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Review

Modern Trends in Recycling Waste Thermoplastics and Their Prospective Applications: A Review

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Abstract: Thermoplastics and thermosetting plastics are two major classes of polymers in that have recently become materials that are indispensable for humankind. Regarding the three basic needs of human beings—food, shelter, and clothing—polymers and polymer-based materials have gained pre-eminence. Polymers are used in food production, beginning with farming applications, and in the health sector for the development of various biomaterials, as well as in shelter and clothing for a variety of applications. Polymers are the material of choice for all modern-day applications (transportation, sporting, military/defence, electronics, packaging, and many more). Their widespread applications have created many negative challenges, mainly in the area of environmental pollution. While thermoplastics can be easily reprocessed to obtain new products, thermosetting plastics cannot; thus, this review focuses more on the use of waste from thermoplastics with less emphasis on thermosetting plastics. Hence, the review presents a concise summary of the availability of waste thermoplastics as raw materials for product development and the anticipated benefits. The prospects for waste thermoplastics and thermosetting plastics, the possibility of cleaning the environment, and the uncovering of opportunities for further research and development are presented. The limitations of the current methods of waste polymer recycling are highlighted with possible future prospects from newly introduced methods. With zero tolerance for polymer waste in our environments, potential uses for recycled thermosetting plastics are described. Waste polymers should be seen as potential raw materials for research and development as well as major materials for new products. Recycled polymers are expected to be processed for use in advanced materials applications in the future due to their availability. This review shows that the major source of environmental pollution from polymers is the packaging, hence the need to modify products for these applications by ensuring that most of them are biodegradable.

Keywords: waste polymers; recycling; environmental pollution; future applications

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1. Introduction

Since the first synthesis of polymers in the early 1900s, the emergence of life and technological advancements has been exceptional due to the vast applications of polymers in every area of life. Polymers have also been used as alternative materials for major traditional materials, such as plywood, metals, glass, and 'light' ceramics in, the production and fabrication of consumer products over the last few decades. This change has occurred because of the excellent properties of polymers, as they are light in weight and durable, good insulators, low thermal conductors, have high impact and corrosion resistance, are resistant to most chemical attacks, are easy to form, are diverse in application, and have a low cost of production. Polymer-based products have provided alternative options due to their unique properties, and they are responsible for reductions in the cost of production [1,2]. The development and fabrication of polymers have significantly increased over the past decades due to their exceptional physiochemical properties, which are suitable for a variety of products for different forms of technological development [3]. The polymer revolution, in which chemists have developed new methods to overcome polymer limitations, has resulted in exponential growth in polymer production and applications [4]. Consequently, the continued development of polymer technologies is observed each year. Recent statistical data show that, during the last decade, global production of polymers grew by an average of 37%, which is equal to an increase at a level of about 100 million metric tonnes of polymeric materials. Financial data indicate that the size of the global plastic market in 2020 was approximately 579.7 billion US dollars, indicating a value growth of approximately 15.5% over the last five years. According to estimations, by 2028, the value of the global plastic market should reach 750.1 billion US dollars, with a compound annual growth rate of 3.4% between 2021 and 2028 [5,6].

However, one major concern for polymer-based materials is disposal because it takes a long time for polymeric materials to degrade. Thus, their long-lasting existence often causes environmental pollution. Although the most cost-effective way to dispose of these polymeric wastes is to recycle and reuse them in the development of secondary materials, this process has not been widely promoted by researchers. While various studies have been performed on the development of biodegradable plastic-based composites using natural fibres as reinforcement, damaged polymeric materials are often incinerated, and the burning process causes the release of toxic gases into the environment. Despite this challenge, the advantages of polymers combined with the incessant increase in the population will increase demand for them for various applications [7]. Thus, the emergence of recycling processes with the introduction of additives to help replenish the physical and mechanical properties of the recycled polymer is a good initiative. It has been emphasized that recycling helps to overcome the environmental problems associated with incineration and ensures the recovery of materials and energy [8].

Diminishing petroleum sources, appropriate regulations, and greater awareness within society have caused the sustainable development of the manufacturing and recycling of polymer-based composites to gain increased attention. Similarly, rising global demand for polymer-based components has increased post-production and post-consumer waste. Therefore, economic management and further recycling of a large volume of waste plastics remain serious environmental problems [6].

The first step in the recycling of polymers is the suitable classification, separation, and cleaning of the waste materials. A comprehensive review of the possibility of applying turbo-charging and electrostatics for the separation of polymers was reported by previous researchers [9,10]. As a result, having a basic understanding of how to process waste from thermoplastics and thermosetting plastics is critical in today’s world for effective handling and processing.

The characteristic features of polymers are solely determined by the degree of polymerization and their structural properties. Elastomers, thermoplastics, and thermosets are the three main classes of polymers based on their behaviour. However, based on their widespread applicability as composite materials, thermoplastic- and thermoset-based composites are predominant. Hence, this review focuses on these two major polymers, from which most polymer-based waste products emanate. Thermoplastics have a linear or branched structure and become soft when heated, whereas thermosetting polymers have a cross-linked and network structure and become stronger and harder when heated. The major distinction between thermoplastics and thermosetting polymers occurs due to their molecular structures and processing routes. The classification of polymer materials based on their molecular structures and fabrication techniques greatly affects the procedures undertaken before and after recycling. Because each step of recycling necessitates a different methodology to maximize recovery potential, major considerations must be made when recycling polymers [11,12].

Many research works have reported the use of recycled thermoplastics for the development of polymer-based composites [8,13,14], with less emphasis on the use of only waste materials. Hence, this review was performed to advance the development of new composite materials (matrix and reinforcement) predominantly from waste as raw material, as proposed by some researchers [15]. This process is expected to promote a more innovative approach to curbing the negative environmental impact of improperly disposed of waste plastics and the conversion of such materials into sustainable materials, thereby finding alternative applications and promoting zero tolerance for waste [15]. Products are now expected to be developed solely from waste in which plastics are predominant. As new environmental regulations begin to target plastic products made from both thermoplastics and thermosetting plastics, there is a need to consider the positive environmental impacts of the materials and their future benefits. Waste plastics account for 11.2% of the annual 84,200 metric tonnes of municipal waste stream generated in Tehran in 2006, while they accounted for 12.4% of the 250 million metric tonnes generated in the USA in 2010 [14]. This predominance is also evident in most African countries, particularly Nigeria, where waste plastics account for the majority of pollution on land and in the oceans [16]. The introduction of traditional residential waste plastic recycling techniques, which use only a few types of plastics, including low-density polyethylene, high-density polyethylene, and polypropylene, has helped to address this issue to an extent [17]. Normally, it is expected that the recycled material must be processed in ways that reduce contamination [18]. Thus, more advanced methods to process other waste thermoplastics and thermosetting plastics are essential for the development of eco-friendly products.

This current review identifies waste polymer-based materials from packaging as the major source of land and sea pollution globally compared to other areas of application. Almost all products are packaged with polymer-based products in one form or another, hence the need to work on how to reduce the negative environmental impacts of products targeted for such applications.

2. Global Production of Polymers

Figure 1 depicts global polymer production from 2016 to 2020, while Figure 2 depicts the percentage distribution of global virgin polymer production. Figure 1 reveals a progressive increase from 2016 to 2019, before a slight decrease in 2020 due to the COVID-19 pandemic. This increase showed that the demand for and availability of polymers during these years were on the rise. Similarly, the global acceptability of polymer-based materials is presented, including thermoplastics, polyurethanes, thermosetting plastics, elastomers, adhesives, castings, sealants, and PP fibres and excluding PET, PA and polyacryl fibres.

