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REVIEW ARTICLE

Plastic recycling: Challenges and opportunities

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Abstract

This review offers an in-depth exploration of current strategies for recycling plastic waste, focusing on mechanical, chemical, and energy recovery methods. It situates these strategies within the context of modern practices by examining ongoing research methodologies and specific case studies related to various types of plastic waste. The global crisis of plastic waste, along with various pre-treatment methods, is thoroughly discussed. The section on mechanical recycling details the processes applicable to different plastics, highlighting key challenges such as thermo-mechanical issues, the use of fillers to enhance certain properties, and material degradation over time. This discussion includes polymers such as polyethylene terephthalate (PET), low-density polyethylene (LDPE), high-density polyethylene (HDPE), polypropylene (PP), and polystyrene (PS). Chemical recycling is analyzed through advanced techniques like pyrolysis, catalytic pyrolysis, solvolysis, and gasification, presenting the state-of-the-art in this field. Additionally, the review touches upon energy recovery and the challenges associated with it. Conclusively, the study delves into the applications of recycled plastics and outlines future challenges. Overall, this

Abbreviations: 3D, three dimensional; ABS, acrylonitrile-butadiene-styrene; AHP, advanced hierarchy process; APAP, acetaminophen; ASTM, American Society for Testing and Materials; BG, butylene glycol; BHET, bis-2-hydroxypropyl terephthalate; CMC, critical micelle concentration; CTAB, cetyltrimethylammonium bromide; DEG, diethylene glycol; DMT, dimethyl terephthalate; DPG, dipropylene glycol; DTAB, dodecyltrimethylammonium bromide; EB, elongation at break; ECO, epoxidized castor oil; EDG, electron donating groups; EG, ethylene glycol; EPDM, ethylene propylene diene monomer; EPS, expanded polystyrene; FFF, fused filament fabrication; GO, graphene oxide; HC, hydrocarbon; HDPE, high-density polyethylene; HIPS, high-impact polystyrene; HTL, hydrothermal liquefaction; IAS, intentionally added substances; IR, infrared; LDPE, low-density polyethylene; LIBS, laser-induced breakdown spectroscopy; MAD, methanol assisted depolymerization; MAH, monocyclic aromatic hydrocarbons; MAP, microwave-assisted pyrolysis; MCO, maleated castor oil; MD, magnetic density; MFI, melt flow index; MIBC, methyl isobutyl carbinol; MIR, mid-infrared; MWD, molecular weight distribution; NIAS, non-intentionally added substances; NIR, near-infrared; NMR, nuclear magnetic resonance; PAH, polycyclic aromatic hydrocarbons; PB, polybutadiene; PC, polycarbonate bisphenol A; PCB, polychlorides biphenyls; PCDF, polychlorinated dibenzofurans; PCR, postconsumer recycled; PE, polyethylene; PET, polyethylene terephthalate; PG, propylene glycol; PHA, polyhydroxyalkanoate; PI, polyimide; PLA, polylactic acid; PMMA, polymethyl methacrylate; PO, polyolefins; PP, polypropylene; ppm, parts per million; PS, polystyrene; PSW, plastics solid waste; PTFE, poly tetra fluoroethylene; PVAc, polyvinyl acetate; PVC, polyvinyl chloride; PVOH, polyvinyl alcohol; RSM, response surface methodology; rPP, recycled polypropylene; rPS, recycled polystyrene; SAN, styrene-acrylonitrile; SBS, styrene-butadiene-styrene; SEB, styrene-ethylene-butylene; SEBS, styrene-ethylene-butylene-styrene; SEM, scanning electron microscopy; SMA, styrene-co-maleic anhydride; SOx, sulphur oxides; SPI, society of the plastics industry; TPA, terephthalic acid; TPE, thermoplastic elastomer; TTAB, tetradecyltrimethylammonium bromide; UTS, ultimate tensile strength; UV, ultra violet; VOC, volatile organic compounds; WEEE, waste electrical and electronic equipment; WF, wood flour; WPC, wood plastic composite; XRF, X-ray fluorescence spectroscopy.

Pradeep Sambyal, Parisa Najmi, and Devansh Sharma contributed equally to this study.

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review aims to provide a thorough overview and practical guidance on the recycling of plastic waste, offering essential insights for further development in this area.

KEYWORDS

chemical recycling, circular economy, mechanical recycling, plastic waste, pyrolysis, sorting

1 | INTRODUCTION

From the top of Mount Everest to the deepest ocean trenches, plastics are omnipresent in every corner of the Earth. This widespread use of plastics has driven economic growth in many countries, largely due to their lightweight nature, tuneable properties, durability, and ease of processing, leading to a wide range of applications. However, the most significant ecological threats arise when the plastic fragments are transported through the atmosphere and oceans and are mistakenly considered food by animals. While the impact of plastics on humans and the ecosystem is yet unclear, plastic pieces have even been found in human blood and breast milk.^[1] This pervasiveness and durability are just one aspect of the global crisis related to the life cycle of plastics, which has reached epic proportions.

