



**Sustainable Recycling Technologies for Thermoplastic
Polymers and Their Composites: A Review of the State of
the Art**

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Sustainable Recycling Technologies for Thermoplastic Polymers and Their Composites: A Review of the State of the Art

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Abstract

This review article discusses the environmental and economic effects of recycling, as well as sustainable thermoplastic polymer recycling technologies. Several researchers have utilized recycled thermoplastics as matrices in the production of a variety of natural and synthetic-based composites, which is also the focus of this study. All of the industries (food and packaging, construction and building, transportation, and indoor usage) where recycled thermoplastics have a large market share (food and packaging, construction and building, transportation, and indoor usage) are covered in this review. The desirable properties of thermoplastic polymers, such as corrosion resistance, low density, and user-friendliness, have caused plastic production to surpass aluminium and other metals in use over the past 60 years. Furthermore, recycling is one of the most important measures available to mitigate these effects and is one of the most dynamic segments of the plastics industry at present. Increased landfilling and incineration of plastics have a negative impact on the ecosystem, and the continued increase in the production of virgin fossil plastic also has a negative impact on the environment. Consequently, this continuous production could lead to the depletion of fossil fuel resources, an increase in environmental emissions during processing, and eventual incineration. Increasing numbers of nations are adopting the circular economy concept in an effort to avoid all of these problems. This concept emphasizes the reuse of products and resources, as well as the recycling of materials according to the waste hierarchy, rather than their cremation or disposal in the environment.

Keywords: Recycling, Thermoplastics, Recycling technologies, Plastic waste, Depolymerization, Environmental impact, Circular economy.

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11 **1.0 Introduction**

13 Polymeric materials are becoming increasingly popular in a variety of manufacturing industries
14 due to their superior mechanical, barrier, and thermal properties, which result in longer component
15 durability. The biodegradability and recyclability of some polymers has piqued the interest of
16 industrial and academic researchers in developing environmentally sustainable materials for the
17 packaging, construction, and food industries [1,2]. Polymer production is always linked to future
18 challenges regarding material utilization after use. Because of slower developments in recyclable
19 processes, ecological and social issues have arisen during the annual disposal of one million tonnes
20 of polymeric materials. Furthermore, unsustainable recycling processes resulted in the omission
21 of certain types of polymers from the economic cycle [3,4]. Thermoplastics, for example, were not
22 recycled and thermoset polymers have been popular polymeric materials in composite industries
23 due to the requirement for potential alternatives in place of metallic materials. The thermoset
24 polymers are synthesized through irreversible polymerization and are cured with the application of
25 heat or through chemical reactions, but they eventually become insoluble and infusible materials.
26 The recycling of these materials is difficult due to cross-linked structures where the chemical
27 bonds are stronger than the thermoplastic materials [5–7]. The thermoplastic polymer materials,
28 on the other hand, are made up of linear molecular chains that soften when heated and harden when
29 cooled. The recyclable nature of these polymers is good due to the non-degradation of polymer
30 chains after the application of heat. There are three types of thermoplastics that can be recycled
31 easily using different mechanical, thermal, and chemical processes [8,9]. The first category is
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3 crystalline thermoplastic materials. Usually, these are transparent with molecular chains and
4 exhibit regular arrangements. These polymers possess higher impact resistance, and the examples
5 are HDPE (high-density polyethylene), PP (polypropylene), and LDPE (low-density
6 polyethylene). Random arrangements of molecules are common in amorphous thermoplastics, and
7 examples are ABS (acrylonitrile butadiene styrene), PVC (polyvinyl chloride), and PC
8 (polycarbonate), PS (polystyrene), and PMMA (polymethyl methacrylate) [10,11]. The third
9 category of thermoplastics is semi-crystalline thermoplastics, where the properties lie between the
10 above two categories, and the examples are PAI (polyamide-imide) and PBT (polybutylene
11 terephthalate). In the current scenario, the recycling cost exhibited by the polymers is low, and
12 hence the huge amounts of utilized synthetic textiles and plastic recycling value fall below the
13 economic value. In comparison to ceramics and metals, polymers lose more of their original
14 properties after recycling. However, there is a way to improve the properties of the products that
15 are made with major changes [12,13].
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33 The production of waste polymers from the household, industrial, and agricultural activities
34 is increasing day by day. It was reported that more than 12% of solid municipal waste was made
35 up of plastic in recent days, whereas 1% of the plastic waste stream was recorded in the year 1960
36 [14,15]. In recent years, it has been difficult to effectively treat polymer waste. Traditional disposal
37 methods, such as landfilling, burying, or combustion, harmed the environment by releasing toxic
38 gases, dust particles, fumes, and pollution into underground water. To save the environment,
39 traditional techniques are no longer used, and new recycling methods are preferred to manage
40 polymer waste. The different polymer waste management strategies in the USA, Europe, and LAC
41 (Latin America and the Caribbean) are illustrated in **Figure 1**, which indicates that the sanitary
42 landfill was the most commonly employed strategy in all three countries. In polymer recycling,
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popular recycling processes such as mechanical grinding, pyrolysis, solvolysis, and other thermal processes were used and reported in the literature [16,17]. The incineration and recycling are the main recovery aspects in thermoplastic polymeric materials. The incineration process results in the production of cadmium and leads through the ash contents after the burning. The recycling of thermoplastic polymers involves four steps, viz., collection, separation, manufacturing, and marketing. These simple recycling steps reduce manufacturing costs and facilitate mass production [18,19].

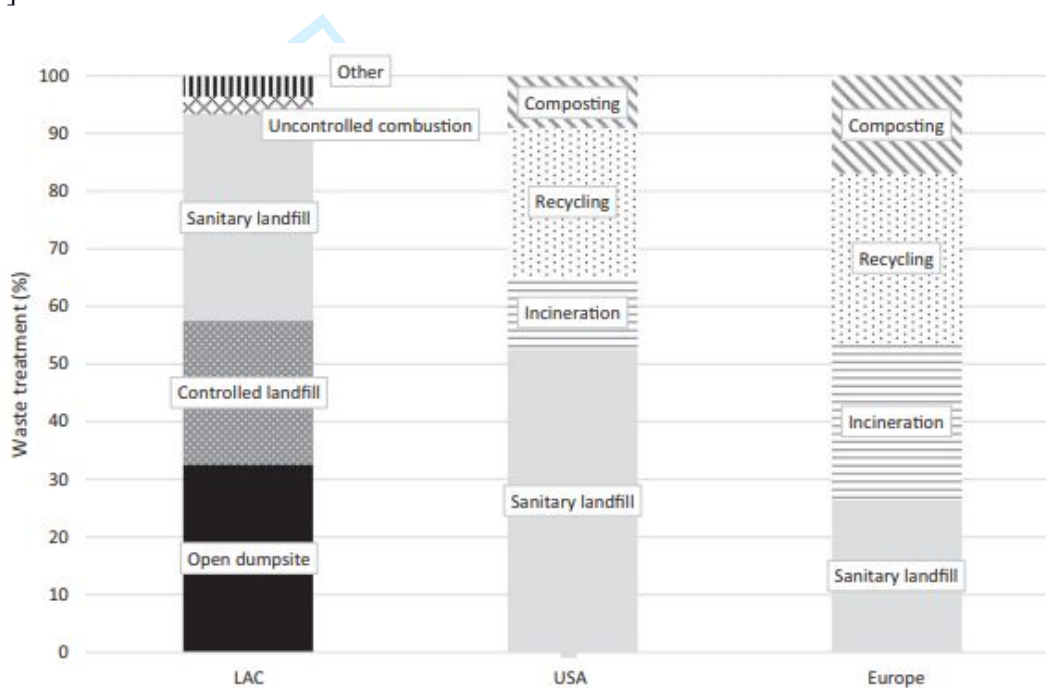


Figure 1. The waste management strategies adopted in LAC, USA and Europe [16].

[<https://doi.org/10.1016/j.scitotenv.2019.06.393>]

The different polymers with their applications, recycling codes, and municipal waste percentage are illustrated in **Figure 2**. The maximum municipal waste was from HDPE material due to regular usage by human beings. The degradation of plastics in the environment was observed through four mechanisms, such as hydrolytic degradation, biodegradation through microorganisms, photodegradation, and thermos-oxidative degradation. When the polymers are

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3 exposed to the environment, the process begins with photodegradation through the absorption of
4 UV light from nature, which provides the required energy for oxygen atom incorporation and
5 causes thermo-oxidative degradation. In this degradation step, the polymer materials will become
6 brittle and fractured into small pieces till they reach the suitability weight required to be
7 metabolized by the microorganisms. Furthermore, these microorganisms convert the carbons in
8 the chains into carbon dioxide (CO₂), or they may be included in the biomolecules. This process
9 will take a minimum of 50 years, and hence recycling is the best solution for polymeric materials
10 [20–22]. The term "sustainability" refers to the maximum depletion of nonrenewable materials and
11 energy usage in products, as well as recycling optimization. "Sustainable technologies" refer to
12 recycling technologies that use little or no energy and have no negative environmental
13 consequences [23,24]. The suitable selection of recycling techniques depends on various criteria
14 such as the types of additives added to the polymers, polymer stream purity, and the chemical
15 constituents present in the polymers. Even with the different recycling technologies, problems still
16 exist at the end of the recycling stage before reaching the product consumers. These problems are
17 raised due to the degradation of polymers during processing and also incompatibility while using
18 various types of fibers. The solution to avoid these problems is the loading of additives into the
19 polymers during the processing stage; this also enhances the characteristics of the recycled
20 components [25,26]. The term "additives" is defined as organic or inorganic substances that are
21 capable of altering one or more characteristics of the polymer materials. These essential additives
22 provide modifications and maintenance to the properties of polymers for long-term usage. The
23 categorization of these additives is based on many principal objectives, such as preferred in volume
24 or bulk for economic benefits and controlling properties, and also the modification of intrinsic
25 physical or chemical properties for recycled or core polymer materials. Moreover, the mechanical
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3 or structural properties of the polymers are also altered when they are used as reinforcements
4 [27,28]. In comparison among recycling methods, chemical recycling helps in reaching economic
5 targets and producing high-quality fibers as compared to mechanical recycling. The tolerance level
6 is higher in chemical processes for contaminated and mixed plastic waste than in mechanical
7 processes. In a chemical process, the polymer materials are broken down into small monomers for
8 the production of high-quality products. Temperature considerations are very important when
9 processing mixed plastic waste because of the various melting ranges of different polymers. Hence,
10 the preferred temperature will be the maximum melting temperature of the polymer, and other
11 polymers with low melting points will degrade easily during processing because of overheating.
12 The higher operating temperature results in more energy requirements and processing costs. The
13 technology's sensitivity towards waste contamination requires more effort around logistics [29,30].
14 This review work connects the economic and environmental aspects of recycling, the impact of
15 additives on polymer recycling, and the various technologies involved in the polymer recycling
16 process, recent research work, and applications of recycled polymer products with the challenges
17 report based on the review work.
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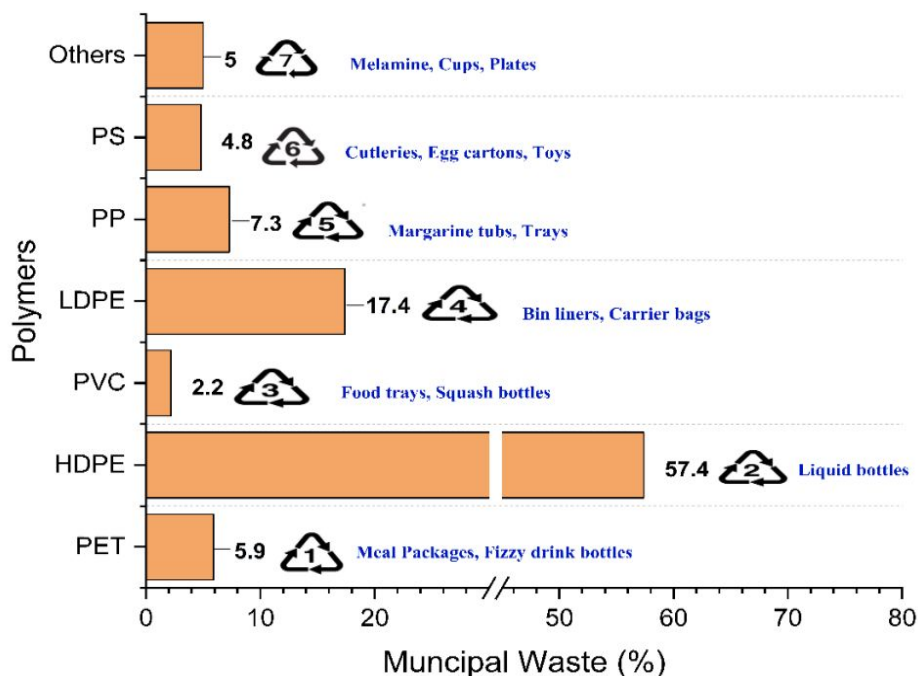


Figure 2. Different polymers with their applications, recycle code and municipal waste percentage [20,31].

2.0 Environmental and Economic aspects of Recycling

In 2018, global plastic production was estimated to be 359 million tonnes, with recycled plastic accounting for only 9 percent of that total. The remainder of the plastic waste was either landfilled (79 percent) or incinerated (12 percent) [32]. Hence more percentage of landfilling effects negatively to the ecosystem, and the continuous increment in virgin fossil plastics production lead to environmental impact as well. Hence this continuous production may cause depletion of fossil sources, more emissions toward the environment during processing, and be subjected to incineration at the end of life. In order to avoid all these problems, many countries are currently moving towards a circular economy concept. This circular economy involves the reusing of products or materials and recycling of materials as per waste hierarchy in place of incineration or disposal into the environment. Some of the interesting facts regarding CO₂ emissions to the

