol/n-hexane 1:1, v/v, 40 KHz, 30 min	[189]
Solvent extraction	TBBPA/36.5% (bromine content)	WEEE plastic	SEM-EDS	Isopropanol (132 °C) and toluene (153 °C), 6 h	[62]
Solvothermal technique	ТВВРА	Waste computer housing plastic (ABS)	Oxygen bomb combustion-IC	Methanol, ethanol, and isopropanol, 90 °C, 2 h, 15:1	[200]
Pressurized liquid extraction (PLE)	PBDE and PBB	PP, PE and ABS	GC-ITMS-MS		[201]
Supercritical-fluid extraction (SFE)	TBBPA	Waste computer housing plastic	Potentiometric titration	Water, methanol, <u>isopropanol</u> , and acetone, 400 °C.	[202]
Ionic liquid	Deca-BDE/92.7%	HIPS		Ethyl acetate, 77 °C, 10 min	[203]

enced by the parameters, such as the solvent, temperature, extraction time, sample size, etc. The popular solvents include isopropanol, n-hexane, etc. [188,189,199]. A mixed solvent of isopropanol and n-hexane results in the high extraction rate due to the high affinity to the analytes and the great swelling effect on the matrix. High temperature is critical for the bromine extraction and the glass transition temperature of the polymer is the demarcation point: higher temperature should be used to obtain higher diffusion rates [199]. The effect of particle size on the extraction recovery was verified, which suggests that the diffusion of the additives in the polymer core may not be the limiting stage in the extraction [188].

(d) Dissolution technique followed by precipitation of Liquid/ Liquid extraction. This method (Creasolv® process at semiindustrial scale) consists of dissolution of polymer into a solvent. The precipitate is the "pure" polymer without any pollutant, such BFR, by adding a non-solvent [209]. The BFR and other pollutants species can be extracted and remain in the solvent-non solvent mixture during the precipitation step. The organic solvents used can be recycled, leading to the use of a small volume of solvent compared with the volume of materials (<1%). Once processed and dried, the polymer exhibits good mechanical properties and is compatible with RoHS regulations. This process was applied to extract the flame retardants (Deca-BDE and antimony trioxide) from a high-impact polystyrene with the extraction efficiency of above 90% and the residue can be used for various applications because the chain length of the polymer is not degraded by the extraction process [203].

Pressurized solvent extraction (PSE) system is another BFR removal process with the usage of gas chromatography coupled to an ion trap tandem mass spectrometry (GC-ITMS-MS), which provides a speed, low cost, simple, and reliable analytical method for the BFR removal. The extraction efficiency from certified reference materials (CRM) was obtained from 79.6% to 93.7% [201].

(e) Supercritical-fluid process can be also used to extract BFR from WEEE. This method is known as supercritical fluid extraction (SFE). CO2 is brought to its supercritical state (Temperature higher than 31.3 °C and Pressure higher than 7.28 MPa) and then introduced into the reactor containing the BFR-containing waste. The extracted BFR can be recovered after condensation in the separator. Additional cosolvent (such as water, methanol, isopropanol, and acetone) can be added to improve the efficiency of the process. The

extraction efficiency depends on both its solubility in sc-CO2 and the diffusion of supercritical fluid through the polymer matrix. In addition, the temperature and density of fluid are important for the extraction performance. It is necessary to find a relation between temperature and pressure as a function of solubility and diffusion of extract [210]. Because sc-CO2 extraction is less efficient at low pressure (10-12 MPa), some studies focus on the mixture of sc-CO2 and organic solvents to improve the solubility of BFR and the extraction yield [185]. Among the four solvents studied (water, methanol, isopropanol, and acetone), isopropanol was the most efficient solvent. The SFE with isopropanol can obtain the excellent debromination efficiency as 95.7%, produce 60% oil under a mild temperature and pressure [202]. The kinetics study demonstrated that the optimum operating conditions were 60 °C and 25 MPa with a medium stirring speed of 1000 revolutions/min [211]. Among the many parameters, temperature and particle size play important roles, while pressure and polymer type (ABS, PS or HIPS) have little impact on the extraction kinetics. To improve the extraction rate, dynamic extraction with circulation of sc-CO2 was recommended to compensate for the low solubility of RFB in the fluid.

So far, the flame retardant extraction has been investigated extensively. Various methods have been developed and some of them have shown promising potential in the application. Further research may put more efforts on the scale-up, further enhance its efficiency, and reduce cost.