Nevertheless, replacing plastics with alternative materials like glass or metal containers is not necessarily an environmentally sustainable solution. In other words, these alternatives are considerably heavier, contributing to higher carbon dioxide (CO₂) emissions during transportation, which might devastate the planet.^[2] On the other hand, the usage of biopolymers and biomass feedstock, thanks to the substantial investment in the past decade, has yielded some progress. Products derived from biomass not only fulfil consumer demands but also degrade within a short span of a few months. Nevertheless, the main drawback lies in their higher cost compared to petroleum-based plastics, and it remains uncertain whether they can adequately substitute specific types of plastics for various applications.^[3] Consequently, much more work is still needed to determine their production scalability and their environmental persistence to realize their potential circular economy process truly.^[4]

Conversely, fossil fuels constituted the primary raw material for over 85% of the plastics produced in 2019, as depicted in Figure 1A.^[5] The rising demand for plastics due to urbanization, increasing wealth, and improved living standards exacerbates the dependency on fossil fuels, aggravating the issue further. Such reliance presents a significant obstacle to environmental sustainability and continues to drive global fossil fuel consumption trends that are both ecologically and economically unsustainable.^[2,6]

Globally, 353 million tonnes of plastic waste was produced in 2019, with approximately half being single-use.^[7,8] Not only was 19% of the generated waste incinerated, negatively impacting air quality, but less than 10% was also recycled.^[9] Almost half of the waste (49%) was disposed of in landfills, and 22% was mismanaged,^[2] burnt in the open, or discarded in the environment through littering or illegal dumping, as illustrated in Figure 1B.

While recycling may appear to be a viable solution for addressing plastic pollution, the low cost of virgin plastics coupled with economically unviable or impractical recycling techniques render it non-incentivizing. However, imposed landfill taxes resulted in an increased recycling rate.^[10,11] For example, China has begun rejecting waste from foreign countries and now focuses solely on recycling its own plastic waste. This pattern is anticipated to expand further, even within plastic waste-importing countries like Malaysia, Thailand, Indonesia, and so forth.^[12–14] A report published by McKinsey states that around 50% of the produced plastic waste worldwide could be reused or recycled by 2030, almost fourfold of the current practices (12%). Additionally, plastics recycling could generate profit-pool growth of as much as \$77 billion by 2031.^[15]

Examining the potential of plastic waste recycling reveals that packaging plastic waste is merely the most visible aspect, much like the tip of the iceberg, as illustrated in Figure 1C. Furthermore, addressing the plastic waste issue requires understanding the primary sources of plastic waste and assessing suitable recovery and recycling technologies.

Table 1 lists a series of recently published review articles that provide information on recycling techniques. In 2017, Ragaert et al. reviewed the chemical and mechanical recycling methods used to manage solid plastic waste.^[16] However, they only focused on the mechanical recycling of polyethylene terephthalate (PET). Vollmer et al., on the other hand, solely concentrated on diverse chemical recycling techniques, including but not limited to pyrolysis and solvolysis, where monomers and oligomers can be re-polymerized.^[2] In 2018, Zhao et al. discovered that mechanical recycling exhibits a reduced environmental burden when evaluating life-cycle