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3 environment during energy recovery from incineration and recycling of different polymers in
4 Europe are illustrated in **Figure 3**. This graph indicates that a reduction of 200 Mtonnes, or 73%
5 of CO₂ emissions, is observed in recycling as compared to energy recovery from the incineration
6 process [33,34]. In previous decades, industries followed the "linear economic model" with a
7 concern for the useful lives of plastic products. This model is dependent on the working principle
8 of "take, make, and dispose of". The linear model relied heavily on two assumptions: one is that
9 fossil resources are infinite materials, and the other is that waste products can be reclaimed and
10 recovered after they have served their purpose. During that period, the industry's focus was on
11 production efficiency improvement and the design and quality of the products that were reliable
12 for the customers [35,36]. Waste management was included in the linear economy model, so the
13 plastic waste was not disposed of properly and filled the environment. Moreover, the complex
14 design of the products causes many losses during recycling loops. In recent years, the awareness
15 of the environment at legislative and social levels has resulted in the coining of the "Circular
16 Economy Model". This model reports the efficient and effective ways to recycle plastic waste after
17 the shelf life of the product. This model also reports the economic benefits from consumers to the
18 production industries [37,38].
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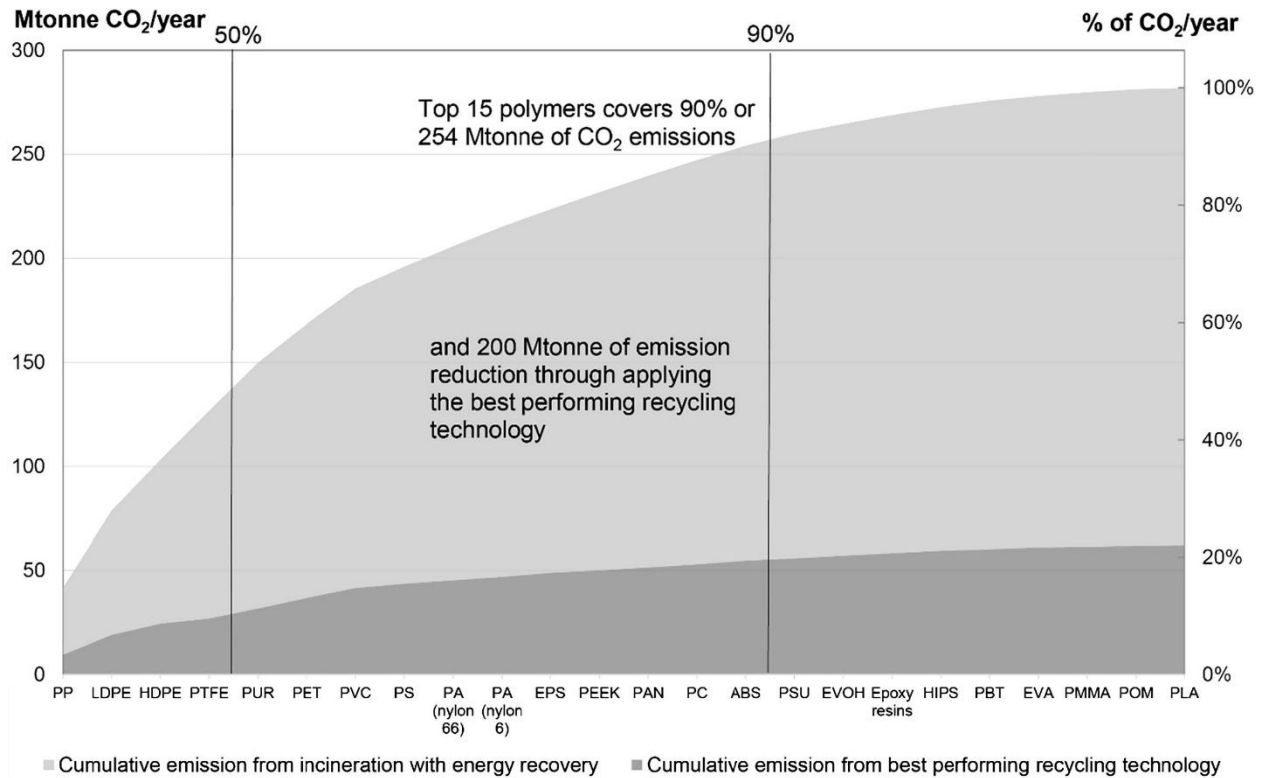


Figure 3. CO₂ emissions from recycling technology and incineration process in Europe for the year 2018 [34]. [<https://doi.org/10.1016/j.wasman.2020.12.020>]

3.0 Sustainable Technologies

3.1 Mechanical Recycling

Mechanical recycling entails the direct reuse of unsoiled polymers and their composites via granulating, squashing, grinding, and finally milling. The step-by-step mechanical recycling process and its description are illustrated in **Figure 4**. This milled polymer waste is again melted and subjected to the extrusion process without altering the material properties. To get additional potential, sometimes these recycled materials are mixed with virgin materials during the production process. Before the beginning of mechanical recycling, waste materials needed to be sorted manually in order to remove unwanted materials, followed by a cleaning and drying process [39,40]. Broken materials integration and additive loading are easier in this process. This process

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3 is limited to only thermoplastic materials such as PE, PP, PET, and PVC. The process of
4 mechanical recycling is inexpensive, but it needs more initial investment for the machinery. The
5 shaft shredding equipment is relatively preferred to cut thermoplastics into uniform sizes, and the
6 cut dimensions can be easily controlled by changing the blade distance inside the equipment. The
7 schematic illustration of the mechanical shredder with top, front, and inside views with reference
8 materials is illustrated in **Figure 5**. The major disadvantage is the lowering of polymer molecular
9 weight due to broken chain links under an acid and moisture environment. Hence this may cause
10 a reduction in the mechanical properties of the final products. This recycling process is sustainable
11 and most effective with parameters like time, carbon footprint, impact on the environment, and
12 cost management. The other disadvantages of this process are the non-homogeneous nature of
13 mixed plastic waste and slight property variations in every cycle due to the reduction of molecular
14 weight [41–43].
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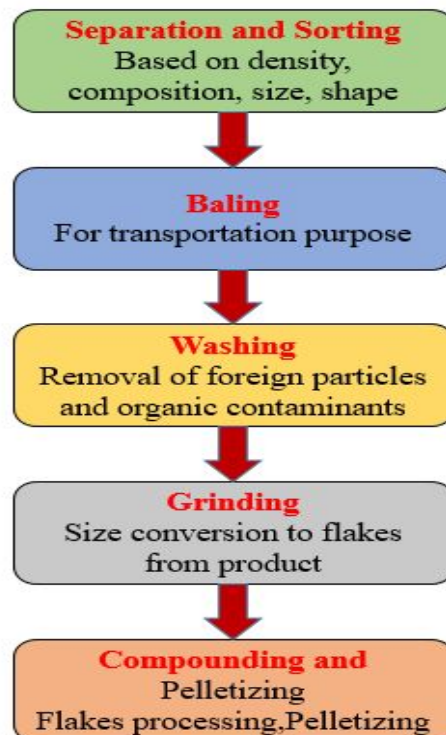
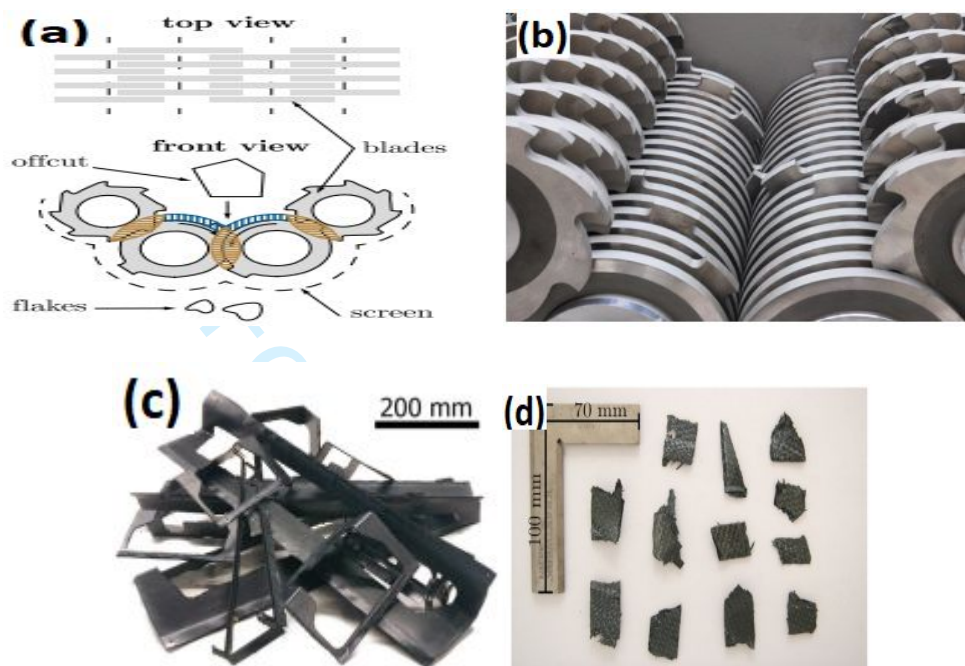


Figure 4. Mechanical recycling flow chart [29,39].

Colucci et al. [44] studied the microstructure and mechanical behavior of mechanically recycled PA66 thermoplastic reinforced short carbon fiber composites. The study reveals that the recycling of aged PA66 has no significant effect on the tensile strength and Young's modulus of the composites. However, the reduction of fiber length during processing has shown a slight reduction in mechanical properties. The microstructural images reveal the inhomogeneous distribution and alignment of carbon fibers along the material flow direction. The mechanical recycling process majorly affects the recycling of polymer waste with different polymer combinations. The different stable natures of polymers influence the resultant properties when the temperature exceeds the melting temperature [45]. Another study revealed that there were no significant changes in the polymer properties after injection molding with polypropylene, glass, and thermotropic liquid crystalline polymers. The temperature requirement for mechanical recycling was determined using thermogravimetric and rheological studies to avoid the degradation of polymers [46]. Mengeloglu and Karakus [47] analyzed the thermal degradation and mechanical properties of wheat straw flour reinforced with recycled PP and HDPE thermoplastics. The inclusion of the MAPE coupling agent enhanced the properties of recycled thermoplastic composites, but the enhancement of properties was more in PP-based composites than in HDPE-reinforced composites. As per ASTM criteria, these recycled plastics have reached the mechanical and thermal properties required for plastic lumber decking board applications. There are different challenges in mechanical recycling for both single and mixed plastic waste. The major issue with these polymers is degradation at a certain condition. These conditions could be related to mechanical shear, heat, ionic reaction, or hydrolysis. Involved in two types of major degradation: thermo-mechanical degradation during reprocessing and degradation over a lifetime. The high

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3 impact degradation is thermo-mechanical degradation, where the degradation occurs due to the
4 application of mechanical shear and heat. Polyimide (PI) and Polycarbonate (PC) thermoplastics
5 majorly undergo this type of degradation [48,49].
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32 **Figure 5.** Schematic illustration of shredder, (a) Top and Front view, (b) inside view, (c) Scrap
33 before shredding, (d) Scrap after shredding [41].
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37 3.2 Chemical Recycling

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40 This chemical recycling process is included as an adjunct to the mechanical recycling process.
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42 This chemical recycling can be defined as a process by which the polymer structures are
43 chemically converted into monomers or partially depolymerized into the final oligomers via a
44 chemical reaction (polymer structure change). These obtained monomers are again subjected to
45 new polymerization to get the same polymers or compatible polymer products. This process is
46 capable of transforming the thermoplastic material into small molecules suitable as feedstock
47 materials, beginning with oligomers, monomers, or hydrocarbon compound mixtures. The
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3 complete adoption of this process is difficult in industries due to the requirement of more expert
4 panels for handling the process and higher investments [50,51]. As per the reports, the recycling
5 of one tonne of plastic solid waste (PSW) can save nearly 130 million KJ of energy, which is also
6 expressed as 123 million BTU (British Thermal Units). This energy is equivalent to the energy
7 released when combusting nearly 22 barrels of oil. Effective and advanced recycling methods such
8 as chemical recycling can save 3.5 billion barrels of oil for processing of global PSW with a
9 savings of 176 billion US dollars [52]. Moreover, plastic waste was suggested as a promising feed
10 material during the production of fuels and chemicals. The current industrial interest is not
11 restricted to energy recovery or mechanical recycling, but also in the delivery of petrochemical
12 feedstock or valuable monomers. Feedstock examples such as nylon, PUR, and PET exist with
13 chemical recycling options [53]. The different processes involved in chemical recycling are
14 discussed in the following sections.

3.2.1 Dissolution/precipitation

34 In this recycling process, the plastic has impurities and additives of other materials or polymers
35 that are dissolved. A solvent for this process has been selected to dissolve the required polymer.
36 After this, the unwanted additives are removed, and the remaining polymer is precipitated. This
37 process is not completely chemical in nature because it does not involve the splitting of chemical
38 bonds. However, basic chemical knowledge is required to analyze the polymer/solvent interaction,
39 solvent recovery, and solvent design to proceed with the dissolution process [54,55]. **Figure 6**
40 depicts the two methods of dissolution using a single solvent and a solvent plus anti-solvent. The
41 polymers obtained from this process are free from pigments, and other recovered flame retardants
42 are reusable. During the process, the added solvent particularly dissolves the specific polymer and
43 is followed by anti-solvent addition for polymer precipitation for recovery. Between the
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dissolution step and the precipitation step, the non-dissolved pigments are distinguished from the polymer solutions. The solution of anti-solvent and solvent needs to be separated once again for reuse in the next process. This separation requires more time and energy for high boiling point solvents [56,57]. Sometimes the residual solvents are left out with the polymers, which leads to changes in the polymer properties. For this process, the solvents should have some characteristics like being capable of dissolving in selected polymers, shouldn't be hazardous, and should be easily recyclable and recoverable. In the case of organic solvents, the process of dissolution is relatively slow because of the small contact areas between the solvent and plastics. Finally, the obtained polymer is in crystallized form, and hence requires upgrading to get reliable virgin quality. In addition, precipitated polymers must be extruded to produce granulates for the manufacture of new plastic materials [58,59]. Poulakis and Papaspyrides [60] conducted a dissolution process for polypropylene (PP) pipes. Initially, the PP pipes were cut into small pieces and soaked in xylene solution (0.15 kg/l) at 135⁰ C. This was the optimum concentration for this process. Beyond that, the solution will become viscous and it will be difficult to process it further. The solution was further filtered with an exclusion size of 20–70 μm. Furthermore, the acetone was added to the filtrate and then polymer precipitation was done. The grains of polymer were separated through filtration inside the vacuum. Again, the washing was done several times with acetone. In the end, the polymer grains were obtained after drying in an oven for 10 hours at 90⁰ C.

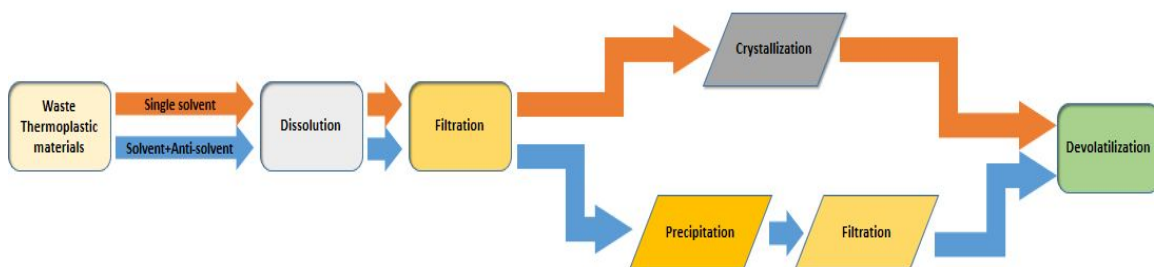


Figure 6. Dissolution process flowchart [54].