Metal analysis and extraction

RoHS (Directive 2002/95/EC) has launched the regulations of the heavy metals including lead (Pb), mercury (Hg), cadmium (Cd), and hexavalent chromium (Cr (VI)) [17,173,212]. A great variety of procedures for metals determination in WEEE have been published but mostly focused on PCBs, batteries, or other devices with rich metal concentrations [16,85,213–217].

Firstly, several methods have been developed to determine metal concentrations in WEEE plastics. The determination and extraction of metals are not easy. X-ray fluorescence (XRF) is a powerful technique to test metal concentrations in soil, coal, vegetables, water, glass, paints, and hairs including Zn, Cu, Pb, As, Ni, K, Ca, Mn, Fe, Sr, Ti, Rb, Zr, Ba, Co, Se, Mo, Hg, Cd, Sn, etc. [218–229]. This technique significantly cuts off the time required for sample characterization and provides the fast test for large scale

Table 6The analytical methods for heavy metals from e-waste plastic.

Method	Detection method	WEEE plastic sample	Metals/Removal efficiency	Optimization condition	Ref.
Wet acid digestion	flame atomic absorption spectroscopy (FAAS)	European Reference Material ERM-EC680	Cd/98% Cr/99.1% Pb/94.1%	0.3 g sample, HNO ₃ (4 ml, 155 °C, 30 min) + HClO ₄ (1 ml, 175 °C, 20 min)	[238]
Leaching	AAS	Waste mobile phone	Cr/0.11 mg/L, Cu/1.78 mg/L, Ni/ 1.57 mg/L, Pb/5.33 mg/L Zn/ 1.78 mg/L	Milli Water, 160 days	[246]
Leaching	ICP-OES	ABS computer housing	Sb/47.9%	Sodium hydrogen tartrate (0.5 M) dissolved in either DMSO, (S/L = 1:14) 100 oC, 20 h	[247]
Microwave digestion	ICP-OES	in-house polymer reference materials (RMs).	Cr(VI)/96%	pH = 2.0, NaOH + NaCO3, room temperature, 60 min	[248]
Microwave digestion	ICP-OES	small WEEE	Pb: 9.15 to 21.31 mg/kg; Cd: 0.49 to 19.61 mg/kg; Cr: 1.47 to 14.85 mg/kg.	250–320 mg solid sample, 5 ml 65% HNO $_3$ with 0.5 ml of 30% $H_2O_2,\ 190\ ^\circ C$ for 15 mins	[239]

samples. However, it requires certified reference materials (CRM) for calibration, leading to the high cost of this technique. Neutron activation analysis (NAA) is more sensitive and reliable for the qualitative and quantitative multielement analysis of major elements and also trace elements in samples [230]. NAA can only give the total element concentrations, but the structure of the chemical compound and physical state cannot be achieved. Furthermore, the rare access to the nuclear reactor slows down the application widely [231]. There are different types of NAA. Instrumental neutron activation analysis (INAA) is commonly used and offers high sensitivity. However, heavy metals in the environment, such as Cd, Pb, and Ni, are not easily determined. Radiochemical neutron activation analysis (RNAA) possesses high sensitivity with accurate value but requires chemical decomposition of sample and testing time is long [232-240]. Moreover, graphite furnace atomic absorption spectroscopy (GFAAS) was applied directly to detect metal concentrations of solid samples. The limits of detection of Cd and Pb are at the level of ppm [240–242]. Apart from the direct analysis techniques, atomic absorption spectrophotometry (AAS), AAS, ICP-OES, ICP-MS, and GFAAS are commonly used to measure metal concentrations in solution after the sample decomposition [225,243-245]. The methods of solid sample decomposition include acid leaching, wet acid digestion, microwave digestion, etc.

Table 6 lists some removal processes for heavy metals in WEEE plastics. Wet acid digestion was developed to determine the concentrations of Cd, Cr, and Pb in plastic materials using flame atomic absorption spectroscopy (FAAS). The metal removal efficiency for the European Reference material was 98% for Cd, 99.1% for Cr and 94.1% for Pb using a solvent of HNO₃ under 155 °C for 30 min, followed by digestion in HClO₄ under 175 °C for 20 min. The method was applied in the analysis of several commercial packaging materials and plastic toys [238].