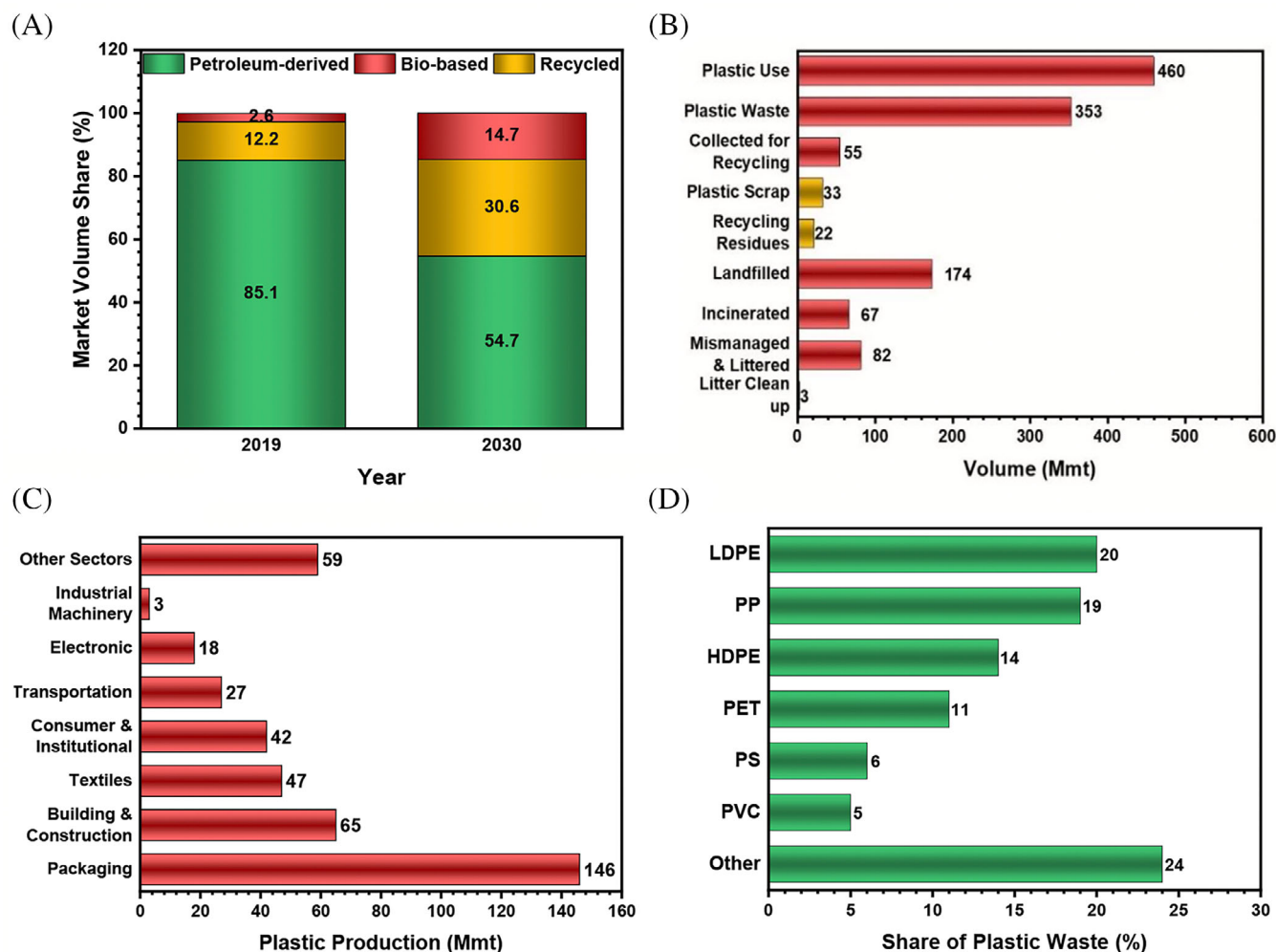


FIGURE 1 (A) Market volume share worldwide in 2019 and its projection in 2030 by type of feedstock, (B) lifecycle of plastic waste worldwide in 2019, (C) global plastic consumption in 2017 by industrial sector, and (D) worldwide distribution of plastic waste in 2015 by plastic type. Source: Statista.^[9,21,22]

assessments with respect to environmental impact, economic factors, and technological performance. Moreover, their literature review revealed that among diverse separation methods, such as flotation and manual sorting, solvent extraction has proven to be more efficient in sorting and separating mixed plastic waste.^[17] In 2021, Schyns and Shaver reaffirmed that mechanical recycling is the most effective method when considering time efficiency, carbon footprint, and environmental impact.^[18] Nonetheless, according to Vollmer et al., significant degradation in the mechanical properties of recycled plastics is caused by chain scission during mechanical recycling. Within the framework of the circular economy, they argued that chemical recycling outperforms mechanical recycling in terms of efficiency. This is due to the fact that the chemical compounds obtained from the chemical recycling of plastic waste serve as polymerization precursors. The resulting polymer demonstrates properties comparable to those of virgin plastics.^[2] Hence, there remains a debate

regarding the efficiency of various recycling techniques for specific types of plastics that need to be addressed.

To the best of our knowledge, no comprehensive summary has yet been provided that covers all recycling techniques, along with the processes of collection, separation/sorting, decontamination, applications, and the appropriate recycling methods for specific types of plastic. Many review articles have been published, each focusing on specific methods such as the exclusive mechanical recycling of particular plastics, chemical recycling, processing of mechanically recycled polyolefins, or strategies for separation.^[2,4,5,8,17–20]

As such, in this review, we aim to address the gap between research and industrially implemented processes. The challenges in plastic recycling are exacerbated by a shortage of collection and sorting facilities, presenting a significant barrier that hinders the widespread adoption of plastic recycling initiatives. In this review, the major focus is on sorting, separation techniques,

TABLE 1 Overview of review papers published on plastic recycling.

Process	Key findings	Year	References
Solvolysis	Solvolysis and re-polymerization of PET	2008	Sinha et al. ^[23]
Mechanical and pyrolysis	Overview of both processes and comparison of their shortcomings, pros, and cons Primarily focused on the degradation of polymeric chains during mechanical recycling Fundamental industrial methods and recycling of specific waste stream	2017	Ragaert et al. ^[16]
Solvolysis and pyrolysis	Catalyst efficacy and financial support were identified as key barriers to commercialization Significance and impact of plastic waste stream on the recycling process	2017	Rahimi and García ^[24]
Pyrolysis	Criticality of reactor design and process conditions in chemical recycling	2017	Lopez et al. ^[25]
Dissolution	Details of appropriate solvents for various polymer types Limitations of dissolution separation for a mixed stream of polymers	2018	Zhao et al. ^[17]
Mechanical	Mechanical recycling emerges as the optimal method in terms of time efficiency, carbon footprint reduction, and environmental impact	2021	Schyns and Shaver ^[18]
Solvolysis, pyrolysis, and gasification	Impact of reaction conditions (reactor type, temperature, and catalyst) with a focus on life cycle assessment	2022	Jiang et al. ^[26]
Solvolysis, pyrolysis, and bio-recycling	Recycling methods for biodegradable plastics like polylactic acid (PLA) Recycling biodegradable plastics, such as PLA, offers a means to mitigate carbon emissions while leveraging existing recycling infrastructure	2023	Kumar et al. ^[27]
Mechanical	Small-scale recycling of PP, PE, and PET demonstrated improved efficiency and quality product outcomes	2023	Uzosike et al. ^[28]
Mechanical and chemical	Mono-material or degradable materials were efficient options to be used in manufacturing packaging The capability for disassembly proved advantageous, particularly in multi-component packaging designs	2023	Ding and Zhu ^[29]
Mechanical and energy recovery	Potentials and limitations of mechanical recycling Inaccessibility, plastic diversity, and contamination were identified as the main reasons for low recycling rates	2023	Klotz et al. ^[30]

mechanical recycling, and various chemical recycling methods, including monomer recovery, pyrolysis, solvolysis, and gasification. Complementarily, the associated recycling technologies and their economics for the five most consumable plastics (Figure 1D), including HDPE, LDPE, PET, PP, and PS, will be discussed.