Li et al. [61] conducted a dissolution recycling experiment on 3D printed polylactide (PLA)/Lunar Regolith (CLRS-1) composites. This recycling experiment was conducted in an oil bath inserted with a soxhlet extractor. The 3D printed gear-shaped composite specimen was inserted into the extraction thimble. The extraction thimble bottom was provided with a 0.22 μm size membrane filter. The solvent used was tetrahydrofuran and the oil bath was maintained at 80^o C. To avoid bumping, a few porcelain pieces were provided in the bottom flask. The whole recycling process was done with a maximum duration of 2 h. After the process, the CLRS-1 material settled at the membrane filter, and then the recycled PLA powder was obtained through distillation under low pressure. Hadi et al. [62] experimented with the recycling of LDPE, HDPE, and PP thermoplastics using the dissolution method and analyzed the characteristics of the plastics after the recycling process. The petroleum ether/toluene blend was used as a solvent at a 1:15 ratio with waste polymer. The precipitation was done with non-solvents like petroleum and N-hexane. After the complete dissolution, the solution was filtered within a size range of 20-70 μm . Furthermore, the non-solvent was added to the filtrate and then precipitated. Again, the precipitated polymer was cleaned with the same non-solvent. The dried polymer grain/powder has no changes in the IR spectra as compared to virgin thermoplastics. Poulakis and Papaspyrides [63] recycled the used PET bottles through the solvent process. The PET bottles were made into small pieces and added to the N-Methyl-2-pyrrolidone (NMP) solvent (0.2 kg/L) and maintained at a temperature of 165^o C. The overall equipment setup included a thermometer, condenser, and stirrer. The filtration was done with Seitz Merkur EF 6/03 equipment with an exclusion size of 20–70 μm . Further, the precipitation was done by adding n-octane into the hot filtrate, which included polymers. The obtained grains were filtered again within a vacuum and washed twice using a solution of n-

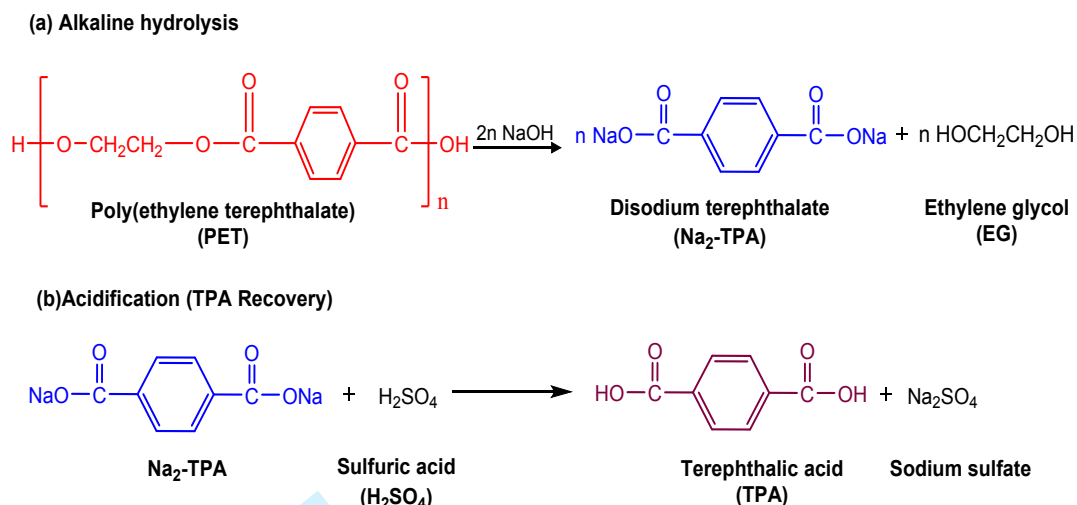
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3 hexane. Further processes included the drying of reclaimed polymer for a duration of 14 h at 90⁰
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5 C. Cella et al. [64] conducted a recycling process on polystyrene and evaluated the recycled PS
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7 properties compared to the virgin material. The polystyrene dissolution was done with ethyl acetate
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9 and the solvent removal was carried out with two processes: vaporization in a tubular evaporator
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11 at 850 C with water and vaporization via the extrusion process. This recycled PS exhibited a
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13 presence of volatile compounds, and it was reduced after the drying process. It was also observed
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15 that the glass transition temperature in recycled PS material increased due to the elimination of
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17 plasticizing additives from the recycled PS material. To go with a green process, García et al. [65]
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19 conducted a dissolution process for PS using d-limonene, phellandrene, p-cymene, and terpinene
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21 solvents. These solvents' actions have no significant effect on polymer chain degradation. The
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23 solubility of PS in vegetable oils increased with increasing processing temperatures, but sometimes
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25 the polymer chain degradation might occur due to higher temperatures.
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3.2.2 Solvolysis

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35 Compared to pyrolysis, solvolysis is one of the potential techniques for extracting monomers from
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37 low-temperature-resistant thermoplastics. In the process of solvolysis, the polymer's hydrolyzable
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39 bonds are broken in the presence of water or alcohol. To get better productivity, yield, and reaction
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41 conditions, catalysts were preferred to promote the reactions [66,67]. The solvolysis process can
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43 be achieved with various processes like hydrolysis, methanolysis, glycolysis, and aminolysis.
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45 These processes are limited to polymers having ether, acid amide, and ester bonds. The advantages
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47 of different solvolysis processes are the maximum possibility of further purification of monomers,
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49 re-polymerization possibility equal to virgin grade, and filtering of colorants and additives. These
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51 advantages are most suitable for polyurethane because it can't be recycled using mechanical
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3 methods. If the recycled monomer quality is not acceptable, then it can be mixed with conventional
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5 polymers for synthesis [68,69].
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8 The process of hydrolysis is good for environmental protection and requires more energy
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10 as compared to other processes of solvolysis. This reaction is possible to execute in various
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12 conditions like alkaline, acidic, or neutral mediums. In the case of polyurethane hydrolysis, the
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14 reaction takes place between the water molecules and polyurethane at high pressure and
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16 temperature to form CO₂, amines, and polyols. At first, the carbamic acid was formed during the
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18 reaction, and it was subsequently decomposed into CO₂ and amine due to thermal instability. As
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20 per the report, this hydrolysis recycling is very limited due to the need for high pressure and
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22 temperature. In previous decades, neutral PET hydrolysis was known, and it was processed in the
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24 molten phase at temperatures above 245⁰ C with a PET/water ratio (w/w) greater than 1/5.1. Then
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26 this process and reaction rate were further improved with the addition of catalysts like zeolites,
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28 organophosphorus compounds, or alkaline metal acetates [70,71]. With the help of hydrolysis, the
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30 waste PET bottles can be directly depolymerized into TPA (terephthalic acid) and EG (ethylene
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32 glycol) in the presence of an alkaline medium. Further, the obtained TPA was recovered from
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34 disodium terephthalate (Na₂-TPA) via an acidification process with the help of sulfuric acid.
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36 Finally, the obtained products of this acidification process were terephthalic acid and sodium
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38 sulfate. The two alkaline hydrolysis and acidification processes are illustrated in **Scheme 1** [72].
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Scheme 1. (a) Depolymerization PET by alkaline hydrolysis, and (b) TPA recovery by acidification [72].

Ügdüler et al. [73] experimented the depolymerization of polyethylene terephthalate (PET) and obtained a maximum yield of nearly 95% at a temperature of 80⁰ C with less than 500 µm particle size in a solution bath of EtOH: H₂O (60:40) and with NaOH solution (5 wt%) for 20 minutes. In terms of environmental purposes, chemical recycling has gained more interest for post-consumer PET through hydrolysis to get main reaction products like ethylene glycol (EG) and terephthalic acid (TPA). For this purpose, the waste PET was synthesized via these precursors from the PET waste. Moreover, this could be helpful in fossil fuel conservation. Initially, the NaOH solution was heated in a flask at 90⁰ C, and then flakes of PET were added to the heated NaOH solution. This reaction was kept for 9 hours, and then the heat supply was turned off, followed by keeping the solution in an ice bath. Then the solution was added to sulfuric acid (concentrated) to neutralize the reaction, which led to TPA formation. The obtained yield was calculated using Eq. (1). Finally, this precipitate was followed by filtration, purification, washing, and drying processes [74]. Kosimidis et al. [75] experimented with the recycling of PET via depolymerization reaction from waste PET bottles by using a trioctylmethylammonium bromide catalyst, and the experiment was

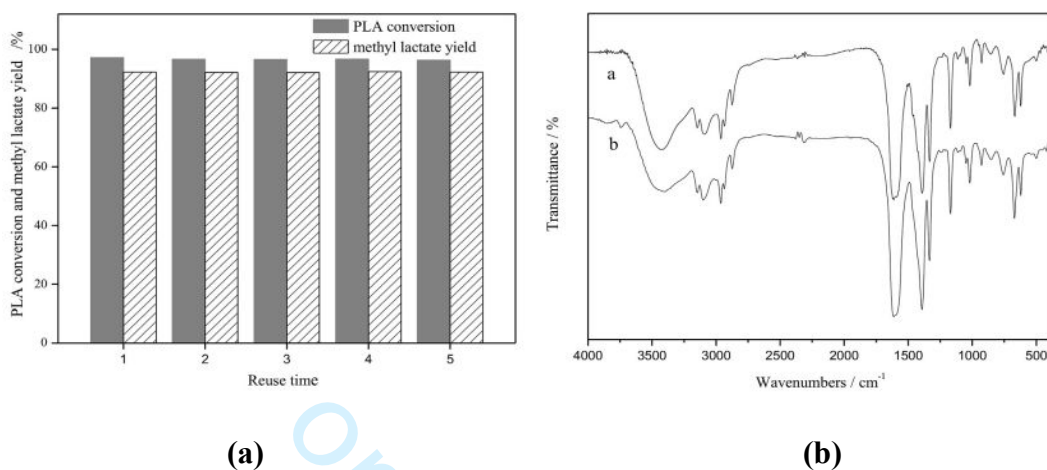
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3 conducted with different concentrations of NaOH solution (5-15% w/w) and temperatures (70-95⁰
4 C). The depolymerization process was executed for a maximum NaOH concentration of 15% w/w
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6 and a temperature of 95⁰ C. Moreover, the TOMAB catalyst also influenced the depolymerization
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8 process, and the maximum yield obtained was 98%.
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$$11 \quad TPA \text{ yield } (\%) = \frac{\text{Produced moles of TPA}}{\text{Theoretical moles of TPA}} \quad (1)$$

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15 Methanolysis is the commonly preferred and effective solvolysis process for the recycling
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17 of polycarbonates, PET, and polyamides. Most of the utilized PET waste is being recycled using
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19 mechanical processes, and the drawbacks of mechanical recycling are the reduction of molar mass
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21 and physical-mechanical properties of the recycled polymer. Therefore, it is commonly preferred
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23 in lesser amounts for bottle production and a majority for carpet production. The commercial
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25 mechanical recycling viability of PET is dependent on the price of oil in the market. To overcome
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27 these drawbacks, chemical depolymerization is an effective process to get high-quality oligomers
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29 and monomers [76,77]. Liu and coworkers [78] worked on the recycling of polycarbonates into
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31 bisphenol A (BPA) using SIILS (succinimide-based ionic liquids) media. A mixture of PC,
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33 CH₃OH, and catalyst [HDBU] [Suc] was subjected to magnetic stirring in an autoclave and
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35 maintained an oil bath temperature of 70⁰ C. After the reaction, the PC residual was separated
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37 through filtration, and this filtrate was further concentrated to remove DMC and CH₃OH. Finally,
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39 the H₂O/diethyl ether was utilized for the separation of the catalyst and BPA. Han [79] reported
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41 the depolymerization of PET bottles using methanol under standard pressure (20–40 atm.) and
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43 temperature (180-280⁰ C) using transesterification catalysts like lead dioxide, zinc acetate, cobalt
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45 acetate, and magnesium acetate. This reaction was maintained at a higher pressure to maintain the
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47 liquidity of methanol. The obtained DMT yield was within the range of 80–85% and the
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49 methanolysis products were separated via crystallization or distillation. The final product of
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3 purified DMT can be used for the second polymerization of PET. The whole process of
4 methanolysis is possible to execute in both continuous and batch conditions. But the problem with
5 continuous conditions is the requirement of complex equipment for PET waste addition to the
6 methanolysis reactor that was maintained at higher pressure. Pham and Cho [80] executed the
7 methanolysis of PET using a low-energy catalytic reaction to get a product of dimethyl
8 terephthalate. The methanolysis was begun in the round flask by adding methanol (8.31 g),
9 dichloromethane (22.1 g), K_2CO_3 (143.52 mg), and the water-PET molar ratio was maintained at
10 0.4 with the addition of deionized water. The initiation of PET methanolysis was done with the
11 addition of PET chips (1 g) under stirring conditions for 24 hrs. Finally, the reaction was completed
12 with the separation of the catalyst when the reaction was used with a solid catalyst or with the cold
13 water addition when it was provided with a liquid catalyst. The mixture was separated and filtered
14 to get pure DMT with the identification of insoluble solids and liquid filtrate. Song et al. [81]
15 reported the influence of different catalysts on the methanolysis of PLA materials and analyzed
16 the maximum yield for the product. In their work, the catalyst metal IL 2[Bmim] [OAc]-
17 $[Zn(OAc)_2]$ brought more PLA conversion and product yield. Moreover, this catalyst reduced the
18 temperature for reaction and also reduced the reaction time as compared to other inorganic salts
19 and catalysts. With the 1 h reaction time, the PLA conversion and methyl lactate yield were 93.46%
20 and 87.75%, respectively, due to good catalytic action. Further increasing reaction time to 2 h gave
21 the conversion and yield of 97% and 92%, respectively, and more than 2 h of reaction didn't
22 increase both conversion and yield. The catalyst reusability concerning PLA conversion and yield
23 is illustrated in **Figure 7 (a)**. With the reusability of the same IL catalyst 5 times, there is no
24 significant decrement in the yield and conversion of PLA as observed in the plot. The authors also
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3 compared the IR spectra of the catalyst for a fresh one and a reused catalyst 5 times. It was observed
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5 that there was no significant difference in the spectra, as shown in **Figure 7 (b)**.
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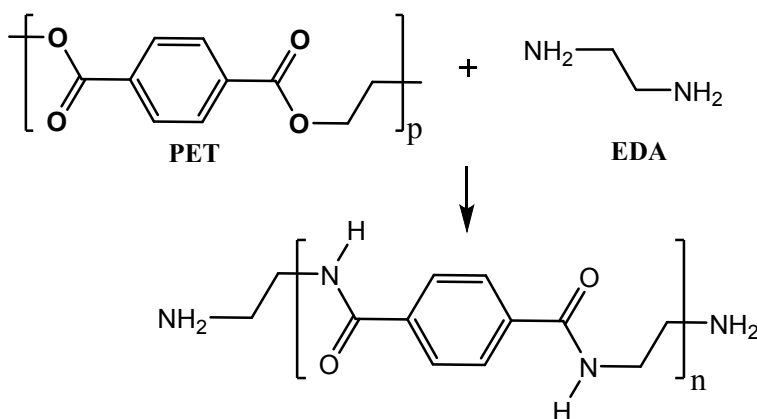


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24 **Figure 7. (a)** Reusability of catalyst 2[Bmim][OAc]-Zn(OAc)₂, (b) IR spectra for catalyst (a.
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26 fresh, b. 5 times reused) [81].
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29 As an alternative to mechanical recycling, glycolysis is also one of the promising recycling
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31 processes with environmental considerations and low energy requirements. This process involves
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33 a transesterification reaction in which the carbonyl carbon in the urethane is attached to an ester
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35 group, which is then exchanged with glycol hydroxyl groups that underwent depolymerization by
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37 the glycols at elevated temperatures [82]. This process can be executed with or without the
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39 presence of catalysts, and the majority of the reactions require catalysts. Catalysts like amine
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41 groups, phosphorus-based compounds, inorganic salts, inorganic acids, etc. are preferred.
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44 Glycolysis is more popular for recycling polyurethane (PU) and is followed by PET recycling. In
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46 the glycolysis process, the waste PUF or PU waste was crushed into small pieces and reacted with
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48 glycol (ethylene glycol, diethylene glycol, and polyethylene glycol) at a higher temperature. In
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50 this solution, the added solvent and reactant were glycol, and hence the optimum ratio needed to
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52 be maintained between glycol and PU. This process takes more time, and layers of the mixture are
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3 obtained after the process. The top layer has recovered polyol and is being used for the new
4 synthesis of polyurethane [83–85]. Al-Sabagh et al. [86] conducted an experimental study on the
5 glycolysis of PET using Zn and Cu acetate-based ionic liquid catalysts to check the optimum yield
6 and PET conversion. This is what happened when the best conditions were met. They were: 1 kg
7 catalyst, 20 kg ethylene glycol (Ethylene Glycol), 3 kg PET, and 3 hours of glycolysis at 190⁰ C.
8 The Cu and Zn acetate catalysts respectively achieved a bis (2-hydroxyethyl) terephthalate
9 (BHET) yield of 53.95% and 45.6%. The PET conversion had no significant decrease in BHET
10 yield on repeated use of catalysts 6 times. Fuentes et al. [87] published an interesting paper on PET
11 bottle glycolysis to obtain a BHET yield using recycled cobalt (CoO) and zinc (ZnO) from lithium
12 and alkaline ion batteries. The reaction was kept in the presence of ethylene glycol for
13 approximately 2 hours at 200⁰ C and obtained a maximum 80% BHET yield. A recent study was
14 reported regarding the use of Fe₃O₄ nanoparticles (magnetic) for PET glycolysis to BHET. These
15 catalysts exhibited almost 100% yield at 12 consecutive reaction cycles at 180⁰ C without any
16 purification process [88]. Lalmangaihzuala and coworkers [89] reported a new study of the
17 glycolysis of PET to BHET using a biomass waste catalyst such as orange peel ash. The advantages
18 of this catalyst are that it is non-toxic, economically friendly, easily available, and renewable. This
19 bio-catalyst depolymerized the PET within 90 minutes and achieved a recrystallized BHET yield
20 of 79%. Hence, the authors called this recycling process the "greener route" for recycling PET.
21 Kathalewar et al. [90] reported the use of neopentyl glycol (NPG) for the glycolysis process of
22 PET with a zinc acetate catalyst at 200–220⁰ C. The NPG was an effective glycol for the
23 depolymerization of PET and generated a valuable product for coating applications. Because of its
24 brittle nature, the recycled PET exhibited good performance, except for its impact resistance
25 properties.