Among toxic heavy metals, lead (Pb) can harm almost every organ, especially the nervous system. Children's toys and products mostly contain plastics, stabilizers, and paints (containing metal pigments, which contain various heavy metals that were added during manufacturing to improve their properties) [249]. Unfortunately, eight toxic elements (Sb, As, Ba, Cd, Cr, Pb, Hg, and Se) were found in baby toys sold in Japan [250]. The high level of Pb was detected out in the metallic jewelry collected from retail stores located in California and also in the low-cost plastic toys bought in Beijing [249,251]. The high concentration of Cd was found in the low cost jewelry purchased from American market [252]. Waste mobile phones have become serious problems because of their rapid increasing quantities. The analysis of Pb concentration in the plastics shows that it has exceeded the regulatory limits (5.0 mg/l) and can be categorized to hazardous materials [246,253]. Milli Q water was applied to leach metals in the sam-

ples, but the leaching performance was limited [246]. The acid digestion with the mixture of H2SO4 and HNO3 was used to detect high Pb concentration of (5-340 mg/kg), and Cd from 4.6-1005 mg/ kg, etc. [2]. Antimony (Sb) has been commonly used in various applications, such as lead-acid accumulators for rechargeable batteries, one of components of catalysts for polyester production, primers in ammunition, and additives in plastics. The recovery of Sb is necessary for two reasons: 1) antimony presence in the environment is not safe, and 2) the recovered Sb can be used as a raw material. A weak acid solution (heated sodium hydrogen tartrate in DMSO) was chosen to leach ABS based computer housing and the leaching efficiency was 47.9% at 100 °C for 20 hours [247]. Hexavalent chromium (Cr (VI)) compounds are genotoxic carcinogens. A optimized extraction process for in-house polymer RMs achieves a recovery of 97% [248]. Microwave digestion was employed for the small WEEE by Dimitrakakis et al. under an optimization condition of 250-320 mg solid sample, 5 ml 65% HNO3 with 0.5 ml of 30% H2O2, 190 °C for 15 min. The concentration range of Pb is 9. 15 \sim 21.31 mg/kg, Cd is 0.49 \sim 19.61 mg/kg, Cr is 1.47 \sim 14.85 m g/kg [239].

In short, various technologies have been developed and tested for the metal extraction. However, due to the complex contents of different metal elements and different sources of the WEEE, a standard method hasn't been developed at current state. Thus, it is still far from the industrial application, considering its efficiency and cost, thus, more research efforts are still needed in this area. A possible direction is developing a universal method for various metal extraction and recycling.

The WEEE plastic recycling has the potential to make the EEE industry become more circular and at the same time produces high valued products, such as virgin polymer, fuels and chemicals. Therefore, there are some industries helping to validate and commercialize the developed technologies recently. MBA Polymer inc. [254] of U.S.A. uses physical processes to separate and recycle polymers (equivalent to virgin polymers) from WEEE after removing metal, dust, wood, rubber, and glass. There are recycling plants in China and Austria are also realizing the same technology. Butler MacDonald inc. [255] recovers HIPS and ABS from e-waste plastics. The plastics were collected from e-waste recyclers. They produce the polymer pallets after separating the plastic from the other metal components and impurities. The separation process involves all types of the density separation and electrostatic separation. Spectroscopic techniques, such as LIBS and NIR spectroscopy, have been used for the plastic sorting.

Australia and New Zealand Recycling Platform (ANZRP), with the partnership of UNSW Sustainable Materials Research and Technology (SMaRT Centre) and recycler TES, has built the first microfactory of commercial e-waste in the world. This plant transforms the WEEE plastic into printer filaments for 3D Printing and can process up to 500,000 kg of WEEE plastic per year [256]. Recipo Material produces recycle plastics from e-waste plastic streams across Nordics since January 2020. Four types of high-quality polymers including ABS, PS, ABS/PC and PMMA are produced and used in the production of flat screen TVs [257]. Stena Recycling (Sweden's leading recycling company) process contaminated e-waste plastics, such as Chlorinated plastics and BFR laden plastic [258]. The U.K. recycler Axion Polymers [259] performs automated separation and recycling of WEEE plastic. However, they use mechanical means to recycle plastics. The EU-funded PLAST2bCLEANED provides an environmentally safe recycling process of ABS and HIPS from WEEE plastics. Sorted out plastics separated, and the contaminated plastics were dissolved in solvents at elevated temperatures. The toxic substances, such as BFRs, Antimony oxide, etc., were extracted using a selective solvent [260,261].

Schlummer, Wolff [262] developed the CreaSolve® process that selectively dissolve and reprecipitate polymers from the mix plastic stream and recover pure PC and ABS from the contaminated ewaste plastics. This process is currently commercialized and licensed by Fraunhofer IVV Germany. It is a close loop process and soon after its invention, this process is used by many countries to develop recycle industry. The first fully functional plant was commissioned in Unilever Indonesia in 2018 for the recycling of their multilayer packaging plastics [263].