2 | PLASTIC WASTE COLLECTION

Collection is an indispensable yet underestimated component in plastic recycling. The significance of the collection phase becomes even more pronounced when considering that, globally, only 15% of the total generated plastic waste is collected for recycling—55 million metric tons out of 353 million. Moreover, among the generated plastic waste, a mere 33 million metric tons can progress to the subsequent recycling stages due to deficiencies in proper collection mechanisms. Source collection and post-collection are the primary collection methods, each branching into diverse pathways.

Households bear the primary responsibility of source collection, actively participating in waste collection through various channels, including buy-back services, drop-off locations, and deposit-refund depots. Post-collection is mainly related to the municipalities, requires less infrastructure, and might be more convenient for the citizens, including curbside and vehicular collection methods. Martinho et al. found that mixed collection systems yield a higher recycling rate and reduce contamination levels in collected plastic waste.^[31] Picuno et al., on the other hand, estimated that employing separate collection techniques rather than mixed collection methods could achieve a collection efficiency in the vicinity of 75%.^[32]

While vehicular collection presents a potential alternative to drop-off and curbside collections, its implementation faces challenges such as odour management and the logistical complexity of daily household waste collection in densely populated urban areas, rendering it impractical for large cities.^[33] The comparison of waste collection systems should not be oversimplified, as the

literature offers various indicators to gauge each method's performance.^[34] For example, indicators such as operational costs, social impact, environmental impact, ease of sorting at the facility, and ease of collection by workers/households are some of the valuable metrics for scrutinizing the most optimal method for waste collection. Tin et al. employed the advanced hierarchy process (AHP) method as a multicomponent decision-making method to compare waste collection methods. Their study identified the deposit-refund system as the optimal approach. This system operates as a mutually beneficial mechanism, allowing the public to earn money while assisting governing bodies in collecting waste for more effective utilization.^[33] The shortcoming here arises from the deposit-refund collection method's reliance on consumer participation. Aslani et al. researched individuals' tendency toward bottled drinking water consumption.^[35] Their findings revealed that over 46% of the participants not only advocated the use of bottled water but also recommended its consumption to others. Interestingly, these individuals exhibited higher income levels and did not perceive the disposal of empty bottles as an environmental concern. Thus, encouraging proper plastic waste collection involves more than just raising public awareness. Contextual factors such as lifestyle choices, economic circumstances, and social beliefs also significantly influence individuals toward responsible waste management practices. While deposit-refund and buy-back initiatives have succeeded in encouraging waste returns and reducing littering, it is essential to establish a universally accepted collection strategy and policy, improve collection rates, and reduce the costs associated with sorting and cleaning plastic waste.

3 | PLASTIC WASTE SORTING

Sorting emerges as the second crucial step in recycling plastic waste, as several analyses have indicated that substantial material losses occur during this stage. The loss is primarily attributed to the complexity of the design of materials and the absence of suitable sorting facilities.^[32,36] The quality of recycled products is highly dependent on the effectiveness of the sorting process. Various factors, including type of plastic waste, size, dimensions, density, additives, adhesives, and pigments, influence the choice of a particular sorting method.

3.1 | Manual sorting

Manual sorting is the most economically feasible and extensively employed technique for sorting plastic waste.

It involves identification based on criteria such as type, shape, appearance, and Society of the Plastics Industry (SPI) codes. It provides visual cues for operators to effectively categorize materials, including separating plastic from contaminants (e.g., metal, glass) or sorting different types of plastic. Despite its viability and cost-effectiveness, manual sorting is carried out by the labour force. Thus, the potential human error cannot be overlooked.

3.2 | Density sorting

The sink-float or gravity (density) sorting technique (Figure 2A) can be employed in the separation of plastics based on the disparities in the density, thanks to its easy processing, high-loading capacity, automatic nature, and cost-effectiveness.^[37] Despite all the advantages of the sink-float technique, its efficiency is somewhat compromised by its inherently slow processing speed and environmental concerns due to the disposal of the densification medium. Additionally, due to its reliance on density disparities, the technique encounters limitations in its applicability to a wide variety of plastics, particularly those with identical density profiles. A mixture of small pieces of plastic waste with different densities was dispersed in a medium, resulting in less dense components floating and denser ones sinking.^[38] The selection of a suitable densification medium, along with the identification of parameters such as reagent concentration, pH, conditioning period, and particle size, plays a critical role in plastic flotation.^[39] For example, polyolefins such as LDPE, HDPE, and PP, with densities in the range of 0.89–0.96 g/cm³, float in water (density of 1 g/cm³), whereas polyvinyl chloride (PVC) and PET, with an average density of 1.38 g/cm³, will sink. Typically, high-density polymers are treated with water, but saline solutions are also favoured for polymer separation because of their precise density control, resulting in excellent outcomes.^[40]

Meneses Quelal et al. used tap water, ethanol, and sodium chloride solutions at different concentrations as densification mediums to sort high-density polymers (PS and ABS) and low-density polymers (HDPE and PP). The findings demonstrated that a 23% v/v ethanol solution successfully achieved complete separation of PP and HDPE, whereas a 40% w/v sodium chloride solution was more effective for the separation of PS and ABS.^[41] Pongstabodee et al. studied a three-stage sink float method to separate high- and low-density polymers, where 30% w/v calcium chloride solution was utilized to separate PET/PVC from PS/ABS.^[42] For the selective separation of each plastic material, surface treatment of one or two species was recommended. The results indicated that