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3 The Aminolysis process is well known for its energetic and environmental criteria. It uses
4 ionic liquids with ammonium content, which makes the whole process expensive. Because
5 aminolysis is essentially a high-temperature process, quick depolymerization is possible due to the
6 high-speed reaction [91,92]. More et al. [93] utilized ethylene diamine for the depolymerization of
7 PET into amino-oligo (ethylene terephthalamide), and a comparative study was also conducted for
8 different depolymerization techniques like solar energy, ultrasound irradiation, UV radiation, and
9 microwave irradiation. Due to its eco-friendly nature, the solar energy was utilized for the
10 depolymerization process for 8 days by keeping the required ingredients inside a tight container.
11 In terms of efficiency, the ultrasound technique was less efficient as compared to other techniques.
12 However, the overall conversion (100%) of PET was observed in all four of the above four
13 techniques. Hoang and coworkers [94] reported the detailed aminolysis process of PET with an
14 excess of ethylenediamine (EDA) for 17 h at 100^o C. The aminolysis of PET in the presence of an
15 excess of ethylenediamine could be postulated as the chemical reaction depicted in **Scheme 2**. The
16 reaction was begun by adding 50 mL of methanol to the mixture of PET flake (3 g) and EDA (15
17 g). This was refluxed again for 3 hours and then cooled to a normal temperature for filtration. The
18 remaining insoluble compounds were rinsed with acetone and methanol. Again, the recrystallized
19 materials at room temperature were subjected to filtration and drying processes. The obtained
20 oligomers have higher melting temperatures due to their extreme molecular weight.
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Scheme 2. PET's reaction with EDA. $n \geq 1$: bis(2-aminoethyl) terephthalamide (BAET); $n \geq 2$: α,ω -aminoligo(ethylene terephthalamide) (AOET) [94].

3.3 Thermo-chemical Recycling

3.3.1 Pyrolysis

The pyrolysis process, also known as thermal cracking, occurs at 500⁰ degrees Celsius without oxygen. Various types of catalysts were utilised in the pyrolysis process to reduce the duration and temperature of the process and to accelerate the specific reactions. In recent years, this process has grown in popularity at the industrial level. In the pyrolysis process, thermoplastics decomposed into solid mixtures, gases (CO₂, CO, and CH₄), polyaromatic chars, and oils (phenols, benzene, and toluene). The released gases and liquids have higher calorific values and varying molecular weights, so they can be used as fuels and sources for various manufacturing processes [95–97]. Unlike the other thermo-mechanical processes, the pyrolysis gives a hydrocarbon rich wax or liquid mixtures that are suitable as a raw materials for refinery industries. This process is typically preferred for harsh depolymerization polymers and those polymers that are difficult to recycle using mechanical recycling methods like mixtures of PS/PE/PP, reinforced fibers, and multilayered

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3 packaging materials [98]. As per the review work done by Armenise et al. [99], the works related
4 to plastics pyrolysis between 2001-2020 were 549 articles (82%), 31 review (4%), and 90
5 proceedings (14%). The clear illustration of publications related to plastic pyrolysis is illustrated
6 in **Figure 8**. The major advantage of the pyrolysis process is the high temperature process, because
7 it guarantees the molecular bond breakage that depends on the random fragmentation,
8 depolymerization, and polymer nature. In other words, the catalytic pyrolysis is also for the same
9 polymers through carbocation and isomerization at lower temperatures. This conventional
10 pyrolysis process is also integrated with plasma pyrolysis, where the transformation of waste
11 plastic takes place into syngas. The recycling temperatures are too high in the plasma pyrolysis
12 that ranges between 1730 and 9730⁰ C for the conversion of plastics to monomers via
13 decomposition. This is an extremely fast process that takes 0.01-0.5 seconds depending on the type
14 of plastic and process temperature. The resultant syngas from plasma pyrolysis is mainly
15 composed of H₂, CO, and hydrocarbons. Additional advantages of plasma pyrolysis are
16 comprehensive polymer breaking, low tar content in the produced gas, and the generated heat is
17 convenient for the generation of electricity in hydrogen production or in turbines [100,101].
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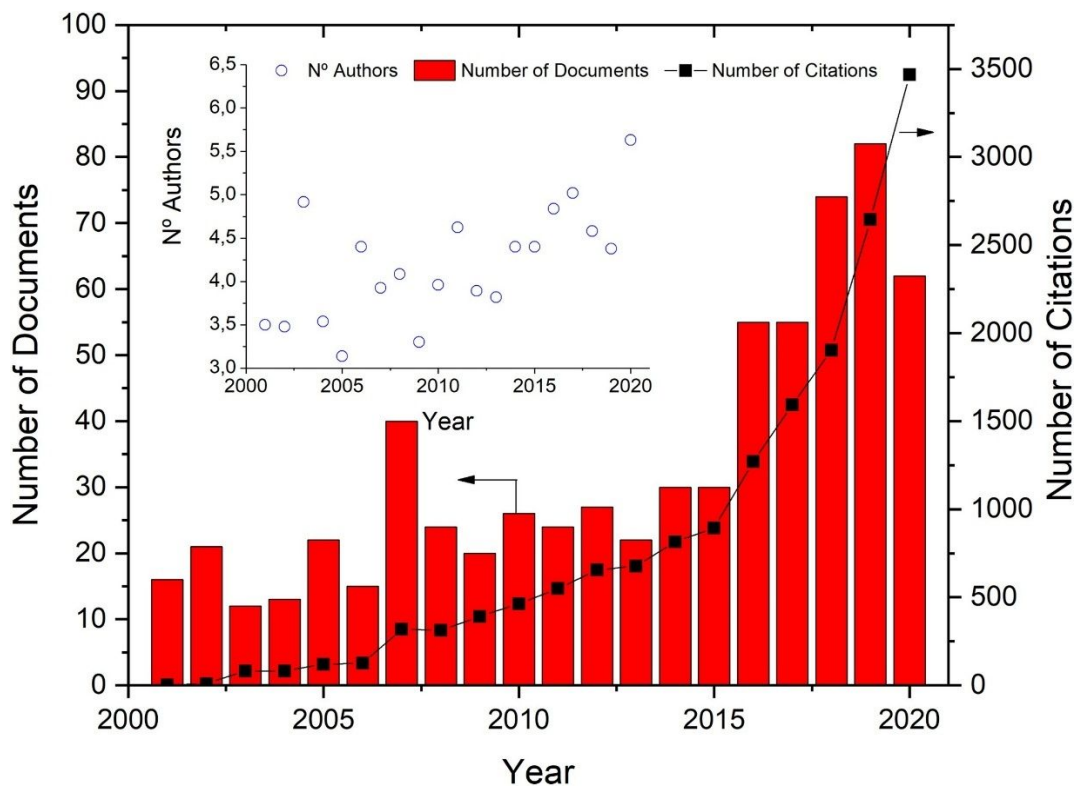
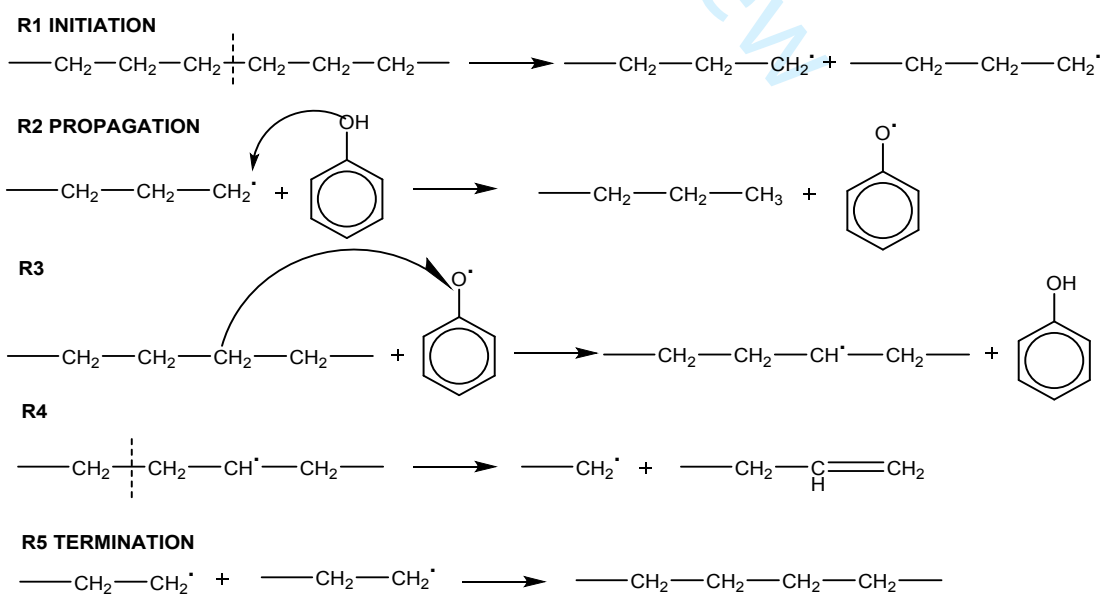


Figure 8. Publications and citations in the field of plastic pyrolysis. Bar graphs associated with number of documents and continuous line associated with number of citations [99].

[\[https://doi.org/10.1016/j.jaap.2021.105265\]](https://doi.org/10.1016/j.jaap.2021.105265)

The pyrolysis process begins with the pretreatment of waste plastic materials to avoid non-plastic contaminants like wood and metal particles. This phase is required to assure the financial viability of the plastic-to-fuel (PTF) facility, and it is normally accomplished by sorting, crushing, or sieving the waste based on its origin. Another key consideration stems from the fact that the various types of plastic waste might vary in shape and size and must be uniformly sized into grains before being fed into the pyrolysis process. This procedure adds to the overall cost of the process. Conducting pyrolysis in the presence of a suitable solvent is another fascinating way to improve it. It is feasible to boost selectivity toward desirable products, in this way Vicente et al. [102] studied thermal cracking of HDPE at 400^o C in the presence of phenol, which supports random

scissions and chain reactions and so accelerates this reaction. Olefins, which are extremely valuable in the petrochemical industry, are the major products of this process. The processes depicted in **Scheme 3** are the most significant ones in the suggested mechanism for HDPE heat degradation in a phenol solution. The HDPE degradation process occurred in multiple reactions: R1 (Initiation), R2 (Propagation), R3 (Propagation Step), R4 (Propagation Step), and R5 (Termination). Thermal degradation is begun by random homolysis of carbon-carbon bonds, resulting in the production of two main radicals [R1]. The presence of the solvent does not affect this reaction. The polymer radical that forms first accesses a hydrogen atom from the phenol, forming an n-paraffin and a phenoxy radical [R2]. This reaction is responsible for the high yields of C5–C32 n-paraffins observed in processes utilizing phenolic solvents. Additionally, the phenoxy radical generated in R2 may steal a hydrogen atom at a random point from an HDPE string, forming a secondary transitional radical and reproducing a phenol molecule as per R3. The obtained secondary radicals may again proceed with C-C bond homolysis concerning R4. Finally, the short radical could be finished off by reacting with another basic radical to make n-paraffin [R5].



Scheme 3. Degradation mechanism of HDPE with the presence of phenol [102].

Miandad and coworkers [103] conducted a groundbreaking study on the topic of investigating the effect of Saudi natural zeolite on the yield and product quality of recycled plastics. Thermal activation (TA-NZ) at 550 °C and acid activation (AA-NZ) with HNO₃ improved the Saudi natural zeolite catalyst. The pyrolysis materials composed of PET, PP, PS, and PE, either mixed or in individual form, were fed in the presence of both modified NZ catalysts. The maximum yield obtained was 60% (AA-NZ) and 70% (TA-NZ) for PS waste as compared to PE and PP. Duque et al. [104] studied the effect of recycling stress history on the pyrolysis of LDPE waste. The major findings indicate that thermal and mechanical stress encourage the growth of long carbon chains while reducing the barriers between compounds. This could be explained by the fact that the need for activation energy for recycled waste was less than the thermal degradation of recycled samples. The waste of regenerated polyethylene had the lowest pyrolysis enthalpy of around 205 J/g. The conflict between evaporation and pyrolysis reactions dominates the pyrolysis route, which is a series-parallel phenomenon. Lim et al. [105] experimented with PET pyrolysis using crushed concrete trash (high in CaO). The authors found that the catalytic activity did not affect the maximum decomposition temperature, but that successful neutralization of decarboxylated chemicals resulted in higher aromatic contents in the final liquid product. With the higher concentration of CaO, Al₂O₃, and SiO₂ in the burnt brick powder catalyst, Hussain et al. [106] observed a marginal improvement in liquid and gaseous product yields and quality in PS pyrolysis. These findings could indicate that CFA and concrete/cement waste have the potential to be low-cost catalyst sources. However, the work done by Jung et al. [107] reveals that the PE pyrolysis with a Ni-based catalyst improved the production of H₂ under an N₂ environment, despite the fast deactivation of the catalyst due to the formation of coke. This problem can be solved by undergoing

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pyrolysis in a CO₂ atmosphere, in which coke is converted to CO and light hydrocarbons. In other words, in-situ catalyst renewal took place, resulting in a longer catalyst lifespan and, as a result, more H₂ production. These studies show that catalytic pyrolysis of PE waste in a CO₂ environment can result in the creation of syngas or H₂. For all types of feedstocks, CO₂ and CO are the major constituents of the gas delivered from the pyrolysis process, and they account for around 60% of overall gas production. These compounds are very small in the case of commercial wastes at around 30%. This is owing to relative feedstock purity and their lower content of chemically bonded oxygen [108]. Dewangga and coworkers [109] worked on the pyrolysis of polystyrene waste to get styrene monomers with the help of a bentonite catalyst. This pyrolysis was executed in a batch reactor at 300-550^o C with polystyrene waste and a catalyst percentage of 0, 5, 10, 15, 20, and 25% waste polystyrene. The 25% catalyst gave a maximum yield of 88.78 wt% as a liquid product, and the analysis of this liquid using GC-MS confirmed the presence of C₈H₈, benzene, and toluene compounds. Overall, 32.34% of the maximum styrene compound yield was obtained at 400^o C for 25 wt% of the catalyst. The majority of the PE pyrolysis methods were supported by heterogeneous acid catalysts like silica, zeolites, and alumina. These are typically nonselective, resulting in a wide range of gaseous hydrocarbons (C₃ and C₄), liquid products like oligomers, cycloparaffins, aromatics, and other solid products. This is caused by the radical process of C–C bond scission, which results in a complicated combination of olefin and cross-linked compounds [110,111].