All these case studies are still limited in terms of primary and secondary recycling. The issue of recycling contaminated e-waste is still not fully resolved in various industries, either due to economic or technological limitations. Because these contaminated e-waste plastics pose a threat to health and the environment, it is common for future researchers to develop new and sustainable processes that can recover or recycle these plastics and add value to the recycling economy.

Reuse of the recycled plastic

The recycled plastic can be either used as virgin polymer to produce new products, or used in roads, plastic lumbers, concretes, wood plastic composites, plywood, and bitumen in which the requirements of physical and chemical properties are not demanding [44,85,264–274]. Plastic lumber is another utilization direction of the recycled plastic. The main advantages of plastic lumber over real wood are that the former does not age or splinter as the real wood does: the mature manufacture and intrinsic stability of plastic lumber. However, the manufacturing cost of plastic lumber is much higher than the traditional lumber materials. Fortunately, the use of recycled virgin plastic brings hope and ensures that the product is more commercially competitive. The principal use of plastic lumber is to replace wood in areas with extreme weather. For example, plastic lumber is generally made from recycled PE in which it may have other plastics and/or fillers, such as PVC, PS, PP, PET, and other materials [44,264]. In addition, traditional concrete needs high cement content, which emits a large amount of CO2 during the production process and causes environment cost. Ecoconcrete is a kind of concrete with less cement usage and application of waste materials. For instance, plastics from computers (PP, PET, polyurethane, HIPS) can substitute natural aggregates and the amount ranges from 15% and 50% in volume [85,265]. In fact, starting in 1990s, recycled waste thermoplastics have entered the scope of research to produce wood plastic composites (WPCs). In recent years, the usage has increased significantly in the developed and developing countries [266,273]. There are some commercial applications of recycled plastics to wood plastic products in the USA [267]. Plywood is another application of recycled plastics and a kind of green and environmentally friendly wood-based panels. The hot-pressing technique for plywood production can reach optimization conditions when temperature is 150 °C with a pressing time of 5 minutes. Compared with the traditional adhesives, the formaldehyde emission was found to be very low [268]. Recycled polyethylene can be introduced to modify bitumen with the improvement of plasticity and viscosity. Compared to the bitumen, the advantages of blends of bitumen and polyethylene (PMBs) include enhancement of softening point, high flexibility and viscosity under low-temperature, low penetration grade, etc. [269]. Recycled WEEE plastics can be also used as engineering plastics [270], filler materials [271], floor carpets, flower vases, waste paper baskets, park benches, and picnic tables [272]. In all, the reuse of recycled plastic has been investigated intensively in recent years, but actual applications need to be further explored.

Managerial and institutional aspects of plastic recycling business

The recycling of plastics, including e-waste, is a socio-economic problem that must be solved through the participation of private and public entities. There must be a sustainable business model that is both economically and environmentally feasible. There are two main approaches to dealing with the plastics recycling business. One is a triple bottom line approach that considers environmental, economic, and social benefits [275]. The other approach is the circular economy model, where the concept of an economic system that replaces the idea of "end-of-life" with the concept of reducing, reusing, recycling, and recovering materials during the generation, distribution, and consumption cycle [276]. Apart from these two approaches, each business model relies on four basic frameworks: the value proposition, the customer interface, the infrastructure including supply chain management, and the financial structure. Despite generating economic value, the plastic recycling business serves a greater benefit to its stakeholders by an invaluable perspective of saving the planet earth.

Value proposition for the recycling of e-waste plastics

The value proposition is the value the company promises to deliver to stakeholders and customers. It includes marketing strategy, brand equity, operational integrity, etc. Most positive aspects of the value proposition for the recycling business are that it serves the larger objective of managing plastic pollution and solving global sustainability. One of the major challenges that plastic recyclers have is the low price of crude oil, which impacts the price of virgin plastics. When the price of virgin plastics is low, the value of the recycled plastic or the transport oil from the pyrolysis of plastic waste needs reduction. This is the financial sustainability of the recycling industry. In this scenario, public–private partnership or government intervention comes into play. The following steps could be taken to tackle this issue [277].

- i. The manufacturing industry must meet its obligation to use recycled plastics, and any failure to do so must result in serious accountability.
- Make recycled products a differentiator in the marketplace by creating awareness and making consumers proud to use recycled plastic products.
- iii. Governments around the world must support the recycling industry through incentives and tax breaks and protect it through laws and regulations.