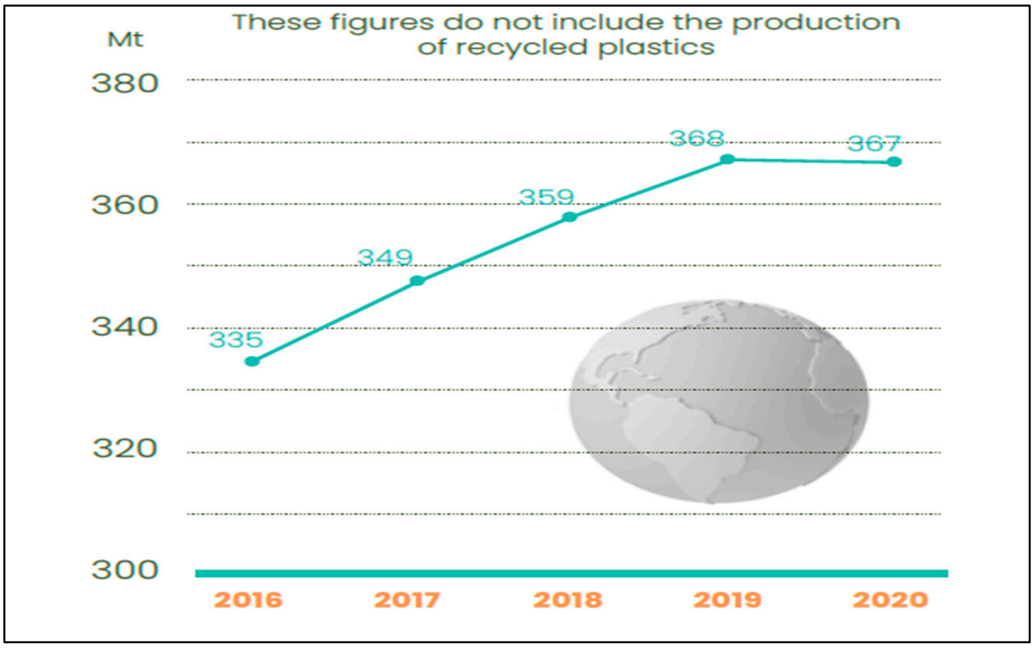


Figure 1. Polymer production from 2016 to 2020 (Mt) worldwide [19].

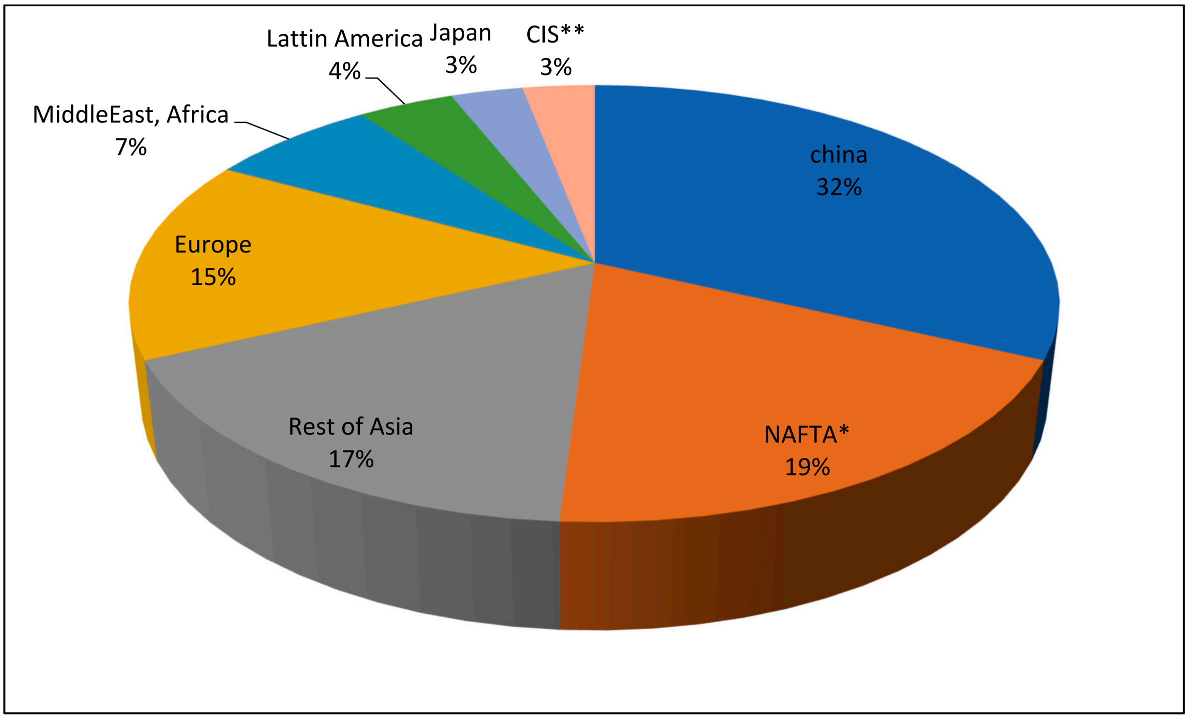


Figure 2. Distribution of polymer production worldwide [19]. * North America Free Trade Agreement. ** Commonwealth of Independent States. Not included are PET-, PA- and polyacryl fibres.

Over the years, polymer production has increased in a huge surge, as have polymer wastes. According to the United Nations Environmental Programme, about 300 million metric tonnes of plastic waste are generated annually from industrial and domestic activities globally, and around 9% of that production is recycled. This percentage is relatively low, and more attention needs to be given to how to reduce the amount of waste from polymer-based materials in our environments. It is projected to be a billion metric tonnes by 2050 [20]. Globally, polymer waste has constituted an unprecedented pollution issue due to a lack of suitable disposal techniques, ranging from litter on land and in landfills [2] to clogging of water resources and waterways, leaching into the body, and transferring through the air [21]. This process has resulted in the death of a huge percentage of aquatic and terrestrial creatures due to the hazardous nature of polymer waste, its non-biodegradability, and the emission of toxic gases when the waste is exposed to or heated without control [12]. Figure 3 presents the demand in Europe from 2019 to 2020.

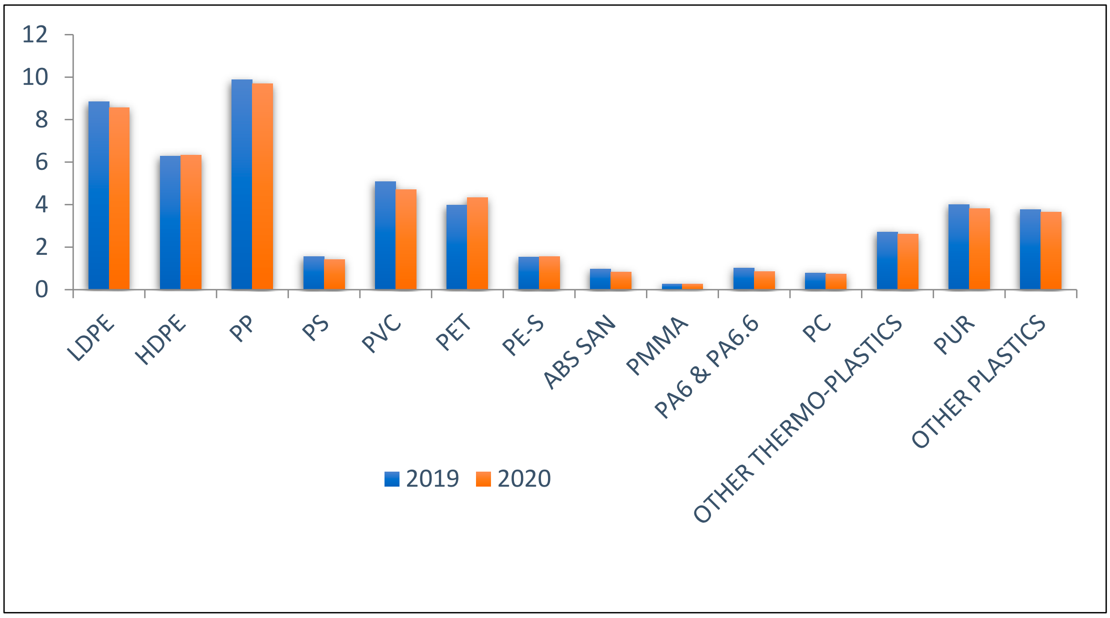


Figure 3. Demand for polymers in Europe from 2019 to 2020 [19].