3.3.2 Microwave-Assisted Pyrolysis

Microwave-assisted pyrolysis (MAP) is one of the promising recycling techniques used to convert biomass and waste polymers into raw fuels and chemicals. The efficiency of the microwave thermochemical process is greater than the traditional organic synthesis method. As a result, MAP

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3 has developed into a very effective technique for promoting the quick conversion of a wide variety
4 of feedstocks into fuels and recycled raw materials. Moreover, microwave-based technology
5 makes the process cost-effective and helpful in developing compact units. Generally, the MAP
6 process requires specific materials that are very good at microwave absorption. However, MAP
7 has a power density barrier over which materials cannot be pyrolyzed without the use of susceptors
8 (usually $> 108 \text{ W/m}^3$) [112–114]. The primary benefit of microwaves is that they heat the material
9 from within, resulting in rapid thermal transfer and energy savings. The MAP approach provides
10 several advantages over more traditional methods. When compared to traditional pyrolysis, it
11 provides more even heat distribution, faster heating rates, more supervision over the technique,
12 and enhanced output speed. The MAP process requires materials to have more dielectric strength,
13 but the lower dielectric constant of plastic waste leads to the requirement of microwave absorbers,
14 which are capable of generating hotspots [115]. Bartoli et al. [116] employed a polystyrene
15 material for depolymerization via the MAP process using carbon as a microwave absorber.
16 Working with less pressure, the liquid yield was enhanced by more than 75% above that produced
17 at ambient pressures. The maximum concentration of styrene in the liquid was achieved (71.9 wt
18 %) using a fractionating system at a residual pressure of 21.3 kPa, which corresponds to the
19 maximum quantity of styrene that was recovered from PS (60.6 wt%). Undri et al. [117] conducted
20 microwave-assisted pyrolysis for PLA under an anoxic atmosphere inside an oven with a
21 condensing system using the tire, carbon, or Fe microwave absorbers. The pure l-lactide was
22 crystallized via the liquids obtained from the pyrolysis of PLA, while lactide in meso forms was
23 detected in the liquid using GC–MS analysis. At the end of the process, 27.7% of the lactide was
24 recovered from PLA pyrolysis. PLA was pyrolyzed in the presence of additional polymers that
25 may pyrolyze into aromatic compounds, such as those found in tires where the cross-reactions
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3 occurred during the pyrolysis, altering both yield and liquid properties. Lactide did not precipitate
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5 due to aromatic chemicals produced during tire pyrolysis.
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7 8 **3.3.3 Fluidized-bed Pyrolysis** 9

10 This process involves the thermal decomposition of polymer matrices followed by the collection
11 and release of unwanted particles. The fluidized bed offers the attrition required to liberate
12 monofilaments from the reinforcing phase and is potentially a convenient technique for rapidly
13 heating feed material in an airstream [118]. Imani Moqadam et al. [119] investigated the catalytic
14 degradation of PS using a silica-alumina catalyst in the fluidized bed reactor. The catalytic
15 performance of silica-alumina was good for polystyrene degradation, with an aromatic selectivity
16 of over 95%. The temperature was the major significant pyrolysis parameter since increasing
17 temperature improved styrene production and conversion. The selectivity of styrene increased as
18 the catalyst amount was raised, but the liquid yield was lowered. Gases, liquids, and residues were
19 released when the plastic degraded. The carbonaceous substances that stay inside the reactor and
20 are put on the reactor body are referred to as residues. A semi-batch reactor was used to catalyze
21 the breakdown of polystyrene. In a Pyrex vessel with a volume of 1 L, a mixture of polystyrene
22 and catalyst was loaded. To ensure that the environment was oxygen-free, the reactor was purged
23 twice with nitrogen. Heating was begun after checking that there were no leaks. The catalyst and
24 polymer combination was heated at a rate of 20⁰ C/min until it reached the appropriate temperature
25 and then kept there until pyrolysis was complete. After cooling down, the volatile compounds are
26 separated in the condensing system. Liu et al. [120] investigated polystyrene plastic pyrolysis in a
27 laboratory fluidized bed reactor at temperatures ranging from 450 to 700⁰ C. At 600⁰ C, he was
28 able to achieve a maximum yield of 78.7% wt% styrene. Kaminsky et al. [121] used a fluidized
29 bed reactor at 580⁰ C to achieve a 65 wt% yield of styrene. In a later investigation, he employed
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3 the same experimental reactor at 520⁰ C and reported the formation of oil containing 82.8 percent
4 w/w of styrene after pyrolysis, utilizing additional distillation of the liquid fraction. The fluidized
5 bed process is ideal for pyrolysis because it provides excellent heat and material transfer rates, as
6 well as consistent temperatures. The features of this process are uniformity in recovered products,
7 no movements in the equipment parts during the hot region, and the reactor can be easily sealed in
8 the closed system. In other words, the fibers, as well as excessive concentrations of metals and
9 other additives, are sensitive to the fluidized bed [122].
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19 **3.3.4 Gasification**

20 Gasification, also known as "indirect combustion," is the process by which solid municipal plastic
21 waste is converted into fuels or gases through gas-forming reactions. This gas-forming reaction
22 involves partial oxidation of solid waste with the help of an oxidant, and the oxidant amount
23 shouldn't promote stoichiometric combustion [123]. Gasification has various potential advantages
24 over traditional solid waste combustion, the most important of which is the ability to combine
25 operating conditions (particularly temperature and equivalence ratio) and reactor features (fixed
26 bed, plasma reactor, fluidized bed, vertical shaft, rotary kiln) to produce a syngas suitable for use
27 in several applications. The gasification process for solid waste is a complicated one that involves
28 many interactions, both physically and chemically, that takes place at temperatures generally
29 above 600⁰ C, with the exact temperature varying depending on the characteristics of the waste
30 and the type of reactor, particularly melting and ash softening temperatures [124,125]. Solid waste
31 gasification has many sequential exothermic and endothermic reactions, and it is graphically
32 shown in **Figure 9**. The initial heating and drying stage can reach a temperature up to 160⁰ C and
33 has many events via steam and liquid migration. In the next step, the devolatilization occurs at
34 700⁰ C with mass and heat transfer in the thermal cracking reaction. This stage delivers char (waste
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residue), tar (hydrocarbon vapors), and some permanent gases like NH_3 , CO , CO_2 , and CH_4 . Moreover, the thermal cracking converts some of the generated vapors into gas and char. Volatiles make up a large amount of the carbonaceous fuel that produces an easily combustible environment of fuel gases all-around solid waste and also the portion of gases produced from the gasification process, as shown schematically in **Figure 9** [126,127].

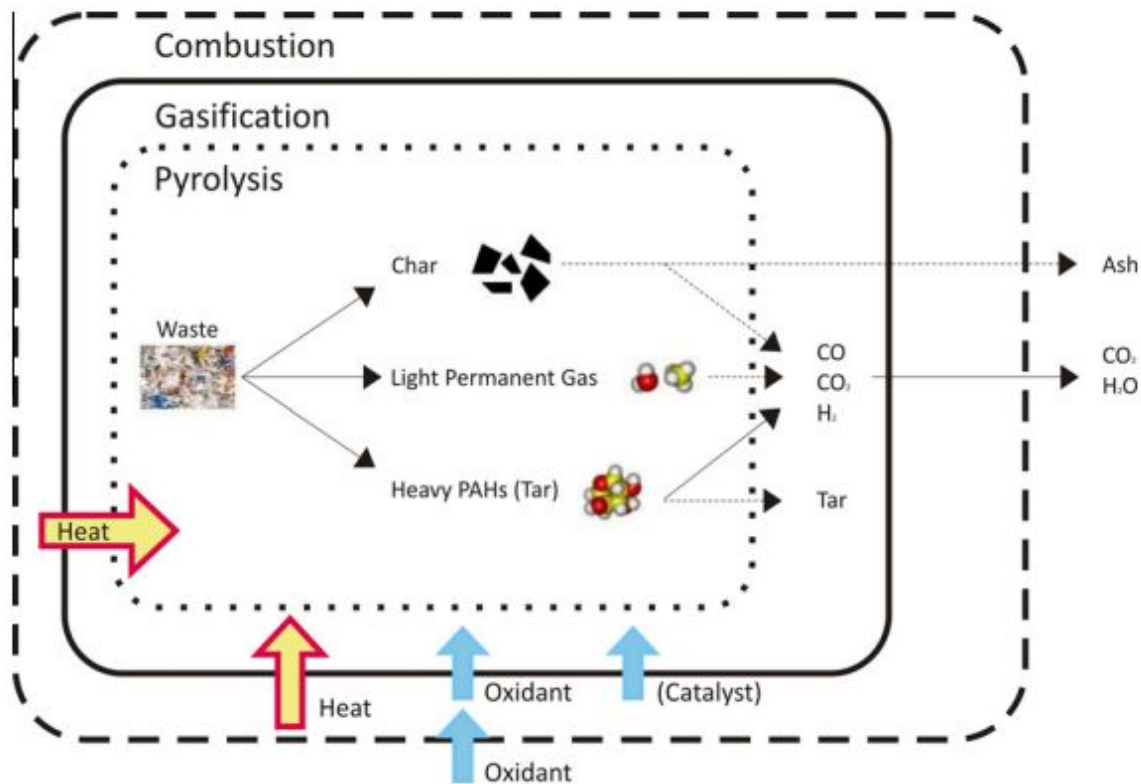


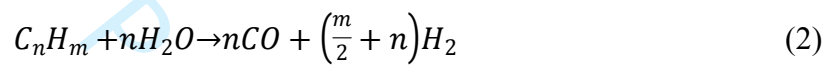
Figure 9. Schematic illustration of pyrolysis process, gasification and combustion stages [126].

[\[https://doi.org/10.1016/j.wasman.2011.09.025\]](https://doi.org/10.1016/j.wasman.2011.09.025)

There are different gasification processes based on the oxidation medium, such as partial oxidation with atmospheric air, oxidation with pure oxygen, plasma gasification, and steam gasification. The partial oxidation with air produces a producer gas with a calorific value of between 4 and 7 $\text{MJ}/\text{m}^3_{\text{N}}$, which is diluted by ambient nitrogen (up to roughly 60%). This value was deemed too low to be used in a gas turbine (due to the fact that it is significantly lower than

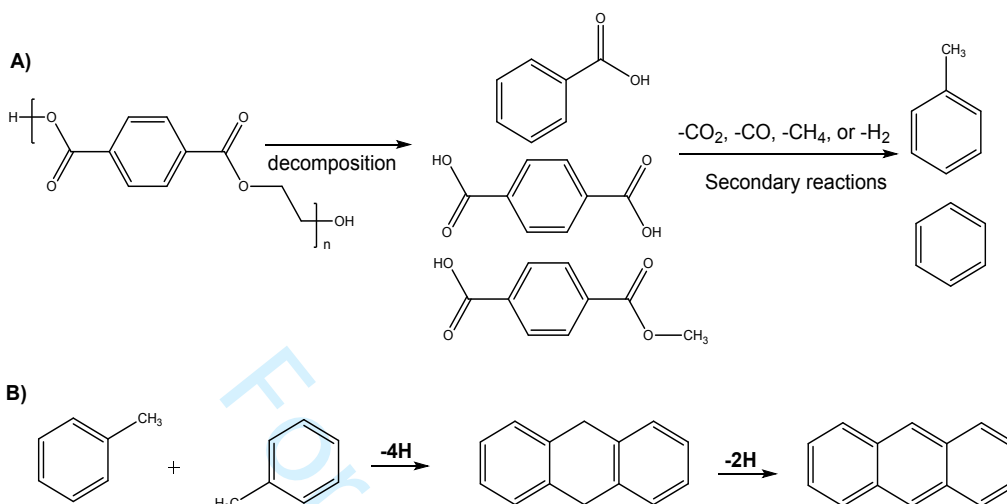
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3 that of natural gas, which is close to 38 MJ/m³_N). Some operations require oxygen-enriched air,
4 which is a mixture of oxygen and nitrogen with an oxygen level of more than 21% and up to 50%.
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6 The purpose is to achieve a greater heating value for gas as a result of the lower nitrogen
7 concentration, allowing auto-thermal processes to be carried out at higher temperatures without
8 the need for expensive oxygen. Steam gasification produces nitrogen-free syngas with a higher
9 hydrogen concentration and a moderate heating value (15–20 MJ/m³_N). Steam will be the only
10 gasifying agent in this scenario; the reaction does not contain exothermic reactions, so the
11 endothermic gasification processes require an external source of energy. In plasma gasification,
12 the heat sources are plasma arc torches that produce electric arcs with a temperature of up to 1500⁰
13 C and they can be controlled with the supply of gasification agents [128–130]. Li et al. [131]
14 conducted a PET recycling experiment through steam gasification and analyzed the influence of
15 gasifying agents on the product composition. The findings revealed that while steam was the
16 primary source of H₂ in steam gasification, increasing the steam to fuel ratio did not significantly
17 enhance the H₂ yield. With the increment of temperature to 800⁰ C from 750⁰ C, the improvement
18 of H₂ yield by 87.7%, biphenyl yield by 123%, and CO₂ gas product yield by 40.3%. During the
19 PET steam gasification process, the temperature influence was greater than the steam/fuel ratio
20 and gas residence time. The major advantage of PET gasification was the liberation of 3 to 4 times
21 less CO₂ than PET burning. Gasification does not require a rigorous sorting process. It is believed
22 to be considerably easier and more flexible than traditional recycling processes. However, not
23 every step in the sorting process is fully automated, so they are supposed to be less labor-intensive.
24 Another benefit is the use of char residue output as a fuel or feedstock material for other
25 petrochemical processes [132]. Liu et al. [133] reported the use of SCW gasification technology
26 for the production of fuel gas using ABS thermoplastic. The optimum temperature range and
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3 pressure found were 450-700⁰ C and 23 MPa respectively. The recovery of styrene monomers was
4 possible under the optimum reaction conditions of 400⁰ C and 3 min duration. Regrettably, the
5 steam gasification process is endothermic as represented in Eq. 2 [132], and it necessitates the
6 presence of air or oxygen to produce the necessary heat via exothermic reactions (Eq. 3) [132]. In
7 terms of cost and environmental impact, gasification in clean air is less expensive than steam
8 gasification. However, the existence of N₂ in an elevated temperature environment causes the
9 formation of hazardous nitrogen oxides (NO_x), resulting in NO_x management challenges and
10 greater expenses. Furthermore, air gasification necessitates bigger volumes, which negatively
11 affects investment costs [134].



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32 Choi et al. [135] conducted an air gasification experiment on PET via a two-stage gasifier, and the
33 carbon was utilized in the two-stage gasifier for the reduction of tar. Moreover, carbon played a
34 critical role in H₂ production during the gasification process. The impact of steam as a gasifying
35 agent on H₂ production was less than the impact of air. The temperature of the tar-cracking reactor
36 had a considerable impact on the gas quality, resulting in increased CO and H₂ generation at high
37 temperatures. The increment in temperature of the tar-cracking reactor resulted in reduced tar
38 content to 23 mg/Nm³ from 394 mg/Nm³ in producer gas. The production of polyaromatics (or
39 char) during PET gasification is seen in **Scheme 4**. The production of char on the AC could have
40 been aided by the PET's structural properties, which include an aromatic ring in its repeating unit
41 and are extremely difficult to dissolve during gasification because of their great thermal stability.
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PET's chief breakdown products include aromatic chemicals such as terephthalic acid, benzoic

acid, and monomethyl terephthalate. It is possible that more decomposition of these aromatics could lead to secondary decomposition products like benzene and toluene being formed.



Scheme 4. Formation of (a) Products from primary and secondary decomposition, (b) char during PET gasification [135].