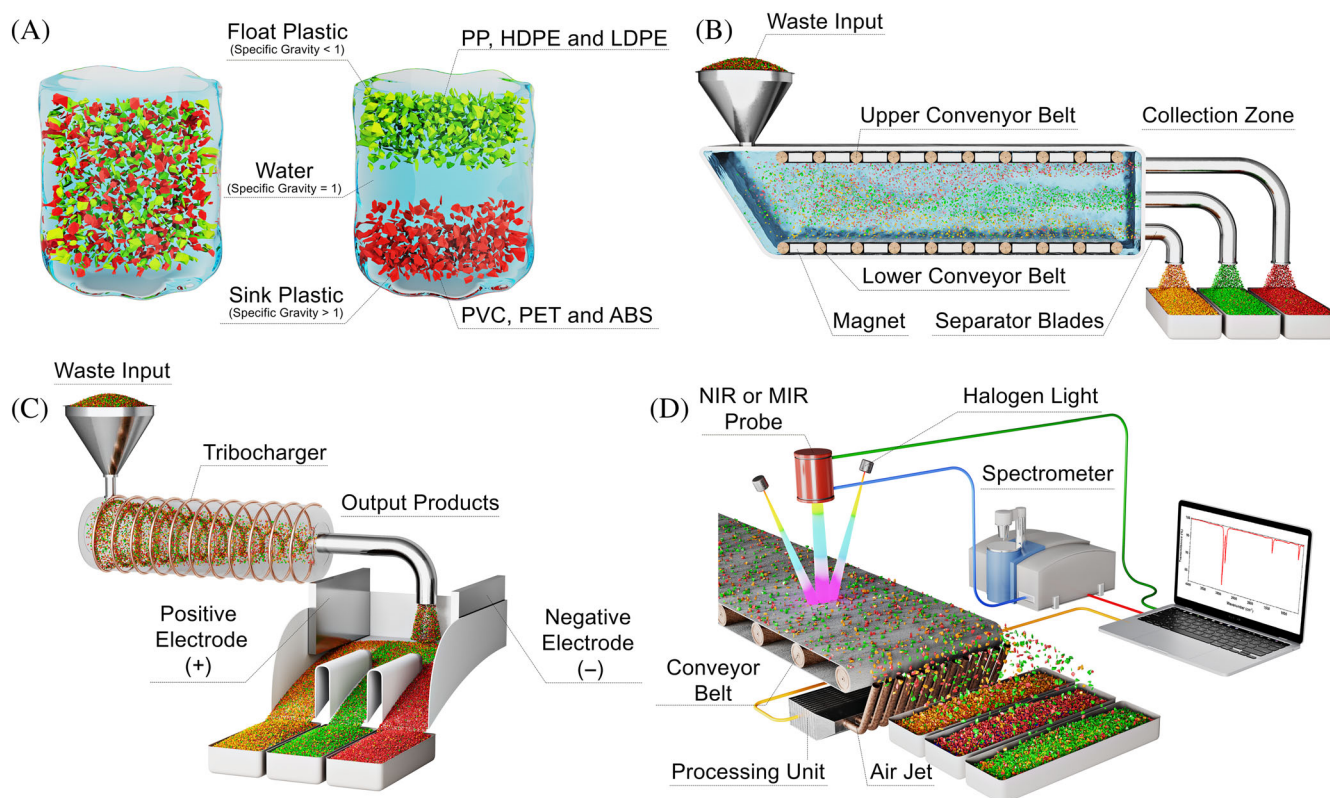


FIGURE 2 Separation of plastic waste using different sorting techniques: (A) sink-float sorting, (B) magnetic density sorting, (C) triboelectrostatic sorting, and (D) infrared spectroscopy. ABS, acrylonitrile-butadiene-styrene; HDPE, high-density polyethylene; LDPE, low-density polyethylene; NIR, near-infrared; PET, polyethylene terephthalate; PP, polypropylene; PVC, polyvinyl chloride.

500 mg of calcium lignosulphonate, 0.01 ppm methyl isobutyl carbinol (MIBC), and 0.1 mg calcium chloride at pH 11 would separate PET from PVC.^[42] Thus, in the flotation process, the wettability of the plastic materials plays a crucial role.

3.3 | Magnetic-density sorting

Magnetic density (MD) represents a significant advancement in density-based sorting that employs a magnetic fluid composed of water and magnetic particles (e.g., iron oxide particles of 10–20 nm). Using a magnetic field produces an artificial gravity where the force varies exponentially in the vertical direction. Consequently, plastic particles with different densities would float at various heights within a liquid medium, as depicted in Figure 2B. A key advantage of MD sorting over conventional density sorting is the ability to sort materials with similar densities, such as PP and PE. Additionally, wetting, feeding, separating, and collecting are the four main steps essential for successful sorting. Wetting is employed to prepare the surface of plastic waste particles for interaction with the medium, typically by making them hydrophilic.

Additionally, it is crucial to avoid turbulence in the flow stream during the sorting process, as it can adversely affect particle movement within the magnetic fluid.^[43,44]

3.4 | Triboelectrostatic sorting

The triboelectrostatic sorting technique enables the separation of various materials by exploiting disparities in their electrical characteristics. This method provides excellent efficiency and affordability while not yielding any secondary pollution. The triboelectrostatic sorting technique utilizes the tribo-charging phenomenon to impart opposite charges to distinct materials. Subsequently, these charged materials are introduced into an electric field, facilitating their separation based on their opposite charge polarities. Furthermore, how materials deflect in the electric field relies on their charge polarity and density. Numerous researchers have proposed triboelectric series, which illustrate the direction of charge transfer between the surfaces of different materials, as shown in Table 2.