2.1. Thermoplastics

Thermoplastics are physio-chemically characterized by a broad range of plastic materials that, when subjected to heat, will melt and that will flow and solidify when cooled. They maintain their ability to melt and flow on subsequent reheating [22]. These properties are essential and aid in the recycling of thermoplastics by re-melting them. However, to better appreciate this ability, a clear understanding of the morphology is needed.

In polymer chemistry, morphology is a key factor in describing the distinction between amorphous and crystalline solids. Polymers with an amorphous morphology have their atoms held together in a loose structure that is not orderly or predictable; hence, amorphous solids have no long-range order. However, in crystalline polymers, the chains behave differently by forming orderly stacks of folded chains, known as lamellae. Lamellae bring long-range order to polymers, which is more like the orderly arrangement of atoms in typical crystals. Nevertheless, some lamellae in certain polymers have small numbers of chains that loop out from the orderly stacks and create amorphous regions in an otherwise crystalline polymer. Hence, most crystalline polymers have amorphous regions, which means that crystalline polymers are never completely crystalline. Crystallinity can range from 0% to 100% to denote entirely amorphous and entirely crystalline, respectively, with most polymers falling somewhere between these two extremes. Chain flexibility, both along the entire chain and in bonds between atoms, plays a large role in polymer crystal formation. As chains flex and bend against each other, various attractive and repulsive forces affect how polymer chains arrange themselves, in either more orderly or less orderly processes. Although 100% or 0% crystallinity is rare, some polymers fall close to the extremes. Those that tend towards high crystallinity are rigid with high melting points and are less affected by solvent penetration. Conversely, those that tend towards high amorphousness are softer with glass transition temperatures and are penetrated more by solvents than their high crystalline counterparts. Examples of crystalline thermoplastics are acetals, nylons, polyethylene, polypropylenes, and polyesters, while amorphous thermoplastics include polymethylmethacrylate, polystyrene, acrylonitrile-butadiene-styrene, and polycarbonate [8,12].

Processing Conditions for Thermoplastics

To effectively recycle thermoplastic polymers, knowledge of the degree of crystallinity is essential. The degree of crystallinity is directly related to whether a polymer melts like a typical solid or whether it transitions between glassy and rubbery states. Highly crystalline polymers have a more traditional melting point, so when they are heated, they reach a certain temperature at which the orderly arrangement of their long-chain structure transits to a random and disorganized arrangement. This value is usually a specific number, which is known as the melting point (T_m).

Amorphous solids, on the other hand, do not flow suddenly when heated. Instead, they reach a range of temperatures over which the material becomes less glassy and more rubber-like or vice versa. As a result, amorphous polymers do not have a melting point; rather, they have a glass transition temperature (T_g). The glass transition temperature of a specific polymer may be listed as a single temperature, but this number is a representative value representing a range of temperatures. Hence, to adequately recycle any thermoplastic materials, knowledge of the melting status is indispensable. Understanding this property will guide in the sorting of waste thermoplastics and the selection of the appropriate processing techniques. Table 1 highlights some of the above-mentioned thermoplastics and their properties.

Table 1. Properties of basic industrial thermoplastic [8].

Properties	Limits	PP	LDPE	HDPE	PC	PBT	PAI
P (g/cm^3)	Upper	0.920	0.925	1.000	1.24	1.35	1.451.28
	Lower	0.899	0.919	0.941	1.19	1.23	
Tg	Upper	−10.000	−125.000	−100.000	150.00	45.00	290.00
	Lower	−23.000	-	−133.000	140.50	20.00	244.00
σ_{max} (MPa)	Upper	41.400	78.600	38.000	72.00	55.90	192.00
	Lower	26.000	4.000	14.500	53.00	51.80	90.00
E (GPa)	Upper	1.776	0.380	1.490	3.00	2.37	4.40
	Lower	0.950	0.055	0.413	2.30	-	2.80

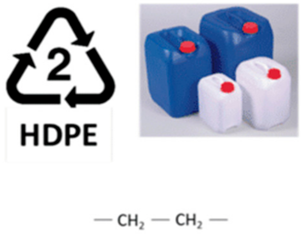
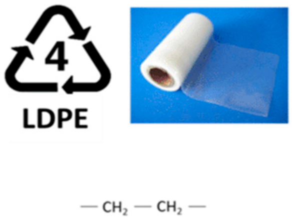
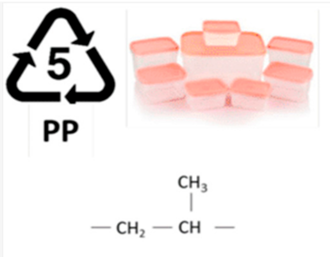
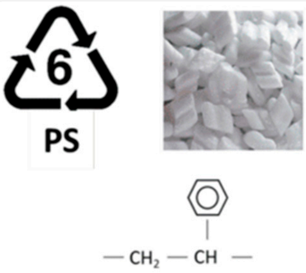
PP—Polypropylene; LDPE—Low-density polyethylene; HDPE—High-density polyethylene; PC—Polycarbonate; PBT—Polybutylene terephthalate; and PAI—Polyamide-imide.

The Society of the Plastics Industry (SPI) has assigned codes 1–6 to different thermoplastics for easy identification of the polymer used in the production of different materials, as well as to expedite the recycling process. The polymers that are classified in this way and some of their most familiar uses are summarized in Table 2.

Table 2. The Society of the Plastics Industry code, structures, and their most common applications [23].

SPI Code	Polymer	Structure	Uses
1	Polyethylene terephthalate (PET)		Soda bottles, water bottles, medicine jars, and salad dressing bottles

Table 2. *Cont.*

SPI Code	Polymer	Structure	Uses
2	High-density polyethylene (HDPE)		Soap bottles, detergent and bleach containers, and trash bags
3	Polyvinyl chloride (PVC)		Plumbing pipes, cables, and fencing
4	Low-density polyethylene (LDPE)		Cling wrap, sandwich bags, and grocery bags
5	Polypropylene (PP)		Reusable food containers, prescription bottles, and bottle caps
6	Polystyrene (PS)		Plastic utensils, packaging peanuts, and Styrofoam
7	Others		

2.2. Thermosetting Plastics

Thermoplastics are polymers with a high molecular weight that can be cured by heat, ultraviolet irradiation, electron beam processing, or chemical reactions using a hardener or catalyst [24]. The curing process causes non-reversible chemical reactions that cause the polymer chains to become cross-linked. Hence, they do not melt when exposed to high temperatures and offer superior mechanical strength. They also do not deform or lose their shape when exposed to cold temperatures. Therefore, they can be used successfully in different environments in which extremely variable temperatures are recorded. Another benefit of using thermosetting plastics is that they have enhanced properties that are obtainable due to the low cost of production when combining thermoset polymers with fibres such as carbon, glass, or aramid to create thermoset-based composites [25–27]. Hence, thermoset structural composites have been widely used in the aircraft components and shipbuilding industries because they ensure high performance in the final products, as well as allow for savings in fuel consumption due to their light weights [28,29]. Furthermore, because of their high thermal, chemical, and mechanical stability, they are suitable for structural and protective applications, such as wind turbines [30]. It has been reported that using fibre-reinforced plastic composite materials instead of metals in airplane structures contributes to a 25% CO_2 reduction [31].

The large-scale use of thermosetting plastics has led to an accumulation of thermoset waste. Currently, thermoset and thermoset-based composite wastes are being ground into fillers, incinerated, or digested using environmentally friendly technologies, while the vast majority is sent to landfills because it is considered difficult to recycle [32]. In recent years, many technologically advanced nations globally have required recycling of this waste instead of landfilling it, but still, no satisfactory way has currently been found for thermoset-based end-of-life products. The difficulty in recycling is due to the cross-linked, three-dimensional chemical nature of the thermoset matrix, which cannot be easily re-melted using heat or solvent, as happens for thermoplastic matrices; hence, recycling is often an expensive and low-rate process. Mechanical recycling allows for the recovery of lower-performance reinforcements, while chemical and thermal recycling has proved to be more commonly used and functional. Thermoset chemical recycling consists of chemically catalysed reactions that break down the polymer chains into building blocks that are reused in the same product as before the recycling process or in other, different products [33]. Similar to thermoplastics, they can be processed by melting, but their formation and cooling bring about an irreversible physical change, making them very difficult to reprocess; thus, thermosetting plastics naturally decompose before re-melting [20]. These findings are supported by the chemical structure of thermosetting plastics. The process known as curing brings about the chemical cross-linking of thermosetting plastics. As the curing process progresses, the thermosetting plastics become stiffer and, in most cases, brittle, with a dense molecular network [8,20].