Recycling and management of plastic waste vary from region to region, depending on the economic status and environmental regulations of the country. Eco-labelling and green purchasing are such approaches to encourage industry to adopt recycling and



Fig. 3. Examples of Ecolabels from different countries [278].

environmentally friendly technologies. The Environmental Protection Agency (EPA) of the U.S. is one such agency that regulates and addresses environmental and ecological issues related to various industries. The EU has enacted several laws and directives to regulate industries, including the e-waste recycling industry. The Waste Electrical and Electronic Equipment (WEEE) Directive 2012/19/EU is one of these regulations that set the rules and instructions for member states to comply with various aspects of the recycling process that includes disposal, collection, transportation, shipping, and recycling standards [157]. Fig. 3 shows some of the ecolabels from different countries.

Customer interface

The customer interface is the component of the business model that refers to the interaction between the firm and its customer. The customers of plastics recycling firms are generally the remanufacturing industries, and very often the source industry which created the product in the first place. The secondary stakeholders of the plastics recycling industry are the direct consumers who are the end-users of the products [279]. A survey was conducted on 2000 American residence about their plastic habits and 59% responded that they are more likely to purchase something that is made from recycled plastics and willing to pay more than the retail price of the same items made from virgin plastics [280]. However, this survey can vary greatly from country to country. Recyclers must use new technologies to produce recycled plastics that are more competitive with virgin quality, while reducing the overall carbon footprint in the recycling process.

Infrastructure

The infrastructure and supply chain of plastics recycling are the major constraints towards the growth of the industry. One of the biggest problems in e-waste plastic recycling is the treatment of contaminated plastics such as plastics containing BFR or plastics contaminated with toxic metals such as Pb, Cd, Cr, etc. The cost of decontamination of these plastics could lead to an increase in the price of recycled plastics. The growth of the recycled plastic

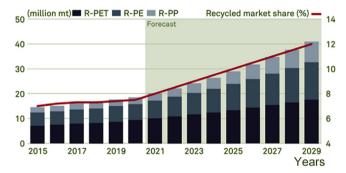


Fig. 4. Global recycled plastics set for continued growth [281].

industry is slow so far, but it is predicted to grow very rapidly in the coming years. (Fig. 4) [281]. One of the issues regarding plastic recycling is the collection and sorting of plastic waste, which is a supply chain problem. Only 9% of all plastic generated is recycled each year [282], with the rest either incinerated or sent to landfills. Poor infrastructure, poor municipal waste management and lack of awareness are the main reasons for low plastic recycling. A proactive approach by the government, raising public awareness about recycling, and using digitalization and blockchain approach to develop a new waste management system could significantly improve plastic waste recycling [283,284]. França, Amato Neto [284] presented a possible application of Ethereum's digital blockchain architecture for municipal waste management in the city of Sao Paulo, Brazil, aiming to use a digital social currency for municipal waste management and addressed a possible contribution of blockchain to sustainable development goals. Blockchain enables smart contracts between two entities in the form of computer codes that are only executed when certain conditions are met in the real world. This could lead to better coordination between producers, importers, retailers, and recyclers [283].

Financial structure

Financial structure is the hard and sole of any business or commercial institution. Access to capital or investment is one of the

biggest obstacles to the development and growth of recycling enterprises. The recycling industry needs to update its process technology and business models as consumer behavior is constantly changing specifically in the area of electrical and electronic equipment. For small businesses or startups, access to capital is a challenge. The government needs to find a way to encourage these small startups by providing loans or join them based on the public-private partnership (PPP) financing model. It is easier for a profitable recycling company to get money from private investors, banks, and other financiers. In the long run, each company must have its own financial strategy to grow larger. To ensure a constant inflow of funds, the company needs to tap into different sources of money. One way to do this is to share ownership with other investors to raise funds, getting debt from banks, and equity financing through the sale of stock, warrants, and bonds [285]. Once the corporation grows enough to seek more financial inputs, it can move to Initial public offering (IPO) [286]. All these financial transactions are controlled, regulated, and supervised by the government authorities of every country.

Conclusion

With the advancement of technology, the consumption of electrical and electronic equipment will continue to increase in the future. To achieve the goals of the circular economy, a sustainable approach to recycling all components of WEEE must be developed. A significant portion of e-waste consists of plastics, which are often ignored and disposed of with other municipal plastic waste. The recycling strategy for e-waste plastics requires special attention because they are often contaminated with BFRs and toxic metals (Cd, Cr, Pb, and Hg). Conventional leaching and extraction processes are not sufficient to treat these contaminated plastics in terms of polymer recovery and removal efficiency. More comprehensive solutions involving physical, chemical, and thermal processes are recommended. The challenges of the recycling industry in terms of financial and business outlook can be mitigated through public awareness, private and public partnerships, and the application of a sustainable business model.

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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