Wang et al. [136] analyzed the CO₂-assisted co-gasification for HDPE and PET plastics mixture with pinewood pellets (PWP) in a fixed bed reactor at 800^o C. Both the PWP–PET and PWP–HDPE mixtures had a beneficial impact on hydrocarbon output but a negative impact on solid yield. The H₂ yield in the PWP–PET mixture did not differ from the calculated value. However, the restricted porosity of biomass feedstocks from the softened PET had a detrimental influence on CO₂ consumption and CO production. The PWP–HDPE mixture showed a substantial increase over the projected value, whereas the CO yield indicated a modest increase but a slight reduction in CO₂ consumption. The SEM results on the surface morphology of solid residue from individual plastics and with PWP mixtures showed that the thermal reaction under CO₂-gasification impacted the solid-surface structures. Wang et al. [137] compared the gasification process of polypropylene with a catalyst (Ni/Al₂O₃) and without a catalyst. In comparison to non-catalytic gasification, the catalyst-based in-situ and quasi-in-situ gasification generated 120.5% and 137.1% more H₂, 38.2%

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3 and 24% more CO, 57.1% and 98.2% more C_mH_n, and 44.1% and 43.8% more total syngas,
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5 respectively. For non-catalytic, in-situ, and quasi-in-situ catalytic gasification, the total energy
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7 efficiency peak was 45.1%, 69.3%, and 78.9%, respectively, although each unit mass of PP could
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9 have used approximately 0.8, 1.1, and 1 unit of CO₂. Overall, this work reports the effectiveness
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11 of quasi-in-situ catalytic gasification in terms of better yield and the best solution for greenhouse
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13 emissions.
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16 17 **3.4 Enzymatic recycling**

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19 The degradation of various thermoplastic polymers has been studied using the enzymatic activity
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21 of several microbial and fungus species. The fundamental challenge in enzyme breakdown of
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23 thermoplastic polymers such as PP and PE is to overcome their derivative nature, such as
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25 hydrophobicity, inertness, and stability. Hence, the solution for the implementation of reactivity is
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27 thermal oxidation or UV pretreatments [138,139]. Natural occurring microorganisms have evolved
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29 capacities to digest the plastic waste in the environment over time and sustained exposure, allowing
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31 them to metabolize it as an energy source within the high organismal variety that exists in soil,
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33 compost, or marine habitats. Generally, the biodegradation process has four major steps: (i)
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35 microorganism adhesion on the waste plastic surface; (ii) microorganism growth followed by
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37 colonization with carbon-rich biofilm formation via metabolization, (iii) breakage of longer
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39 molecular chains into smaller ones by means of enzymatic hydrolysis, (iv) final degradation into
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41 dimers, oligomers, and other products like H₂O, CO₂, and CH₄. The process is predominantly
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43 mediated by enzymatic action, and hence its efficacy is strongly dependent on the accessibility of
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45 the polymeric chain [140–142]. Enzymatic activity has been found on a range of polymers,
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47 including PP, PE, and, most notably, PET, by a variety of microbial and fungal species. *Bacillus*
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49 *subtilis* H1584, *Kocuria palustris* M16, and *Bacillus pumilus* M27, all isolated from the Arabian
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3 Sea, caused only minor weight loss in PE films [143]. The enzyme Cutinase from *Humicola isolens*
4 may also function at high temperatures, and it degraded the PET film by 97% when it was
5 incubated for 96 h at a temperature of 70⁰ C. This shows the potential for commercial PET
6 conversion under mild conditions [144]. Singh et al. [145] experimented with the enzymatic
7 depolymerization of PET plastic into ethylene glycol (EG) and terephthalic acid (TPA) with
8 techno-economic considerations. The cost analysis results estimated the recycled TPA production
9 cost at \$1.93/kg. Moreover, the expenses for pretreatment, processing costs, and feedstock prices
10 contribute more toward overall depolymerization costs. Meanwhile, the amount of PET
11 breakdown, solids loading, enzyme pricing, and enzyme loading is all-important cost factors in the
12 depolymerization process. When compared to vTPA, the supply chain analysis demonstrates that
13 PET recycling via the enzymatic approach requires less energy and emits fewer greenhouse gases.
14 Then and coworkers [146] reported the enzymatic recycling of PET using TfH, BTA-2, Tfu_0882,
15 TfCut1, and TfCut2 catalysts at 55-65⁰ C for a duration of 48 h. However, those enzymes are not
16 stable enough in this range of temperature to degrade post-consumer PET materials efficiently. As
17 measured by circular dichroism spectroscopy, the melting temperatures of the enzymes increased
18 between 10.8 °C and 14.1 °C when Ca²⁺ or Mg²⁺ cations were added to them. After crystalline
19 PET degradation, the weight losses recorded were up to 12% and 9%, respectively, for 10 mM
20 Ca²⁺ and 10 mM Mg²⁺ cations. Their replacement with arginine resulted in an increase in the
21 thermal stability of the respective enzyme versions. The developed versions of TfCut2 are
22 stabilized catalysts that are suited for PET hydrolysis processes that do not need Ca²⁺ or Mg²⁺.
23 Sadler and colleagues [147] used modified *Escherichia coli* to establish an enzyme-catalyzed post-
24 consumer PET decomposition to produce vanillin. After comprehensive process optimization
25 studies, optimum vanillin titers of 785 M (79% conversion) were attained, a 157-fold improvement
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over pre-optimization titers. Cell permeabilization, biotransformation temperature, and the application of ISPR to boost flow towards vanillin were all key factors in this high conversion. In the polymer biodegradation process, the first stage is related to microorganisms' attacks by delivering extracellular enzymes for hydrolysis or oxidation of the polymers. This process is especially beneficial in increasing polymer hydrophilicity and improving polymer adhesion by fungi. Further, it proceeded with chain scission and resulted in molecular weight reduction. The proposed mechanism of PP biodegradation is illustrated in **Figure 10** [148,149]. Alcalase, Lipase, and Esterase are the enzymes that are capable of hydrolyzing PLA. The maximum activity of these enzymes is influenced by ideal pH values, crystallinity, temperatures, and chain stereochemistry. As a result, researchers have focused more on enzyme degradation [150].

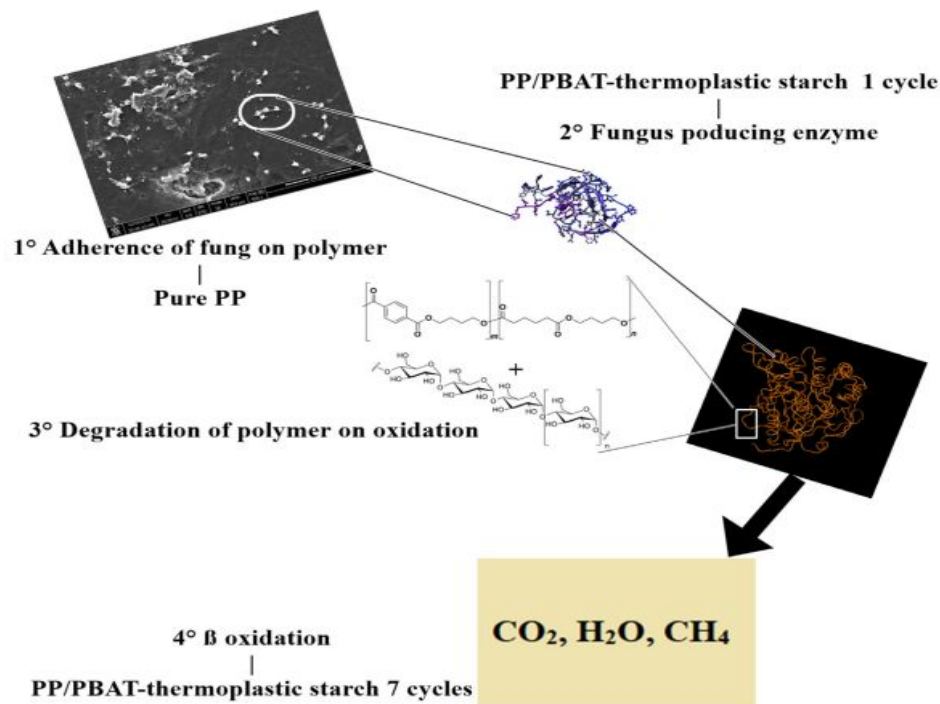


Figure 10. Proposed mechanism of PP polymer biodegradation [148].

Quartinello et al. [151] experimented with the enzyme PET hydrolysis with the help of *Humicola insolens* cutinase (HiC). After 6 hours of incubation, the largest proportion of soluble

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3 TA (6.5 mM) was produced using either 1 or 2 mg ml⁻¹ of HiC, with no further increments until
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5 24 hours. Lower enzyme doses of 0.1 and 0.5 mg ml⁻¹ resulted in TA releases of 0.53 and 1.9 mM,
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7 respectively. After enzymatic treatment of the chemically modified PET, terephthalic acid with
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9 97% purity was produced, which is close to the synthesized TA (98% pure). When PET was
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11 chemically pre-hydrolyzed with the presence of a zinc acetate catalyst, it had a deleterious impact
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13 on enzymatic hydrolysis. In fact, the amount of TA was not increased any further. Ameen et al.
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15 [152] isolated the mangrove fungi from tidal water and utilized these fungi for biodegradation
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17 studies on LDPE plastic. Among the collected isolates, six of them were able to grow with LDPE
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19 in a vitro environment without the presence of dextrose. During co-cultivation with LDPE, these
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21 fungi gathered much more biomass, generated more ligninolytic enzymes, and emitted
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23 significantly more CO₂ than the controls. The isolates, *Alcanivorax* sp., *Cobetia* sp.,
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25 *Exiguobacterium* sp., and *Halomonas* sp., were able to degrade LDPE without the requirement of
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27 prior oxidation or chemical treatment of LDPE. The morphological changes like pits, roughness,
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29 and cracks were confirmed using FE-SEM and AFM, and they were reported by Khandare and his
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31 coworkers [153]. Sen and Raut [154] reviewed the interactions of micro-organisms with LDPE
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33 material and analyzed the biochemical changes. They reported that microbial enzymes are the most
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35 powerful elements for biodegrading LDPEs among different biological agents. Most enzymes'
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37 biodegradation activity is stronger in fungi than in the bacterial stage. Cutinases are one of the
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39 enzymes that have been shown to be capable of degrading hydrophobic PET. Cutinases are found
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41 in plants infected with viral pathogenic fungus, and they aid in fungal penetration of hydrolyzed
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43 cutin [155]. In particular, any pretreatment of the thermoplastic will affect its susceptibility to
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45 enzymatic and hydrolytic breakdown, respectively. Moreover, the efficiency of degradation was
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47 highly influenced by the properties and state of the polymers. The use of biodegradable plastics
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3 has several advantages, including the ability to recycle in a broad yet very specialized manner via
4 biocatalysts (microorganisms and enzymes). Upcycling and recycling degradable and traditional
5 thermoplastics (PET) into added-value chemicals and monomers have been proven using
6 developing microbial-based methods in a bioreactor configuration [156].
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13 **4.0 Role of Additives in Thermoplastics Recycling**

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16 The deterioration of polymers during processing stages and by-products over their lifetimes, as
17 well as the degradation of heterogeneous recycling, are the key issues in post-consumer plastics
18 recycling. To reduce these negative impacts during recycling, there are two solutions for
19 homogeneous materials: slow degradation via restabilization and loading of additives to improve
20 the thermoplastic performance without altering the final cost of recycling material [157,158]. The
21 different classes of additives and their roles are tabulated in **Table 1**. The main issues in the
22 recycling of homogeneous plastics are solely connected to the degradative phenomena that occur
23 throughout the recycling process. Because oxygenated groups produced during processing or
24 usage greatly accelerate the deterioration of plastics, these processes are often far more severe than
25 in virgin polymers [159,160]. The final characteristics of secondary materials are severely harmed
26 as a result of this. Whatever the end use of polymer recycled materials, additives play a critical
27 role in polymer manufacture, acting as processing aids during shaping, and ensuring the completed
28 items' long-term performance. Additives are used in a variety of ways, including terminology that
29 aims to reflect their effect. The majority of the additives possess low molecular weight, and other
30 impact modifiers exist as polymers. In most cases, the additives are not chemically linked to the
31 polymer networks. They may have a propensity to migrate into materials that come into contact
32 with the plastic surface depending on temperature, concentration, and time [161,162].
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55 **Table 1.** Class of additives and their role.
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Class of Additives	Examples	Role	Ref.
Antioxidants and Stabilizers	Calcium stearate, 2-Mercaptobenzimidazole (MB), Dibutyltin maleate, Inorganic lead sulfates.	Prevents oxidation in recycling. Delay thermoplastic deterioration. Stabilizers protect thermoplastics from UV light and heat. Thermal, oxidative, and mechanical reactions are inhibited by antioxidants.	[163,164]
Mineral fillers	Calcium carbonate, Dolomite, Barite, Talc.	Enhances thermoplastic properties and saves money. Surface product gloss improvement. Improves mechanical and thermal properties.	[165,166]
Compatibilizers	Ethylene–ethylacrylate (EEA), Ethylene–butylacrylate (EBA), Maleic anhydride, Glycidyl methacrylate	Improves recycling compatibility of mixed plastic waste. They form reactive bonds with polymers, increasing compatibility. These can also improve recycled thermoplastics' mechanical properties.	[167,168]
Processing aids	hexafluoropropylene-vinylidene fluoride copolymer, Fluoroelastomers based processing aids.	Processes polymers better. Processing aids enable thinner extrusions and shorter moulding cycles. It also reduces plastic melt fracture.	[169,170]

Impact modifiers	Methyl Methacrylate Butadiene Styrene polymer (MBS), chlorinated polyethylene (CPE), ethylene-vinylacetate (EVA).	These are elastomeric materials that absorb and dissipate impact energy. It has the properties of a rigid part. It also increases thermoplastic resin toughness and durability.	[171,172]
Flame retardants, Antistatic agents	Halogenated compounds, Melamines, Aliphatic amines, Ethylene glycols.	Flame retardants are used to prevent or delay the combustion process. They will reduce static electricity buildup on the plastics.	[173,174]

Tsenoglou et al. [175] analyzed the importance of stabilizing additives in the melt recycling of consumed HDPE plastic bottles. For the restabilization process, the granulated hindered amine stabilizer (HAS) having a higher molecular weight was utilized. Reprocessing begins with remelting in the first stage without any stabilizer. The second stage was carried out with 0.2% w/w and final remelting with 0.4% stabilizer. The degradation appears to be almost linearly proportional to the degree of processing and reduces when the concentration of the restabilization agent is increased. The authors also developed an analytical model to predict the optimum stabilizing agent concentration to improve the stability of recycled material. The recycling of the PP/PA polymer blend with ammonium polyphosphate (APP) flame retardant resulted in a decrease in flammability properties, but the obtained properties were better than the virgin PP [176]. Leng