Over the last two decades, the issue of recycling thermosetting plastics has been widely investigated, and although it is still an incomplete solved problem, some technologies are suitable for recycling on a large scale. These technologies apply to the most commonly used thermosets, such as polyurethane foams and epoxy-based composites. Therefore, polyurethane foams are usually converted into polyol, while epoxy-based composites are recycled through catalysed alcoholysis or are converted, under certain conditions, into recyclable thermoplastics using polyamine curing agents able to cleave at cross-linking sites [34,35]. These technologies promote the circular economy and allow for business opportunities, as the waste of low-value products is turned into high-value products. Furthermore, fibre-reinforced thermosetting composites can be recycled to replace virgin materials, reducing both the raw thermoset matrix and the raw fibres used. This process has lowered the environmental footprint, thereby contributing to a more sustainable society.

3. Recycling of Waste Thermoplastics

Thermoplastics in the present have no doubt become not only an essential part of technological development but an indispensable aspect of promoting domestic and home comfort in human lives [36]. Hence, the suitable solution that should be implemented to mitigate thermoplastic pollution is to maximize the four Rs, that is, by reducing, recovering, recycling, and reusing thermoplastic wastes [37]. Recycling seems to be the most rational, economic, and ecological way of mitigating the damage caused by thermoplastic waste materials. To mitigate thermoplastic waste through recycling, the cycling procedures are grouped into four sequential techniques, as shown in Figure 4.

Primary recycling is generally known as a closed-loop system; secondary recycling is known as downgrading; tertiary recycling is chemical or feedstock recycling, which applies when the polymer is depolymerized to its chemical constituents; and quaternary recycling is energy recovery from waste or valorization [8,38,39]. Although these methods have their limitations, they are being adapted in various industrial applications to a definite level. They have, however, improved the recycling of plastics and reduced the incineration of plastic wastes, which usually presents difficulties such as the production of poisonous gases and residue ash, which contains lead and cadmium [40]. Figure 5 shows the possible life cycle of thermoplastic materials from the first production level to the recycled product.

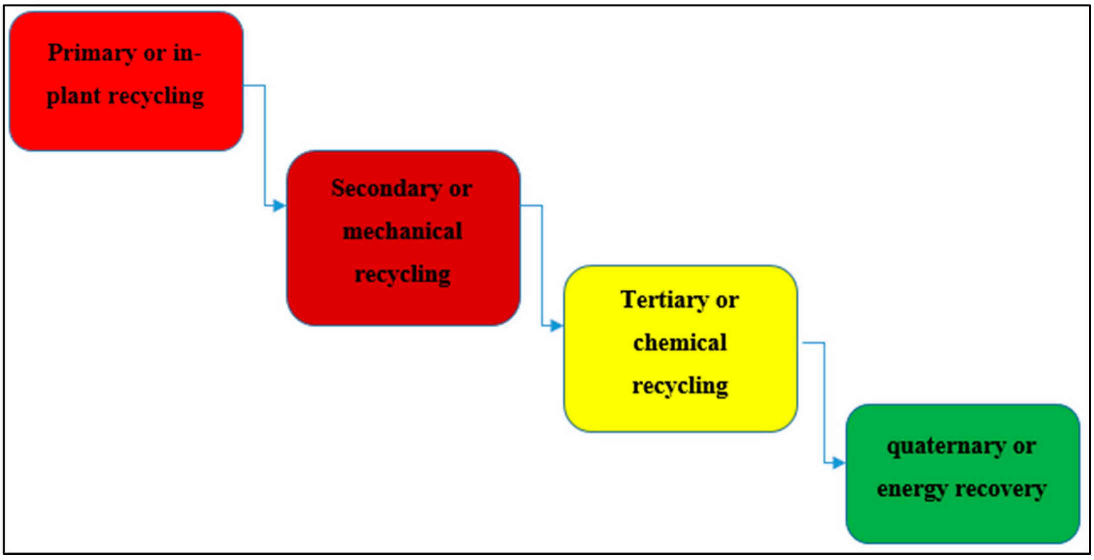


Figure 4. Sequential technique for thermoplastic recycling [8].

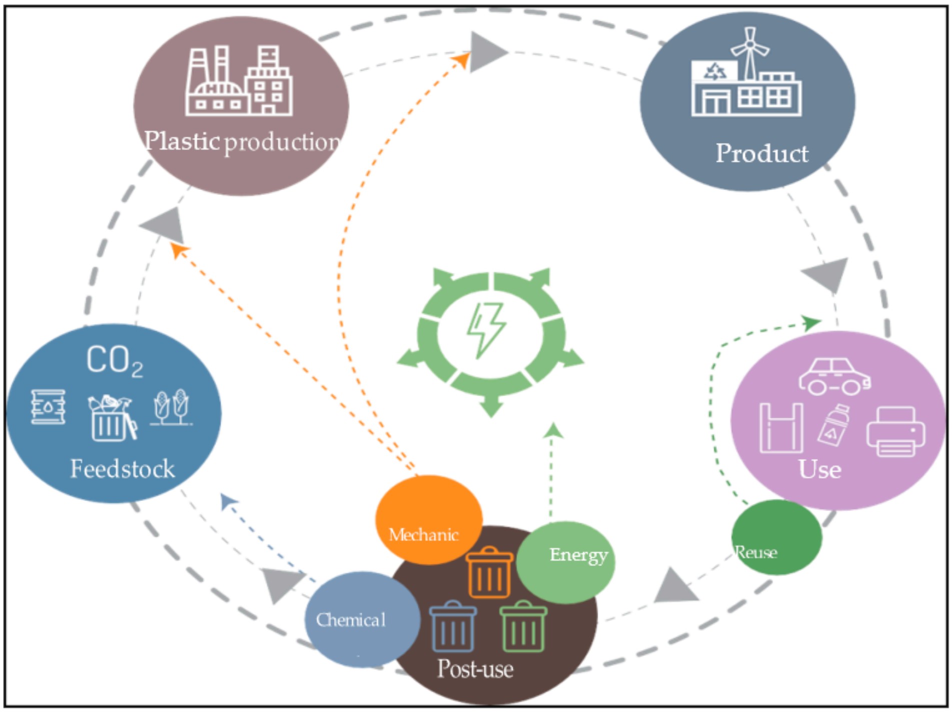


Figure 5. Life cycle of thermoplastic materials [41].

Current and Conventional Methods of Recycling

Photo-degradation, thermo-oxidative degradation, hydrolytic degradation, and biodegradation by microorganisms are the four mechanisms by which thermoplastic materials degrade in the environment. The primary route of thermoplastic material degradation, known as photo-degradation, usually arises naturally. Photo-degradation occurs from the emission of ultraviolet light rays, which draw their source from the sun and offer the activation energy required to introduce the intercalation of oxygen atoms into the polymer, in turn introducing the next route, called thermo-oxidation degradation. In the thermo-oxidation degradation route, the plastic materials tend to possess a brittle nature and begin to disintegrate or fracture into tiny pieces until the polymer chains attain a suitably low molecular weight to be metabolized with the aid of microorganisms [8]. It takes decades or years for microorganisms to convert the carbon molecules present in the long polymer chains into carbon dioxide or incorporate them into biomolecules. Thermoplastics degrade due to changes in their internal morphological structures, which are usually due to mutations in the meso-region caused by a variety of factors, including time [42]. Therefore, there is no doubt that the continued existence of thermoplastic materials after use frequently initiates environmental pollution. The most economical means of disposing of the thermoplastic waste are to recycle and reuse it as secondary materials [8].