As indicated in Table 2, polymethyl methacrylate (PMMA) and acrylonitrile-butadiene-styrene (ABS)

TABLE 2 Triboelectric series for different polymers.

(+) Tribo-electric series (–)	References
ABS-PP-PC-PMMA-PE-PVC-PTFE	Fujita ^[46]
PS-PET-PE-PP-PVC	Matsuhita et al. ^[47]
ABS-PC-PET-PS-PE-PP-PVC	Dodbiba et al. ^[48]
Nylon 6.6-PVAc-PVOH-PMMA-PC-PS-PE-PP-PET-PVC-PTFE	Diaz and Felix-Navarro ^[49]
PMMA-PE-PET-PP-PVC	Iuga et al. ^[50]
PMMA-ABS-PET-PE-PP-PVC-PTFE	Park et al. ^[51]
PMMA-ABS-HIPS-PET-PE-PE-PP-PVC	Park et al. ^[52]
PP-ABS-PVC-PC-PS-PE	Li et al. ^[53]

Note: Adapted with permission,^[45] Copyright 2018, Elsevier.

Abbreviations: ABS, acrylonitrile-butadiene-styrene; PC, polycarbonate bisphenol A; PET, polyethylene terephthalate; PMMA, polymethyl methacrylate; PP, polypropylene; PVAc, polyvinyl acetate; PVC, polyvinyl chloride; PVOH, polyvinyl alcohol.

typically exhibit a positive surface charge, while PVC and polytetrafluoroethylene (PTFE) tend to carry a negative charge. Jeon et al. successfully separated PVC and rubber from covering plastics in communication cable scrap.^[54] In another study, Tilmatine et al. separated 97% of the LDPE (purity of 97.83%) and 92% of the HDPE (purity of 92.45%) from the mixture of 50% LDPE and 50% HDPE.^[55]

3.5 | Spectroscopic sorting techniques

Various spectroscopic techniques such as laser-induced breakdown spectroscopy (LIBS), X-ray fluorescence spectroscopy (XRF), infrared spectroscopy (IR), and Raman spectroscopy are extensively being used for the identification of plastics. The underlying working principles of all these methods rely on the fundamental properties of materials at the atomic and molecular levels, which results in accurate materials identification.^[56] Furthermore, spectroscopic methods offer speed and precision while requiring minimal human intervention, making them ideal for plastic identification and sorting.

3.5.1 | Infrared (IR) spectroscopy

IR spectroscopy depends on the principle that molecules absorb specific wavelengths of infrared radiation due to their characteristic vibrational motions. Previously, IR spectroscopy has been employed to differentiate various polymers such as PE, PVC, PP, and PS. This technique exploits the distinctive absorbance/transmittance spectra from the vibrational processes inherent in different

polymers. The typical configuration of the near-infrared (NIR) or mid-infrared (MIR) spectroscopy-based sorting method is depicted in Figure 2D. Generally, sample fabrication for IR spectroscopy in the 2–25 μm range is challenging as samples usually need to be in thin film or solution form. This limitation hampers IR technique applicability for large-scale plastic waste separation, rendering it impractical for industrial-scale operations.^[57] Therefore, other studies focused on the two regions of the IR spectrum: NIR wavenumbers from 14,000 to 4000 cm^{-1} and MIR wavenumbers from 4000 to 400 cm^{-1} .^[58] NIR spectroscopy rapidly sorts different plastics, as each polymer reflects distinct characteristic spectra. However, NIR spectroscopy cannot separate black or dark-coloured polymers since black polymer absorbs all inbound NIR radiations, resulting in reduced reflections and, hence, worthless spectra for separation.^[59] This issue can be mitigated with MIR spectroscopy, which enables the identification of plastics by inducing localized melting through MIR radiation.^[60]

3.5.2 | Raman spectroscopy

Raman spectra provide characteristic peaks corresponding to the functional groups in the molecules, enabling the identification of the materials even in complex mixtures.^[56] The operational procedure for Raman spectroscopy sorting is similar to that of NIR spectroscopy. However, it has several advantages over IR and NIR techniques, including the capability to sort black plastic samples, less negative impact on the sorting efficiency in the presence of H_2O and CO_2 , and enhanced spatial resolution.^[61,62] However, Raman spectroscopy has drawbacks, including a low signal-to-noise ratio, random background emissions, and potential polymer degradation during separation. Additionally, it is prone to interference from fluorescence.^[63]

3.5.3 | Laser-induced breakdown spectroscopy (LIBS)

LIBS is capable of distinguishing among all six polymer types, including LDPE, HDPE, PP, PET, PS, and PVC. LIBS instrumentation system comprises three essential components: a pulsed laser source, a charge-coupled device spectrometer, and a data processing unit, as illustrated in Figure 3A.^[64] During the sorting process, a focused, high-energy beam heats a small section of the plastic waste intensely. This intense heating leads to the formation of plasma plumes and the emission of characteristic radiation. Subsequently, a spectrometer