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3 and coworkers [177] employed triethylenetetramine to degrade PET, which they subsequently
4 used to alter recovered asphalt. They discovered that adding a terminal amine group to PET's
5 recycled additives aids in the bonding of the binder and aggregate, and as a result, the PET-
6 modified blends did not produce stripping even when large percentages of recovered asphalt
7 pavement were used. The recycling of PP waste from the automotive industry was reported by
8 Bleviso and coworkers [178] using inorganic fillers such as zeolite and sepiolite. To maximize the
9 effect of sepiolite and zeolite addition on the mechanical characteristics of waste PP materials,
10 pellets of the new compounds should be prepared to provide better dispersion and homogeneity of
11 the added fillers with the PP matrix. This should be accomplished through the establishment of a
12 well-organized and regulated recycling process that begins with the granulation of PP wastes, the
13 inclusion of mineral fillers, blending and homogeneity, melting, extrusion, and the final production
14 of recycled pellets. According to the statistics, the use of a small amount (5%) of sepiolite or LTA
15 zeolite enhances some mechanical and rheological characteristics of waste PP recycled materials.
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17 Oksuz and coworkers [179] investigated the mechanical, thermal, and microstructural
18 characteristics of polymer blends (EPDM) composed of isotactic polypropylene and varying
19 amounts of talc (3-15 wt%). It was reported that the yield and tensile strength of the polymer blend
20 were improved with increasing talc percentage up to 6 wt%, but the elongation at break decreased
21 with increasing clay content. However, with the use of talc, the elongation at break value was
22 significantly decreased. Moreover, the talc addition reduced the MFI of the i-PP/EPDM polymer
23 blend. PET recycling via glycolysis with the help of fly ash fillers was conducted by Phung et al.
24 [180] by varying fly ash contents of 50, 100, 150, and 200 phr and evaluating the properties of
25 recycled material. The results showed an increment in mechanical properties for 150 and 175 phr
26 of fly ash content, and they suggested this combination for commercial plasterboard application.
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3 The absence of porosity and a smooth surface was observed in plasterboard morphology.
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5 Moreover, the fly ash additives prevent the penetration of water molecules and act as a barrier to
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7 chemical attacks. Electrical and electronic equipment waste (WEEE) is composed of complex
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9 copolymers including carbon black as well as other mineral materials. They were all identified by
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11 Vazquez et al. [181] and studied the effect of compatibilizers. These minerals have the ability to
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13 alter the mechanical or morphological properties of basic polymers. Physical blends using HIPS
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15 as the primary component exhibited an advantage of "self-compatibility," given the possibility of
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17 direct recycling of mixed WEEE plastics from these types of blends. Thumsorn et al. [182]
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19 analyzed the flame retardancy of recycled PP and PET polymer blends with cockleshell-based
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21 CaCO_3 (CS) renewable filler materials. The results revealed that the addition of commercial and
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23 natural CS improved the stiffness, impact performance, and thermal stability of the polymer
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25 blends. This potential was optimum for the polymer blend with higher CS filler contents. The
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27 thermal oxidative degradation activation energy was found to be proportional to the filler size and
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29 dispersion of RPP particles in the polymer blends. Skrockien and his coworkers [183] investigated
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31 the influence of plasma-treated bentonite filler on the thermoplastic polyurethane blend. The
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33 results confirm the improvement of mechanical properties in the polymer blends, and plasma
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35 treatment improved the dispersion of fillers in the polymer blend. Whereas the unmodified
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37 bentonite filler exhibited poor reinforcing ability because of lower surface activity and the
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39 existence of complete exfoliation difficulties in the polymer blends, The advantage of unmodified
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41 bentonite was the improvement of crystallinity in polymers, which was about 1.4 times more than
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43 the modified fillers' effect. Moreover, the modified filler reinforced polymer blend exhibited 19%
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45 more tensile strength than the unmodified filler reinforcement. The bitumen modification with
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47 both recycled and unrecycled polymer blends (LDPE/EVA, EVA) showed improved binder
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3 rheological characteristics, especially with higher in-service temperatures. In this track, the 5 wt%
4 bitumen modification showed a maximum linear viscoelasticity moduli in EVA/LDPE yield for
5 different temperature ranges [184]. Optical brighteners are the new class of additives that are being
6 used to avoid the formation of yellow color due to usage and aging. These are commonly preferred
7 for post-consumed plastics, specifically PE, PMMA, PU, and PVC. The selection of an optical
8 brightener that is acceptable for material is dependent on a number of parameters, including light
9 fastness and the requirement for the whitening effect. A fluorescent compound's structure must
10 have an aromatic heterocyclic or aromatic condensed ring system in order to be employed as an
11 optical brightener. Inorganic brighteners are capable of absorbing electromagnetic spectrums in
12 the visible range, and the best example of an inorganic brightener is titanium pigments [185,186].
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27 **5.0 Literature on Recycled Thermoplastic Composites**

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29 Many research papers on the use of recycled thermoplastics for composite fabrication have been
30 published in the literature in relation to the above-mentioned recycling techniques. Some of the
31 interesting literature is tabulated in **Table 2**. An interesting study was reported in the literature
32 regarding the use of recycled HDPE and PP plastics with sawdust powder. This work is evident in
33 the use of recycled WPCs for exterior applications with lower moisture absorption and swelling
34 behavior. Furthermore, the results for tensile strength and absorption behavior were comparable
35 to those of virgin plastic reinforced composites [187]. Another study also revealed that the use of
36 compatibilizing agents in recycled HDPE/wood flour composites improved both the physical and
37 mechanical properties of the resultant WPCs. Moreover, the utilization of polyethylene-grafted
38 maleic anhydride (PE-g-MA) strengthened the interfacial bonding in the composites, which
39 resulted in enhanced tensile and flexural properties [188]. The reinforcement of wood and bagasse
40 natural fibers in recycled HDPE resulted in reduced composite thermal stability, crystallization
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3 peak temperature, and a wider range of crystalline temperature as compared to neat recycled
4 HDPE. The added coupling agents (carboxylated polyethylene, maleated polyethylene, and
5 titanium-derived mixture) enhanced the compatibility between natural fibers, and some of the
6 agents played a lubricant role. Positively, the mechanical properties of the recycled HDPE/natural
7 fiber composite were better than those of the virgin HDPE/natural fiber composite. As reported in
8 this work [189], the influence of the coupling agent on the thermal degradation temperature of the
9 composite was very small. The reinforcement of corn husk fibers (125 μ) into recycled LDPE
10 resulted in improved mechanical properties (modulus, tensile strength), moisture absorption, and
11 thermal stability of the resultant composites. The tensile strength was shown to grow with
12 increasing fiber proportion up to 10%, then slightly drop but remain greater than non-loading R-
13 LDPE. The flow and film development of the RLDPE in the composite structure was credited with
14 the increased strength. The drop in tensile strength with higher fiber loading (15%, 20%) might be
15 attributable to a reduction in melted R-LDPE flow in the composite. It's worth noting that internal
16 bond strength is strongly influenced by density, and therefore enhanced internal bond, density, and
17 composite strength are all linked [190]. Gebremedhin and Rotich [191] fabricated a pineapple leaf
18 fiber-loaded recycled LDPE composite for the wall tile application. The best PALF length for
19 producing PALF-LDPE composites with maximum characteristics was discovered to be 30 mm.
20 Because of the establishment of linkage between the OH groups in fibers and water, the moisture
21 absorption by the composites was increased as the PALF weight percentage increased. The
22 acetylation and alkali treatment of natural fibers improved their adhesion with recycled
23 thermoplastics, and hence improved the potential of the resultant composites [192]. AlMaadeed
24 and coworkers [193] analyzed the mechanical and thermal properties of wood flour/glass fiber
25 reinforced recycled PP composites that were produced using the injection molding process. The
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degree of crystallinity of the PP polymer was reduced when the wood particle concentration in the PP was increased. When the glass fibers were added, the composites' tensile strength improved as the percentage of crystallinity increased. Because there is no agglomeration in this scenario, the improvement in mechanical characteristics with increasing crystallinity percentage (and decreasing lamellar thicknesses) may be attributed to the confined area between the lamellae.

Table 2. Literatures on recycled thermoplastic composites.

Recycled thermoplastic composites	Coupling agent	Manufacturing technique	Inference	Ref.
Recycled PP/wood flour	Maleic anhydride-grafted polypropylene	Melt blending, Extrusion	The rPP/RWF composites with 1 wt.% UV stabiliser showed less lightness change and relative loss of hardness, flexural strength, hardness, and maximum strain than those without UV stabiliser.	[194]
Recycled LDPE/pine wood waste (PWW)	Polyethylene-grafted maleic anhydride	Extrusion, Injection molding	The addition of PWW fibres increased the mechanical properties of the LDPE matrix, resulting in greater resistance to photochemical ageing than unfilled LDPE. PWW must be able to absorb most UV rays to	[195]

			protect the LDPE polymer matrix.	
Recycled LDPE/Bean Pod Ash	-	Extrusion, Compression molding	The addition of bean pod ash particles to the RLDPE polymer enhanced the percentage of water absorption while also improving the composites' modulus, stiffness, and hardness characteristics. It took 20 wt% BPAp to make the tensile and flexural strengths reach their maximums of 20.1 and 39.0N/mm ² .	[196]
Recycled polystyrene/ Wood flour	Poly(styrene- co-maleic anhydride) oligomer	Extrusion, Injection molding	A 2 wt. % coupling agent increased the interfacial adhesion between polystyrene and wood flour, as well as the lignocellulosic composites' flexural and impact strengths. Beyond 2 wt. % coupling agent reduced mechanical properties.	[197]

1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20 21	Recycled PVC/Rice husk	N- (n-Butyl)- 3- aminopropyl- trimethoxy- silane	Dry mixer followed by hot compression molding	SEM images show that silane treatment increases adhesion of recycled PVC to rice husk. The improved tensile properties of the recycled PVC-based composite result from the use of rice husk reinforcement and surface treatment.	[198]
22 23 24 25 26 27 28 29 30 31 32 33 34 35 36 37 38 39	Recycled PP/bamboo	Maleic anhydride grafted polypropylene	Extrusion, Injection molding	In composites, alkali treatment increased bamboo fibre contact area, resulting in more uniform fibre dispersion in the PP matrix. The coupling agent improved the adhesion and mechanical properties of PP composites.	[199]
40 41 42 43 44 45 46 47 48 49 50 51 52 53 54 55 56 57 58 59 60	Recycled PP/Curaua fibers	Maleic anhydride grafted polypropylene (MAP 504)	Extrusion, Injection molding	Fibers of shorter lengths (0.01–0.4 mm) were fed into the extruder, resulting in better fiber dispersion and improved mechanical characteristics of the composites. The fibers were partially fibrillated during the	[200]

			extrusion process, which improved their aspect ratio.	
Recycled PP/kenaf/fiber/rice hulls	Maleic anhydride grafted polypropylene	Melt mixing and compression molding	The composites made from recycled PP have equivalent flexural strength and thermal stability to those made from neat PP. Composites reinforced with kenaf fibers demonstrated superior characteristics to those reinforced with rice hulls.	[201]
Recycled LDPE/Pennisetum Purpureum Fibers	Polyethylene-graft maleic anhydride	Extrusion, Injection molding	These natural fibers have a high aspect ratio and were easily extruded into LDPE waste. Incorporating P. purpureum fibers into LDPE waste resulted in a considerable rise in the material's Young's modulus as a proportion of the fiber concentration.	[202]
Recycled HDPE, PET/rice	Ethylene-glycidyl methacrylate, Polyethylene-	Extrusion, Compression molding	The mechanical characteristics of the compatibilized blend and RH nanocomposite were increased by up to 40% and	[203]

1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18	husk/organo clay	grafted maleic anhydride		34%, respectively. Another significant enhancement of the flame-retarding effect was found, in which the burning rate was declined by 29-37% through matrix compatibilization.	
19 20 21 22 23 24 25 26 27 28 29 30 31 32 33 34 35 36 37 38 39 40 41 42 43 44 45 46 47 48	Recycled PP/talc/ EPDM/ bamboo	Maleic anhydride grafted polypropylene	Extrusion, Injection molding	Bamboo fiber boosted modulus, tensile and flexural strengths, and fatigue life substantially, while decreasing impact strength and elongation at break values. The compatibilizer had a beneficial influence on only tensile and flexural strength, as well as fatigue life. The combination of coupling agent and fiber at concentrations more than 2 wt% increased the fiber's degradation temperature.	[204]
49 50 51 52 53 54 55 56 57 58 59 60	Recycled PP/hemp	MAPP Epolene PMG-3003	Extrusion, Injection molding	The results indicate that the inclusion of MAPP improves adherence within the composite.	[205]

			Generally, the fiber quantity and size of the composites had a significant effect on their torsion, tensile, and flexural, characteristics. Although, the addition of MAPP increased the impact properties of composites, the results were always less than those of plain PP.	
Recycled LDPE/olive pits (OP)	-	Extrusion, Compression molding	This investigation revealed that both NaOH and water/DMSO treatment of OP resulted in improved compatibility, but in compared to the water/DMSO treatment, the NaOH treatment exhibited better physical and mechanical characteristics.	[206]
Recycled PP/kenaf fiber/halloysite	Maleic anhydride grafted polypropylene	Solution process followed by hot compression	The kenaf fiber reinforcement improved the overall mechanical strengths due to more resilient and stronger than the halloysite which is more brittle in nature. PP containing a	[207]

			non-polar methyne group capable of reacting with the hydroxyl groups in KF's cellulose.	
Recycled LDPE/wheat straw	Polyacrylate latex (PAL), Blends of PAL and polymethylene polyphenylene isocyanate (PAPI)	High intensity mixer followed by wood moulding	While the PAL was capable of improving composite quality, the coupling agent made of PAPI and improved PAL produced a more substantial improvement. Due to the coupling agent's loaded level of 30/70 PAPI/PAL was 4.5 wt%, the composite exhibited the greatest internal bonding (IB) strength and the IB strength beyond 2 hours of immersion in boiling water.	[208]

6.0 Market Opportunities for Recycled Thermoplastics

By and large, virgin polymers exhibit superior qualities to recycled polymers and the market price of the former acts as a natural ceiling on the price of the reused material. This price differential is a critical aspect in determining the commercial significance of recovered polymers. The price of virgin materials is determined by the price of oil, which has risen significantly in recent years. The

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3 latter problem has increased the economic viability of recycling [209,210]. Current recycling
4 techniques require more development before they can be commercialized. To extend the market
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6 for recycled materials, we need more affordable, less polluting, and efficient recycling methods,
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8 as well as remanufacturing technology for high-performance goods. The current opportunities and
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10 applications of recycled thermoplastics/composites in different sectors are discussed in the
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12 following subsections.
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20 **6.1 Food and Packaging industries**

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22 Global population expansion is increasing demand for food supply and, as a result, the use of food
23 packaging materials. Packaging protects food from external influences such as physical, chemical,
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25 and biological activities. The packaging business needs lightweight materials to minimize the
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27 quantity of material necessary to package a product, hence lowering the product's weight and
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29 transportation costs, as well as decreasing the amount of packaging waste material at end-of-life
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31 [211]. As a strategy to reduce trash dumped into the environment, the packaging industry has
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33 started to include recyclable plastics in its packaging materials. However, recycling plants must
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35 keep impurities in reformed plastic at levels that are suitable for the packaging's intended usage.
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37 This has proven challenging, costly, and time-consuming, as the collected plastic garbage is
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39 composed of a variety of different plastic kinds that must be processed and segregated. Globally,
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41 single-use plastic packaging materials have poor recycling rates, with just 14% of plastic
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43 packaging waste materials collected for recycling and only 5% effectively recycled into new
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45 plastics [212,213]. The European Union has advocated modifications to packaging waste
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47 guidelines that require 75% of packaging waste to be recycled by 2030. Recycled plastics of
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49 excellent quality cost between 60% and 70% of the price of virgin plastic, but their value decreases
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3 as their quality deteriorates due to frequent recycling [211]. Due to its infinite recyclability, the
4 usage of recycled PET (R-PET) as a substitute for "virgin" PET or glass-made products has
5 increased dramatically over the previous decades. Numerous businesses sell bottles comprising
6 between 50% and 100% R-PET, accounting for around 1.4 million tonnes of the packaging market
7 in 2018 [214]. Only PET and HDPE bottles are recycled at a rate greater than 40%, whereas the
8 majority of other packaging is recycled at a rate of less than 10%, owing mostly to collecting,
9 sorting, and recycling issues with mixed plastics. The sorption characteristics and diffusion
10 behavior of various polymer classes or individual plastics influence the interaction between
11 packaging plastics and products. As a result, plastic inertness is the most important factor in
12 determining whether or not packaging plastics can be recycled closed-loop [20,213]. When PET
13 vessels are reprocessed and cleaned using sun-based water cleaning, the plastic filters additional
14 pollutants after being exposed to UV for 6 to 9 hours while filled with water. They also detected
15 an increased infiltration when the containers were exposed to UV radiation and then warmed to
16 60° C. PET bottles are recycled to make new PET bottles, as well as plastic sheets, film, and
17 transportation strapping. Polyester fibers and cloth are also made from them [215]. The liquid
18 packaging boards (LPBs) are comprised of approximately 21% PE, 75% cardboard, and 4%
19 aluminum. The cardboard layer is designed to give stability, while the PE materials guard the
20 package against moisture on both the outside and inside, and the metal functions as a gas barrier.
21 In Germany, 178 kt of LPBs were utilized in 2016 [216].
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50 **6.2 Construction and Building sector**