Recycling is a technique of using waste plastic materials to fabricate a secondary product or recover energy. Energy recovery involves the use of waste plastics as fuel in controlled combustion, in which the high calorific value generated is exploited in the process. This process is not economically friendly and may generate poisonous residues, such as lead and cadmium. Additionally, it is important to know that recycling does not reduce the demand for new materials [38]. The terminologies used and techniques performed in the recycling of plastics are complex and sometimes confusing due to the broad series of recovery and recycling activities. Figure 6 shows piles of waste polymer-based materials that can be used as potential raw materials for secondary products, while Table 3 and Figure 7 present the four major methods and mechanisms of recycling, respectively.

Figure 6. Piles of waste plastics in Veolia, Germany [41].

Table 3. Four techniques for recycling [39].

ASTM D5033 Definitions	ASTM D5033 Definitions	Other Equivalent Terms
Primary recycling	Mechanical recycling	Closed-loop recycling
Secondary recycling	Mechanical recycling	Downgrading
Tertiary recycling	Chemical recycling	Feedstock recycling
Quaternary recycling	Energy recovery	Valorization

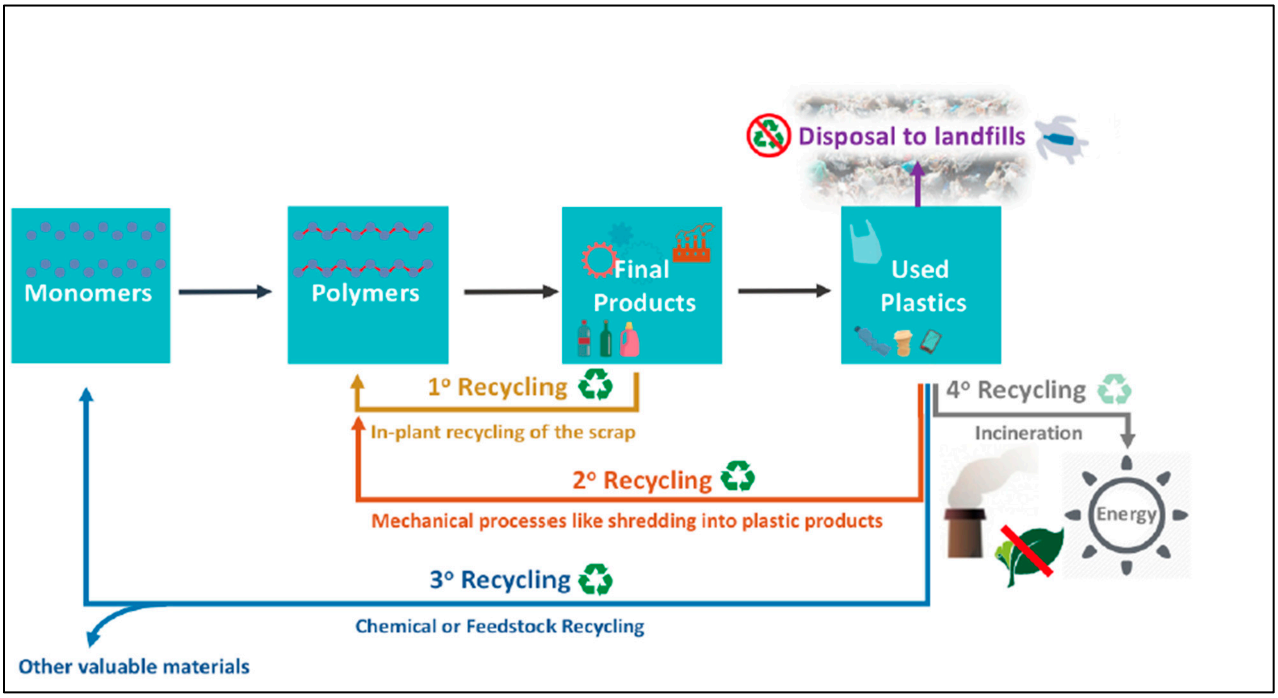


Figure 7. The four mechanisms of recycling [38].

The recycling of thermoplastic materials possesses great advantages that are beneficial to human life, such as the mitigation of environmental issues and the remediation of both materials and energy. The four recycling techniques are discussed briefly as follows:

i. Primary recycling: Primary recycling is a mechanical recycling process that involves the reprocessing of products in their natural state without any significant change to the materials' chemical structures. This method usually consists of the following steps: sorting, shredding, cleaning, processing, and milling [43]. Hence, it is a simple and economical process. This method of recycling is efficiently attained if the polymer components are: (i) efficiently alienated from the components that initiate pollution; and (ii) stabilized against degradation during reprocessing and subsequent reuse. Plastic materials that are not fit to recycle for a particular application can also be used as a starting material for the fabrication of a different plastic product (this process can also be considered primary recycling), so no plastic is wasted [8,39]. Mechanical recycling reuses waste material as the raw material for second-grade products or uses it as filler in composites [44,45]. This process is a closed-loop mechanical processing technique, as stated in Table 3.
ii. Secondary recycling: This process is also a mechanical recycling process in which continuous mechanical recycling could yield low-quality or substandard products, known as downcycling. This method is essentially performed on thermoplastic materials, which are easily re-melted and reprocessed for the development of novel plastic products. This process does not require the modification of the plastic during the recycling process; hence, the products are downgraded, as stated in Table 3. The stages are similar to those of primary recycling. Hence, the purity and quality of recycled polymer by mechanical processing are limited.
iii. Tertiary recycling: In tertiary recycling, which is also known as chemical recycling (Table 3), the polymer structure in the plastic material is chemically transformed into molecular monomers. However, in some cases, the plastic materials are partially depolymerized to oligomers by the chemical reaction catalysed by tertiary recycling, thereby resulting in a change in the chemical structure of the polymer. The resulting monomer is usually applied as the basis for the creation of new products [8]. However, chemical recycling requires a large amount of chemicals and is not possible for

all plastic types, thereby making this process uneconomical and detrimental to the ecosystem. The chemical reaction methods used for this recycling process include the following:

i. Hydrogenation;
ii. Glycolysis;
iii. Gasification;
iv. Hydrolysis;
v. Pyrolysis;
vi. Methanolysis;
vii. Alcoholysis;
viii. Aminolysis;
ix. Chemical depolymerisation;
x. Thermal cracking;
xi. Catalytic cracking and reforming;
xii. Photodegradation;
xiii. Ultrasound degradation;
xiv. Degradation in a microwave reactor.

iv. Quaternary recycling: This method involves the recovery of energy and heat content produced from the recycling of plastic materials (Table 3). In plastic recycling, incineration is said to be the most efficient way of reducing the volume of organic materials. Although it generates considerable energy from the polymer, it is not acceptable because of the health risks associated with the toxic substances generated during the incineration process [8]. Plastics that usually find their way to landfills can be used for energy production. They are used as feedstock for incineration plants that use plastics as fuel. The main drawback of this method is the release of considerable amounts of pollutants into the air [43].

Recycling is a viable solution for reducing the need for new raw materials and decreasing the materials discarded in landfills. Other beneficial effects of waste recycling are preventing environmental pollution from the extraction of raw materials, saving energy, and even providing domestic jobs. The life cycle of waste polymeric materials is shown in Figure 8.