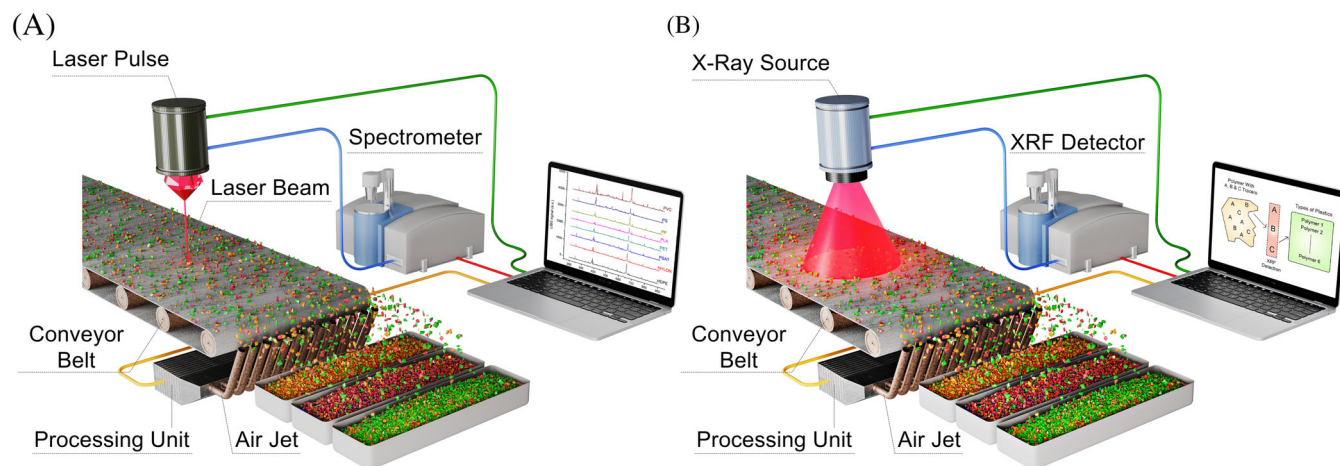


FIGURE 3 Separation of different plastic waste based on different spectroscopies techniques: (A) laser-induced breakdown spectroscopy (LIBS) and (B) X-ray fluorescence (XRF) spectroscopy.

captures these emitted radiations, which are then processed by a data processing unit.^[61] LIBS is well-suited for industrial applications due to its speed, efficiency, and lack of need for sample preparation. Several factors can impede the sorting efficiency of the LIBS technique, including surface contamination, signal fluctuations due to varying plasma generation, and substantial initial investment costs.^[65]

3.5.4 | X-ray fluorescence (XRF) spectroscopy

X-ray fluorescence (XRF) spectroscopy is a non-destructive sorting method that can sort a wide range of materials, including metals, cement, oils, and polymers.^[43] Plastic sorting is accomplished by analyzing fluorescence emissions acquired from the samples (Figure 3B). In XRF spectroscopy, an external X-ray source excites the atoms on the surface of polymers, resulting in the emission of specific X-ray photons. These X-ray photons generate distinctive spectra regarding atomic weight and element type.^[66] XRF can quickly identify and sort dark and even black plastics. In a recent study, Olscher et al. utilized XRF to recover PVC and plastics containing brominated flame retardants, primarily due to the distinctive presence of chlorine or bromine peaks.^[67] Brunner et al. successfully separated PVC from PET using the XRF technique.^[68] XRF offers several advantages over other methods, including the ability to identify additives from trace to ultra-trace levels and differentiating plastics within a mixed mixture based on fluorescence spectra. Moreover, it can analyze the time dependence of fluorescence decay.^[56] However, safety considerations are associated with using X-ray sources.^[69]

3.6 | Cleaning and decontamination of plastic waste

3.6.1 | De-inking/decolouration process

To maintain the quality of recycled plastic materials, it is crucial to eliminate ink from printed plastics before the extrusion process. Understanding the composition of printing inks is vital for effective de-inking. These inks consist of resins, solvents, colourants, and additives, each serving a distinct role in the formulation.^[70] Resins, comprising 15%–50% of the ink, stabilize colourants and ensure substrate adhesion.^[71] Solvents, making up to 65%, dissolve resins for proper ink transfer.^[72] Colourants, constituting 5%–30%, provide desired hues, while additives are added to enhance ink properties. Solvent-based inks are preferred for different applications, but ink composition varies based on the printing method and substrate.

UV-based inks, cured using UV radiation, present unique challenges due to their reactive resins.^[73] The UV-based ink diversity expands plastic packaging functionality but complicates recycling, especially de-inking processes. Generally, solvent- or water-based solutions are employed to remove the ink effectively. Ferreira et al. successfully extracted up to 94% of blue pigments and 100% of orange pigments by employing limonene as a solvent and a wide range of alcohols as antisolvents.^[74] Gecol et al. discovered that cationic surfactants exhibit excellent capability to remove both water-based and solvent-based inks at alkaline pH.^[75,76] They investigated the removal of water-based inks from polyethylene film using various surfactants. They concluded that cationic surfactants were highly effective for de-inking across pH levels of 5–12, while anionic surfactants showed minimal

effectiveness, even at high pH levels. Another study found that non-ionic surfactants can de-ink, but their effectiveness depends on the solution's pH.^[77] Subsequently, Gecol et al. also explored the impact of calcium ion concentration on de-inking efficiency. They found that both synthetic and natural anionic surfactants were effective in removing water-based ink from plastic film at alkaline pH levels, but only in the presence of calcium ions.^[78] In another study, Chotipong et al. investigated the extraction of inks using alkyl trimethylammonium bromides, specifically dodecyl-, tetradecyl-, and hexadecyltrimethylammonium bromide, referred to as DTAB, TTAB, and CTAB, respectively. The authors examined the impacts of pH, temperature, concentration, and the alkyl chain length.^[79] The results demonstrated that cationic surfactant CTAB effectively removed both water- and solvent-based inks. Moreover, critical micelle concentration (CMC), medium pH, temperature, and stirring were categorized as crucial factors affecting de-inking efficiency.^[79]

The literature also showcases several patents focused on de-inking plastic waste during recycling. Fullana and Lozano developed an innovative method for removing ink from plastic surfaces.^[80] Its technical and economic feasibility was validated by establishing a semi-industrial de-inking plant with a capacity of 100 kg/h.^[81] The resulting ink-free pellets matched the original quality and were suitable for high-value product manufacturing. The patent offers an economically viable method for removing solvent, water, and UV-based inks from monolayer or multilayer plastic materials.^[82] The de-inking medium comprises amine and liquid components, such as water, alcohol, hydrocarbons, and organic solvents.