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52 With a market share of 20%, construction is the second most common use of plastics. Construction
53 materials like pipes, insulation materials, frameworks, etc., and interior building components like
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3 mattresses, carpeting, blinds, etc., in contrast to packing, have a typical lifespan of more than 10
4 years. As a result, manufacturers' efforts in the field of eco-friendly design of such goods can only
5 have a long-term impact on recycling. Roof tiles for flat green roofs are already available on the
6 market and are frequently produced from recycled PP or MPO, as they have an extremely low
7 load-bearing capacity if any [217,218]. From a broad perspective, recycling waste PET plastics
8 can make a major contribution to the economy and the environment. It contributes to altering the
9 physical and engineering qualities of construction materials such as asphalt and building concrete,
10 as well as reducing the exploitation of natural resources to reduce environmental pollution and
11 save money and energy [219]. There are many advantages to using recycled plastic waste as a
12 construction material. Because plastic waste is primarily derived from municipal waste, it is
13 predicted to be a low-cost construction and building raw material. Moreover, the plastic waste
14 utility in construction materials not only adds value to the plastic waste but also motivates the
15 industry more towards recycling plastic waste. Because plastic garbage is readily available in all
16 places, the cost of transportation for raw materials will be lower than for other resources [220].
17 Laria and coworkers [221] studied the processing and mechanical properties of recycled LDPE-
18 HDPE and PET composites for building component applications. In tensile, flexural, and
19 compressive tests, the mechanical resistance was roughly 60% that of virgin material. The
20 nonstructural materials standards based on the CIRSOC normative were met by the components
21 made using this plastic composite. Vasudevan et al. [222] investigated the use of waste plastics in
22 the manufacture of flexible pavements. They concluded that coating aggregates with polymers
23 reduced moisture absorption, porosity, and increased soundness. They discovered that using waste
24 plastics for flexible pavement was one of the most effective ways to dispose of waste plastics
25 quickly. The recycled unplasticized polyvinyl chloride (UPVC) was utilized for the manufacturing
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3 of tiles and plumbing pipe fittings. Whereas recycled plasticized polyvinyl chloride (PPVC) was
4 used for industrial flooring [223]. In tandem with the ever-expanding asphalt business, road
5 pavement specialists and scientists are developing new strategies and methods to improve
6 construction and maintenance processes to reduce environmental damage. Studies on the use of
7 plastics on roads have discovered significant advantages in the road's physical and mechanical
8 qualities, such as tensile strength, water resistance, durability, and overall life expectancy
9 [224,225]. Mondal et al. [226] reported the use of energy-efficient recycling of mixed waste
10 plastics as a construction material. The strong heat resistance of waste plastic impregnated bricks
11 might provide economic value to brick makers, motivating them to set up the necessary logistics
12 for collecting and using all sorts of waste thermoplastics. Bricks with a higher percentage of
13 plastics contain larger voids and are thus more thermally resistant, even though their compressive
14 strength decreases. Another study reported by Abu-Saleem and coworkers [227] reveals the
15 efficiency of mixed recycled plastic waste (PET, HDPE, PP) as a concrete performance booster
16 for kerb applications. When compared to the control mixed plastic waste, PET showed acceptable
17 drying shrinkage. Microstructural investigations revealed that PET-containing blends had higher
18 binding strength than HDPE and PP. Due to a weaker bond between the plastic surface and the
19 cement paste, increasing the replacement ratio of plastic aggregate decreased the RPC's mechanical
20 strengths and static chord modulus of elasticity. There was also an interesting literature review
21 regarding the use of recycled polystyrene as a potential substitute for sand in cement mortar [228].
22 Due to its huge scale and existing market for building materials, construction might be considered
23 a potentially substantial and promising area for the employment of recyclates.
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6.3 Transportation sector

The growing demand for thermoplastics as a lightweight material for automobiles and other commercial applications necessitates additional research into the polymer resources available. Oladele et al. [229] developed an eco-friendly composite using recycled waste plastic and snail shell particulates (SSP) for automobile floor panel or door sill applications. The mechanical properties were enhanced within 3–9 wt% of SSP, while wear and water-repellent potentials were greatly boosted with the addition of 15 wt% of SSP. This research indicated the possibility of bio-filler-recycled waste plastic composites for most automotive interiors with reasonable wear, mechanical, and water resistance qualities. Automakers are putting in significant effort to reduce car weight by altering the materials used. Automobile manufacturers must also consider the environmental impact of the vehicle's entire lifespan, including raw material supply, vehicle production, utilization, and waste disposal, due to strict guidelines and legislation in the European Union and Asian countries regarding automotive end-of-life requirements [230,231]. PET recycling is not restricted to the manufacturing of bottles and containers but is also recycled as a fiber in automobiles for seat cover applications. These recycled PET fibers are also ideal for use as "covering textiles" in less contactable regions like peripheral zones and door panels. These fabrics, obtained from waste PET, exhibited resistance to aesthetic wear, tear strength, and structural properties required for seat cover application [232]. The combination of recycled PP and thermoplastic vulcanizates (TPV) blends is popular nowadays in Thailand for the automotive OEM (original equipment market) to reduce the import cost of TPV. All blends may have their mechanical qualities modified by adjusting the recycled PP percentage, and the mechanical properties allow for commercial soft car component applications [233]. The usage of mixed plastic waste (PP, PET, HDPE) with ethylene glycidyl methacrylate compatibilizer as a raw material for

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3 automobile bumper production is a current trend in the automotive industry. Their strength and
4 mechanical properties depend on the compatibilizer quantity. The properties obtained with the
5 mixed plastic blend were more than the individual plastic reinforcements [234]. Bello et al. [235]
6 fabricated an automobile bumper prototype using recycled LDPE and eggshell nanoparticles. The
7 added nanoparticles boosted the structural properties and load-bearing capacity of the
8 thermoplastic composite. This is one of the most cost-effective methods of lowering raw material
9 costs without sacrificing component properties. Bruijn and van Hattum [236] manufactured the
10 world's first completely recycled thermoplastic (Polyphenylene sulfide (PPS)) based composite
11 for rotorcraft access panel applications in the aerospace industry. The redesigned rotorcraft access
12 panel is lighter than the original component, has a lower production cost, and is more
13 environmentally friendly due to the use of an energy-efficient technique and recycled materials.
14 Weight is very important in this type of application, and it is one of the main reasons this type of
15 material is used.
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32 33 **6.4 Indoor applications**

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35 Electronic waste has been noted to comprise a broad and rising spectrum of electronic gadgets in
36 recent years, ranging from big home appliances such as refrigerators, and air conditioners to
37 consumer electronics such as laptops and cellular phones. Furthermore, following the system's
38 introduction, the recycling yield has risen. The only plastics utilized in interior applications are
39 those that can be recycled from high-grade plastics. Some of the recycled thermoplastics such as
40 polystyrene and polypropylene are currently being used in the manufacturing of television back-
41 cabinets, foundation frames, nets for air conditioners, and dewatering tanks [237,238].
42 Additionally, polypropylene goods are recycled in washing machine water tanks, safety
43 equipment, and refrigerator fresh food containers. Due to their size and average plastic content of
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3 19% by weight, large home appliances (LHA) contain rather large plastic parts, and the presence
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5 of brominated flame retardants appears to be less significant in comparison to other electronics
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7 and electrical equipment (1.5% vs. 60% in ICT equipment). The target is based on material-
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9 specific recycling. Belgian experience with a 50% by weight objective for the recovered plastic
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11 fraction leads to taking into account the average plastic content of LHA (19% by weight) in a target
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13 suggestion of 9.5% by weight for the weight of the product [239]. The PP recycling with the help
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15 of all recycling processes covering washing and pelletizing showed better results in impact strength
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17 and ultimate elongation value. Hence, these results suggest the decrement of toughness value
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19 because of contamination. By observing all these properties, the recycled PP could be used for the
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21 external unit panels of the room AC, washing machine cleaner parts, and base unit [240]. Another
22
23 interesting study reports the quality assessment of open-loop recycling of PP to check its
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25 applicability for refrigerator plastics. While closed-loop recycling is prohibited, the open-loop
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27 recycling of post-consumer waste PP is not and can be expanded in proportion to the amount of
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29 RPP utilized. The performance study of open-loop RPP reveals that it is a well-balanced plastic
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31 with comparable toughness and strength to Impact-PP and that it is suitable for refrigerator plastics
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33 with good impact properties at low working temperatures [241]. According to Plastics Europe,
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35 mechanically recycled plastics from EEE waste account for 2% of the overall mechanical recycling
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37 of plastics. This material is primarily derived from major residential appliances [242].
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46 **7.0 Challenges in Thermoplastics Recycling**

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48 Effective recycling of heterogeneous waste plastics is the sector's next significant challenge. The
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50 majority of post-consumer plastic waste collection programs focus on rigid packaging due to the
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52 difficulties associated with flexible packaging throughout the collecting and sorting processes.
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54 Recycling rates are traditionally low for a reason. Rather than that, they are a result of a range of
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3 economic and technical constraints that work against the recycling process. Due to the
4 incompatibility of waste plastic mixes in general, effective waste sorting is crucial to the quality
5 of goods obtained by mechanical recycling [243,244]. The additives that hampered the mechanical
6 recycling of waste plastics may also have a detrimental effect on the chemical recycling of
7 monomers via poisoning the added catalysts that permit depolymerization of post-consumer
8 plastics. The chain growth inside the plastics is again a difficult thing during monomer recovery
9 in terms of constrained functionality and increased stability of the plastics. Every plastic category
10 presents some unique technological recycling issues, such as opacity of recyclates, waste stream
11 quality, and compound and multilayer packaging issues [245]. Chemical recycling is mostly
12 constrained to condensation polymers, and energy recovery techniques are not acceptable
13 ecologically. Whereas mechanical recycling is associated with deterioration of properties in the
14 products. The melting point difference in the mixed plastic waste induced partial degradation of
15 constituents and incompatibility issues resulting in poor mechanical properties. Apart from sorting,
16 recycling is concerned with the thermal breakdown of polymers at elevated temperatures and
17 physical shredding during melt processing [29,53]. The blending or adhesion of normal grade iPP
18 and PE is another challenging task in plastic recycling. This tackles one of polymer chemistry's
19 forthcoming great challenges: creating polymers that can be recycled effectively back into their
20 original elements [246]. The recycling of thermoplastic-based composites is the next level's
21 biggest task. For example, the recycling of wood-plastic composites is critical due to the strong
22 interaction between thermoplastics and wood particles. The majority of plastics from scrapped
23 waste are treated in an auto shredder, which may contaminate foreign particles and other plastic
24 waste. Due to the difficulty of quiescent depolymerization, applying mechanical forces might alter
25 the fundamental thermodynamic equilibrium, favoring chain fragmentation during plastic
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3 recycling [244]. Enzyme recycling of PET plastic trash is the most sophisticated recycling method
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5 now available, but microbiological recycling procedures as an end-of-life treatment for
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7 biodegradable polymers are still in their infancy. Chemical additives are frequently used to
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9 strengthen thermoplastic linkages, making them more difficult to melt and, as a result, reducing
10
11 their possibility for reuse. Numerous end users and recyclers of plastics understand that the
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13 polymers used in consumer durables are frequently more valuable than those used in packaging.
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15 However, the recovery of these plastics is hindered by several specific problems, including a far
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17 broader range of diverse and incompatible polymers; a less developed collecting infrastructure; a
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19 greater variety of end products; and lower total quantities of these materials, especially on an
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21 individual polymer grade basis. Plastics are frequently encountered with coatings, paints, and a
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23 variety of coverings (films, sheets) that are commonly seen with durable goods, posing issues for
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25 both recycling and identification. The coatings and paint presence reduced the properties of
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27 recycled plastics due to the stress concentration exhibited by coated particles. There is also a
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29 possibility of plastic chemical degradation while processing because of coating degradation.
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31 Plastics that have been reprocessed could also have left coatings and paint on them that could affect
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33 how they look and how they feel on the surface.
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41 **8.0 Conclusions**

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44 Plastic waste has become a major issue in modern society, and civilization is nearing a breaking
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46 point. Many industrial start-ups have emerged, and major polymer producers have expressed
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48 interest in or taken steps toward recycling. Environmental and economic impacts of recycling were
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50 reviewed, as were sustainable recycling technologies such as mechanical, chemical
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52 (dissolution/precipitation, solvolysis), thermo-chemical (pyrolysis, microwave-assisted pyrolysis,
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3 fluidized-bed pyrolysis, and gasification), and enzymatic. This review also discusses additives and
4 their roles in thermoplastic recycling. Researchers have used recycled thermoplastics as matrices
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6 to make natural and synthetic composites, which were studied in this study. This study found a
7
8 large market for recycled thermoplastics in food and packaging, construction and building,
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10 transportation, and indoor uses. Despite all of these great sustainable technologies, the current
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12 review report highlights a few challenges. To achieve a successful and profitable recycling process
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14 that produces more acceptable polymers, more research must be conducted on all aspects of
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16 recycling. Plastic pollution has made recycling a critical ecological and economic concern.
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18 However, high recycling costs, low energy costs, and low landfilling costs are limiting recycling
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20 growth. Now, mechanical recycling seems to be the most cost-effective and environmentally
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22 friendly option. As a result, mechanical recycling of mixed plastic waste is feasible if the waste is
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24 compatible. For a circular economy with higher recycled quality, chemical recycling is seen as an
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26 appealing technological approach. Pyrolysis is currently the least preferred method of chemical
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28 recycling, but advances in catalyst and reactor design may lead to pyrolysis that produces
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30 monomers. Also, developing methods like plasma-assisted pyrolysis can produce extremely high
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32 monomer yields. In other cases, microwave-assisted thermochemical recycling outperforms
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34 conventional organic synthesis. A major issue is that combining polymers often results in phase
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36 inversion and poor composites due to chemical incompatibility. Most thermoplastic polymers, like
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38 PP and PE, resist enzymatic degradation due to hydrophobicity, inertness, and stability. Two major
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40 strategies can reduce the negative effects of recycling processes: restabilization during
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42 reprocessing to prevent or slow degradation, and inclusion of modifiers or additives that can
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44 improve performance without increasing material costs or recycling costs. The use of recycled
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46 plastics in composites increases the economic and environmental benefits of new materials.
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3 Almost every industry now allows the use of recycled thermoplastics in product development.
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5 Research chemists and chemical engineers have a bright future developing new and better
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7 procedure for recycling a wide range of commonly used plastic materials.
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11 **Declaration of Competing Interest**

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13
14 The authors declare that they have no known competing financial interests or personal
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16 relationships that could have appeared to influence the work reported in this paper.
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