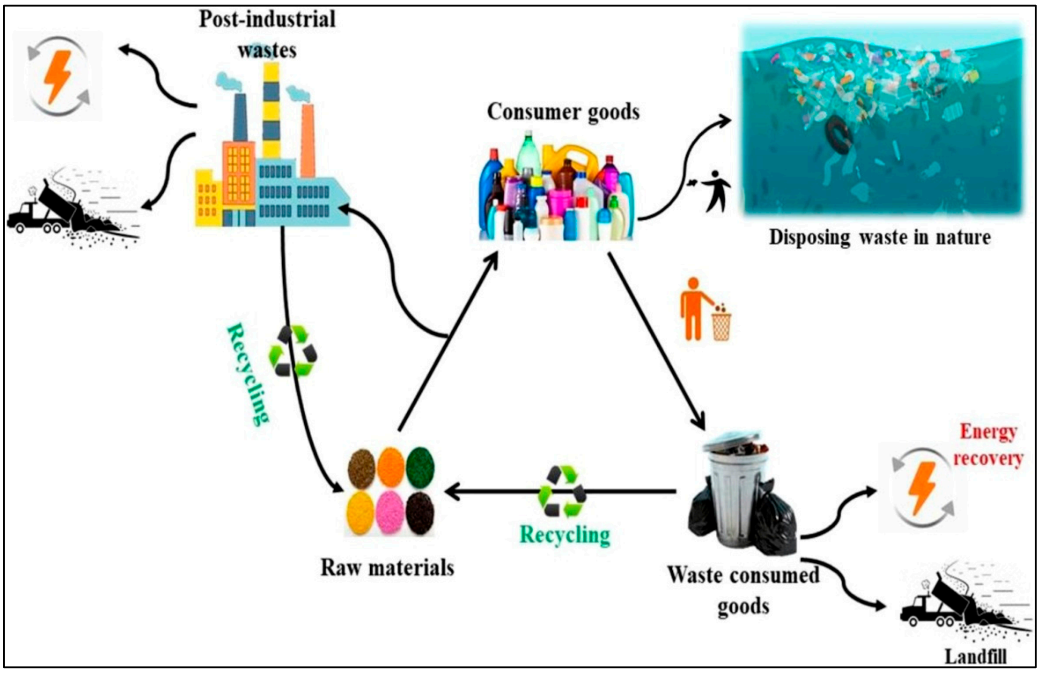


Figure 8. The life cycle of waste polymeric materials [46].

As noted regarding the three basic recycling methods (mechanical, chemical, and incineration), they are all limited by one factor or another, hence the need for an advanced method. Since the mechanical and incineration processes have simple and somewhat fixed steps, more attention can be focused on advances in the chemical recycling methods listed among the tertiary recycling methods due to the greater likelihood of modifying the process. Table 4 presents summaries of some recent developments from various chemical recycling methods to overcome these limitations [46].

Table 4. Summary of the chemical recycling processes for waste plastics [46].

Chemical Process	Main Degradative	Temperature (*C*)	Advantage	Disadvantage
Hydrolysis (alkaline)	NaOH, KOH	120–200	High purity	Requires chemical substances, longer times, and higher temperatures than the acidic method
Hydrolysis (acid)	concentrated sulphuric, nitric, or phosphoric acid	70–120	High purity	Requires a large amount of acid separation of ethylene glycol from acid, which is difficult
Hydrolysis (neutral)		200–300	Environmentally friendly	Low purity; requires high temperatures
Glycolysis	ethylene glycol, diethylene glycol, propylene glycol, butylene glycol, and dipropyleneglycol	180–250		Slow reaction in the absence of a catalyst
Methanolysis	zinc acetate	180–280		Low purity; requires high pressure and temperature
Alcoholysis	methanol, ethanol	180–250	CO_2 free	Requires high pressure; only applicable for plastics without dyes
Aminolysis	methylamine, ethylenediamine, ethanolamine, and butylamine	20–100	High purity; Applicable at low temperatures	Longer reaction time at low temperatures (10 to 85 days)

A comprehensive review of the state of the art about recycling thermosets and thermoplastics was performed by Kazemi et al. [46] with an emphasis on the application of recycled polymers in the construction of built environments. The study revealed amylolysis as the most promising method for thermoplastics, while irradiation recycling was found to be the most common method for thermosets. It was discovered from each group that there are innovative approaches, such as aminolysis and biological recycling, which have not yet been scaled up for market usage, hence the need to consider these methods. Among the major factors rendering one method more prevalent than others are processing costs, the quality of recycled products, ease of recycling, and amount and type of post-recycling waste. All of these factors led to the need to understand the science of polymer functionalization to be able to tune the recycled granules' surface properties for targeted applications.

Based on life cycle assessment studies, mechanical recycling has shown lower emissions than other approaches, making it the most suitable method for environments. However, the mechanical properties of recycled products are much more degraded than virgin plastics; hence, the revolutionary plastic mechanical recycling process was introduced using a new type of twin-screw extruder with the addition of a molten resin reservoir unit applied under various extrusion conditions. As a result, the mechanical properties of recycled products were recovered from their original materials. It was found that steady shear, which is a conventional process, affected the degradation of mechanical properties, while re-extrusion using a new type of twin-screw extrusion with a suitable processing condition is related to the regeneration of mechanical properties and lamellar structures similar to those of virgin plastics. Thus, this method introduces the recent advances in the plastic mechanical recycling process and proposes the future directions for plastic recycling technology [42].

4. Availability of Waste Plastic as Raw Material for Product Development

In 2020, more than 29 million metric tonnes of post-consumer plastic waste were collected, but more than 23% was discharged to landfills, and more than 40% was applied in energy recovery operations, while only approximately 34% was recycled [19]. Sources of waste plastics are industrial, commercial, and municipal waste. Industrial, or primary, waste is acquired primarily from the plastic processing, manufacturing, and packaging industries. Commercial waste is typically post-consumer waste from industries, such as packaging, building and construction, automobiles, electrical and electronics, household, leisure, sport, and agriculture, typically obtained from farms and gardens in rural areas. Municipal waste plastics are also obtained from residential areas, streets, parks, collection depots, and waste dumps [11]. Figure 9 shows the distribution of the percentages of plastic waste from different areas of application in Europe, where it was discovered that most of the plastic products are from packaging, as well as building and construction. Due to the large amount (41%) of plastic products used for packaging and the indiscriminate disposal of products after use, these products have been responsible for major problems in our environment. There is, therefore, a need to intensify efforts regarding how to handle plastic waste coming from packaging.

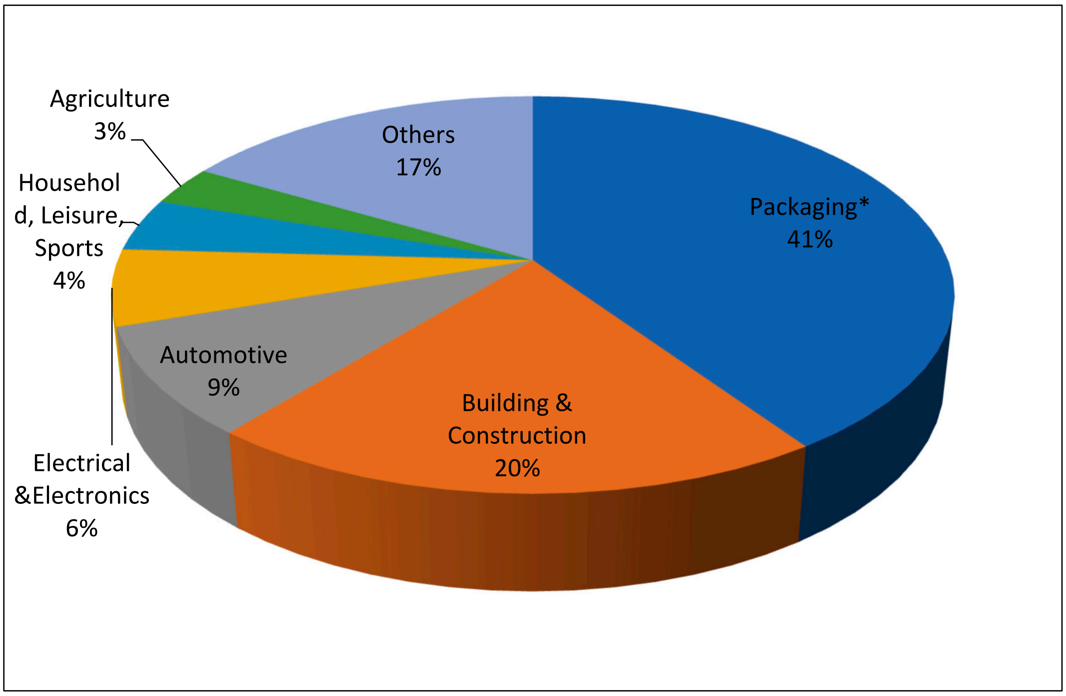


Figure 9. Percentage distribution of waste plastics in Europe [19]. * Including commercial and industrial packaging. Total 49.1 Mt.

Increasing consumption of plastic products in various fields concomitantly generates much plastic waste material. Thus, the predominant availability of plastic waste materials makes it a potential source of raw materials for future product development. In contrast to the previous opinion about waste plastics, these materials need to be researched and used for the development of suitable products for various applications. This research has become necessary due to their ready availability. In contrast to metals, waste plastics are beneficial materials to humanity because they serve as the leading low-cost and readily available materials with future benefits.

Benefits of Accelerating Waste Plastic Recycling

i. Environmental benefits: Recycling waste plastics reduces pollution and climate change by reducing the number of waste plastics that go into landfills or are released into the environment. According to reports, about four to 12 million metric tonnes of waste plastic are washed into rivers and end up in the ocean every year [41]. This process constitutes an environmental threat to marine inhabitants since it leads to depletion of the ecosystem. Additionally, recycling aids in the significant reduction in atmospheric emissions of CO_2 because recycled plastics do not generate the emissions that are generated during the production of virgin plastics.

ii Economic and social benefits: Recycling waste plastic generates employment opportunities and value creation by fostering the local growth of recycling plants. Establishing recycling plants promotes local industrial activities in the recovery of value from recycled materials [41].

iii. Availability of raw materials: Recycled plastics are potential materials for secondary products. Processed plastic waste can be suitably adapted for appropriate applications in various fields.

5. Current Applications of Thermoplastic Waste

Due to the exponential surge in the accumulation of pervasive and persistent thermoplastic waste, which has increased in landfills and affected the environment, scientists and researchers have focused on innovative and sustainable means to recycle/reuse waste plastics to reduce the impact on the environment. Waste plastics are emerging as a viable option in some sectors, such as in construction, energy, domestic goods, automobiles, fabric/clothing, and many more [7,15,47].

5.1. Utilization of Thermoplastic Wastes in the Construction of Bricks, Tiles, and Blocks

Waste plastics have been globally utilized in construction. Recently, polyethylene terephthalate (PET) as scrapped plastic waste (SPW) and foundry sand (FS) was used for the production of green bricks for construction applications. It was observed that bricks from 70:30 of FS:SPW yielded optimum compressive strengths of 38.14 MPa and tensile strengths of 9.51 MPa [48]. In another report, PET plastic waste (PPW) and recycled crushed glass (RCG) were used at different ratios of 80:20, 70:30, and 60:40 of RCG. It was noted that PPW increased the tensile and compressive strengths of the base materials by 70.15% and 54.85%, respectively. These findings revealed better properties compared to the mechanical capacity of conventional ceramic bricks. The average compressive strength and tensile strength obtained were 42.01 MPa and 9.89 MPa, respectively, while the average water absorption value was only 2.7%. Due to their highly hydrophobic properties, neither type of masonry brick made from foundry sand and crushed glass requires water for construction, they are both more resistant to chemical attack, and they are less deformable under strain stress than burnt clay bricks" [49].

Additionally, in the construction of bricks, Mondal et al. [50] used some thermoplastic waste, including polycarbonates (PC), polystyrene (PS), and mixed plastic. Sand, ash, and regular Portland cement were also added in varying proportions. It was recorded that the resulting bricks were porous in nature, light in weight, and thermally resistant. They possessed a compressive strength of about 17 MPa and a maximum water absorption value of 14.18%, and the bulk density decreased from 2.06 to 1.60 g/cm^3 with the addition of waste thermoplastics.

Currently, thermoplastic waste has also found great applications in the manufacturing of different types of marble floors. The production of roofing marbles with recycled high-density polyethylene (rHDPE) as starting material alongside foundry sand has also been investigated by a few researchers, and conclusions were drawn regarding its suitability for industrial applications [50]. Additionally, the application of plastic waste and disposed glass waste was further reported for the production of roof tiles, hollow blocks, and marbles. However, in a few research works, cement was replaced with plastic waste, whereas broken glass replaced river sand, partially yielding stronger and more durable construction materials [49,50].

5.2. Utilization of Thermoplastic Wastes in Concrete and Road Construction

Waste thermoplastics have been employed as a partial or total substitute for concrete in road construction. Different admixtures have been investigated and have been applied in various aspects of global technological development. Recently, technological research reported that recycled plastic-bounded concrete (RPBC) was developed from 100% waste plastic with the exclusion of binder or Portland cement. Additionally, recycled high-density polyethylene (rHDPE) materials and recycled polypropylene (rPP) were applied in research works. The mechanical properties, crack recovery, and thermal and moisture sensitivity of the recycled thermoplastic-bounded concrete were further observed as follows. The compressive strength of recycled polypropylene-bounded concrete was 30 MPa, which was almost three times that of asphalt binder concrete. Recycled PP had three times the bending strength of plain cement concrete (PCC) and five times the bending strength of asphalt concrete (AC). The crack healing performance of RPBCs was approximately 92%. RPBC showed greater resistance to moisture exposure. The strength of recycled PP decreased by 5%, while the strength of asphalt concrete decreased by 17%. The bending power of ACs was only 10%, but the strength of recycled HDPE- and recycled PP-bounded concrete was 85% and 99%, respectively [47]. Additionally, the possibility of using waste polypropylene (PP) plastic as a replacement for bituminous road concrete in pavement construction was investigated, and it was discovered that PP could enhance the performance of asphalt concrete and reduce the cost of road construction [51]. Correspondingly, waste PET was also applied as a plastic aggregate in concrete production. Here, conventional coarse aggregates were substituted with waste thermoplastic materials in quantities of 5%, 10%, and 20% by volume [52]. Thus, substituting waste plastic for cement is more economical and environmentally friendly because the production and development of cement release a very large quantity of carbon dioxide (CO_2) and require a sizeable amount of oil [53].

The use of waste polymeric materials in construction has many advantages apart from being cost-effective, but it still poses some challenges. Waste polymers need to be processed to meet construction requirements. There should be regulations and standards for using these waste polymers. To compensate for the shortcomings of individual types of waste polymers as construction materials, treating the waste materials or combining them to enhance their applicability and compatibility should also be encouraged. Hence, more efforts need to focus on the upcycling of waste thermoplastics and thermosets to support the most recent advances in improving the affinity of polymeric solid waste as construction material [46].

5.3. Utilization of Thermoplastics Waste in the Production of Fuel

An essential application of waste thermoplastics is their valorization into fuels. The process consists of transforming polyolefin into essential products, such as fuels, naphtha, and polymers. The major mechanisms carried out include:

(i) Gasification of waste thermoplastics, which involves the production of gaseous streams for energy or synthesis;
(ii) Pyrolysis for H_2 and pyrolysis for specific purposes;
(iii) Integration of waste plastics into refinery units [54].

Pyrolysis is a more economical process and has been adopted by many researchers because it yields a large volume of liquid oil, up to 80 wt.% at a moderate temperature of 500 °C. The derived liquid oil has found its application in major industries that use high-efficiency machines, such as diesel engines, boilers, and furnaces, without upgrading [55].

5.4. Utilization of Waste Thermoplastics in the Production of Commercial Products

The commercial use of thermoplastic materials for the manufacture of bottles and other goods has become ingrained into human existence. The use of thermoplastic products has consequently increased gradually, resulting in an imbalance between consumption and production. This imbalance has necessitated the reuse of waste thermoplastics to meet the demand for the manufacture of commercial products [7]. Since recycling methods, such as high value-added recycling and horizontal recycling, reduce or totally eliminate contamination while producing high-value products with intrinsic viscosity, good colour separation, and reliable batch-to-batch quality, they are highly sought [56]. Some common contaminants, such as detergents, fuel, pesticides, and stored concentrated chemicals, can be dangerous to human health if not properly treated during the recycling process. Since contamination usually causes the physical and chemical properties of the thermoplastic to deteriorate, its reduction concurrently results in a considerable improvement in the quality of recycled waste thermoplastics [56]. Some household and commercial areas in which waste thermoplastics have been applied are summarized in Table 3, with more commercial thermoplastics and products expected to continue to emerge from derived plastics rather than the existing ones. It can be seen in Table 5 that there is no human being who does not u se polymer-based products daily; hence, polymers remain an indispensable material for human existence [36]. Polymer products were detected to be the dominant materials in our household materials, whether as consumables or non-consumables.

Table 5. Commercial applications of recycled thermoplastics [8].

Thermoplastic	Product Identification Code (SPI)	Applications
HDPE	HDPE	Detergent bottles, mobile components, agricultural pipes, compost bins, pallets, toys
LDPE	LDPE	Bottles, plastic tubes, food packaging
PET	PETE	Drink bottles, detergent bottles, clear film for packaging, carpet fibres
PP	PP	Compost bins, kerbside recycling crates
PS	PS	Disposable cutlery
PVC	V	Packaging for food, textiles, medical materials, and drink bottles.
Others		Containers

6. Future Prospects of Waste Thermoplastics and Thermosetting Plastics

This section provides detailed information about what the future outlook will be with reference to waste polymers, recycling, and applications. Presently, researchers have developed an interest in converting these enormous waste resources into useful products for several applications. Hence, it is envisaged that recycled polymers will not only be used for commodity products but will also be used for advanced materials applications due to their availability as raw materials. However, it is expected that the use of waste polymers for various applications should be classified since the materials have become established now.

6.1. Waste Thermoplastics

Recently, waste thermoplastics have been more of a benefit than a threat. Current research trends have shown that waste thermoplastics are potential materials for secondary production and products. They are possible materials for biodegradable products that can have a positive environmental impact when processed, used, and disposed of appropriately [42]. They can also find suitable applications in building and construction, electronics, sporting materials, energy and power generation, automobiles, and many more fields, in addition to those stated as current applications in this review. Their inherent advantages of ease of processing and global availability enable their continuous reuse. Waste thermoplastics are the leading universally available raw materials worldwide as waste products, with similar compositions or constituents. However, due to varying environmental influences, slight changes may be noted in their properties over time. As a result, more advanced research on the environmental impact and type of thermoplastic waste generated is required. The world is confronting a crisis due to the large amount of waste plastic generated globally; therefore, there is a need for increasing attempts to overcome this challenge due to environmental and economic concerns [3]. Therefore, there is a need to develop new eco-friendly recycling technologies, reduce energy consumption, and decrease or completely eradicate the harm associated with waste plastics in the environment. Researchers and scientists are devising more promising methods and other eco-friendly options that will be more sustainable and economically friendly. More studies are to be conducted, including carbon capture using waste plastic sources, the synthesis of carbon microspheres and nanotubes from plastic waste, natural-based monomers for commodity plastics [36], biodegradation, and composting opportunities [8]. These recycling options will mitigate environmental challenges and enhance the application of thermoplastic products [3].

6.2. Waste Thermosetting Plastics

Until now, waste from thermosets and thermoset-based composites have been considered a threat to the environment. They are usually referred to as a class of materials that cannot be recycled because they have a cross-linked structure. However, this condition has persisted due to a lack of possible processing technology for their adaptation to new products. Hence, these materials can be processed by crushing them into different shapes or powders of varying grades, and they can used as reinforcement materials in thermoplastics, concrete, and bricks. They can also be applied as partial replacements in road construction and other allied areas of applications in which hard and brittle materials are needed with suitable binders. Common examples of these materials are polyester, silicone, melamine, urea formaldehyde, epoxy, and polyurethane. Waste foam from polyurethane can be suitably used as filler, while other materials can be appropriately used for common products and applications using plastics that are needed in construction equipment panels, electrical housings and components, insulators, kitchen appliances, toys, circuit breakers, agriculture, automotive parts, and signage.

6.3. Biological Recycling

Biological recycling, or bio-recycling, is an emerging technology that uses microbes, such as bacteria or fungi, to break down plastic into its basic components for reuse is a process in which the microorganisms attack waste polymer materials, break them down, and make new materials from them using biological processes, such as anaerobic digestion or photosynthesis. Due to the need for more biodegradable polymers in recent years, more products are now being developed from biodegradable sources, and the evolving waste needs to be recycled. Hence, biological recycling, which applies mainly to biodegradable plastics, has recently attracted more consideration from researchers. The process has been classified as a form of tertiary recycling which is also referred to as organic recycling [39]. Biodegradable plastics can find successful applications in many other areas after use [57]. However, if there is not adequate awareness, users may be misinformed and carelessly dispose of biodegradable packaging material in the environment, thus increasing polymer pollution. Although some biodegradable or compostable polymers have grave impacts on the environment, as do non-biodegradable petroleum-based polymers [58], these types of polymers need to be properly identified for easy sorting before processing. A mixture of biodegradable polymers with non-degradable polymers will pose further sorting and waste management problems. Hence, there is a need for adequate information from manufacturers globally in resolving the disposal problem. For example, polylactic acid polymer has been widely believed to easily biodegrade in landfills or home composts or even in aquatic environments. This relief is not true, as the polymer does require an industrial composter and, as such, should be referred to as compostable [59–62].

It was reported by Kosior and Mitchell [63] that bio-based products may not be effectively handled by current end-of-life waste management options. Only drop-in bioplastics, which can be used with current technologies, are seen as ideal, and these materials include bio-PE, bio-PP, and bio-PET. However, other bio-based polymers such as polylactic acid can be used alone or with organic fillers to create compostable food packaging composites, presenting opportunities for green packaging materials that can be biologically recycled. Compost bins may be used to allow for home composting of biodegradable food packaging waste [64].

Some natural and designed microbes were reported by Drzyzga and Prieto [65] to show potential for possible application to biodegrade problematic petroleum-based plastics. Another new field of study reported is the use of enzymes engineered for plastic degradation. Studies of PET, LDPE, and linear-low-density polyethylene (LLDPE) have been performed. Further research is envisioned in the biological recycling field to employ microbes, fungi, and enzymes in the degradation of plastics.

6.4. Reduction in Materials

One of the new innovations in recent times is the reduction principle, which is aimed at lowering the material quantities used in products and processes [66]. It can also be referred to as down-gauging whereby the dimensions of polymer-based materials are altered to reduce and save the materials used [39]. For example, in packaging, this process translates into the reduction in the amount of material used to create a package but still maintaining the optimum function of the packaging. Material reduction principles can result in packaging products with considerably lower environmental impacts. Packaging reduction should be carefully undertaken to avoid compromising the overall product system [67]. Coca-Cola has managed to reduce the material used in making bottles and is now using redesigned smaller bottles with shorter necks [58]. The target is to eventually make 100% recyclable packaging by 2025 [68]. In addition, their plastic usage comprises up to 10% recycled plastics [58], and this figure is projected to rise to 50% by 2030 [68]. In addition, the reduction in the use of non-biodegradable materials and the uptake of bio-based biodegradable polymers for food packaging plastic material are viable options with much research interest and potential to yield a sustainable circular economy in food packaging [69–75].

7. Conclusions

Mechanical recover, chemical recovery, and energy recovery, which have low-grade, expensive, and environmental pollution consequences, respectively, are the three major methods of recycling waste polymers currently in use. This review revealed the current drawbacks related to these approaches. Additionally, it was suggested that mechanical and chemical methods could be enhanced to overcome some of these drawbacks. Furthermore, to produce a high-quality product, it was further established that cleaning and sorting are two fundamental phases that must be given essential attention in all waste polymer-recycling methods. More efforts are expected to be focused on the cost-effective and environmentally sustainable mode of processing waste polymers, as suggested in the review, due to the recent interest in utilizing these waste materials.

The concomitant surge in waste polymers due to their lack of degradability has been perceived as a serious environmental challenge to our ecosystem. However, these waste thermoplastics and thermosetting plastics can be strategically mitigated by recycling them. It was proven in this review that waste polymers can be completely reused, thereby advancing the drive for net zero waste in our environments, particularly from polymers and polymer-based composite products. The review methodically investigates the prospect of waste polymers, including their availability as raw materials for product development, the basic recycling techniques being used, their limitations and ways forward, current waste polymer applications, and prospects for waste thermoplastics and thermosetting plastics. Thus, waste thermoplastics and thermosetting plastics should be appreciated as inexpensive and universally available raw materials for product development. Recycled waste thermoplastics and thermosetting plastics should be investigated for additional potential applications as packaging materials, which are currently the leading source of pollution on land and sea. The blend of these materials should be considered for other areas in which recycled waste thermoplastics and thermosetting plastics might not even be applicable for now. The possibility of using recycled thermoplastics and thermosetting plastics in biomedicine and nanotechnology should be intensified.

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