3.6.2 | Decontamination process

The decontamination process is a highly critical step in recycling plastic waste. Before undergoing the recycling process, sorted plastics must undergo a thorough cleaning. Impurities can result in an inferior recycled product and negatively impact its market value. Contaminants are broadly classified into Intentionally Added Substances (IAS) and Non-Intentionally Added Substances (NIAS). As the name implies, IAS are incorporated into plastics to enhance their physicochemical properties. These substances encompass a range of additives, such as flame-retardants, plasticizers, and stabilizers, among others.^[83] NIAS contaminants may include external foreign particles such as dirt, grease, food waste, or by-products resulting from their breakdown.

Due to their inherent limitations, recycled plastics from post-consumer sources are commonly restricted to applications in construction, agriculture, or piping.^[84]

Meeting industrial standards for high-demand applications like packaging is challenging due to complications arising from colour, odour, and the migration of unidentified organic substances.^[85] Various decontamination methods, such as extraction using polyethylene glycol, hot water extraction with sodium hydroxide, subcritical water extraction, ultraviolet radiation oxidation, and advanced oxidation using ultraviolet radiation with H_2O_2 , were utilized to reduce contaminants.^[86]

Once collected, the plastic waste undergoes a pre-washing tank to eliminate surface and heavy impurities. Moreover, hot water or compressed air efficiently removes greasy trash and stickers. The odorous components of plastic waste are partially eliminated through caustic washing, while organic solvents and detergents are required to remove the most apolar components.^[5,87,88] Detergents are also employed during the mechanical cleaning process to achieve complete decontamination. Alassali et al. successfully extracted organic contaminants from plastic waste using the CO_2 supercritical extraction method.^[89] In a parallel investigation, supercritical carbon dioxide extraction was utilized to extract aliphatic hydrocarbons such as eicosane, tetracosane, and nonadecane from the surface of LDPE. The extraction process was conducted at 22 MPa and 60°C for 4 h, demonstrating enhanced extraction efficiency compared to Soxhlet extraction.^[90]

Efficient recycling was achieved by reclaiming surface-coated metals from discarded ABS plastic waste using ammoniacal leaching and solvent extraction techniques.^[91] Welle employed a water-based solution comprising 2%–3% sodium hydroxide (NaOH) and specific detergents to diminish surface impurities and eliminate labels and adhesive residues from PET bottles.^[92] In another study, agrochemical plastic packaging waste was decontaminated and recycled using a triple-rinsing procedure.^[93] Cabanes and Fullana employed polyethylene glycol at 100°C to eliminate volatile organic compounds (VOCs) from post-consumer recycled HDPE, achieving a 70% reduction in the analyzed VOCs.^[94] The solvent extraction method was utilized to extract polycyclic aromatic hydrocarbons (PAHs), heavy metals, and biphenyl polychlorides (PCBs) from recycled PE samples sourced from agricultural, post-commercial, post-industrial, and post-consumer origins.^[84]

3.7 | Strategies and methods for plastic waste recycling

3.7.1 | Mechanical recycling

Before beginning the reprocessing of recycled materials, it is essential first to convert the waste into new raw

materials. This process starts after waste collection and involves several steps, including sorting, cleaning, grinding, baling, and reprocessing. This process is called mechanical recycling, which only reprocesses the discarded plastic without altering the chemical structure of polymers. Mechanical recycling is commonly employed to clean single-type plastic waste like PP, PE, PET, and PS. However, its effectiveness is limited when dealing with complex, heterogeneous, and contaminated waste. These recycled polymers are typically repurposed for a different application than their initial use.^[95]

Collection and sorting

Plastic waste is collected and sorted according to shape, size, density, colour, and chemical composition, as discussed above in Section 3.

Cleaning

Removing colour, odour, greasy contaminants, and organic contaminants.

Grinding and shredding

Reducing the size of plastic waste into flakes.

Baling

When plastic is not processed at the sorting location, it is typically baled for transportation.

Reprocessing

Extrusion stands as the primary method for reprocessing shredded plastic waste. The plastic waste is fed into an extruder and heated until it reaches the designated temperature, which is typically around the melt temperature of plastics. Subsequently, the material is transformed into either reprocessed filaments or pellets, as shown in Figure 4, facilitating their reuse in various applications. Applying heat and shear forces to polymers within an extruder results in thermo-oxidative and shear-induced alterations such as thermal degradation, chain scission, chain branching, or material crosslinking.^[18,96,97] This degradation of the polymer chains reduces their length, consequently reducing the material's mechanical properties and processability.^[98] The degradation mechanism involves the production of radicals along the polymer chain, originating from oxygen-induced peroxy radicals and thermally induced hydrogen atom abstraction. These radicals, influenced by the applied shear forces, can induce β -scissions in chains, thereby reducing chain length and decreasing viscosity.^[96] Due to this degradation, the mechanical properties of the recycled polymer, including tensile strength, elongation at break, and impact resistance, were significantly reduced.^[96]

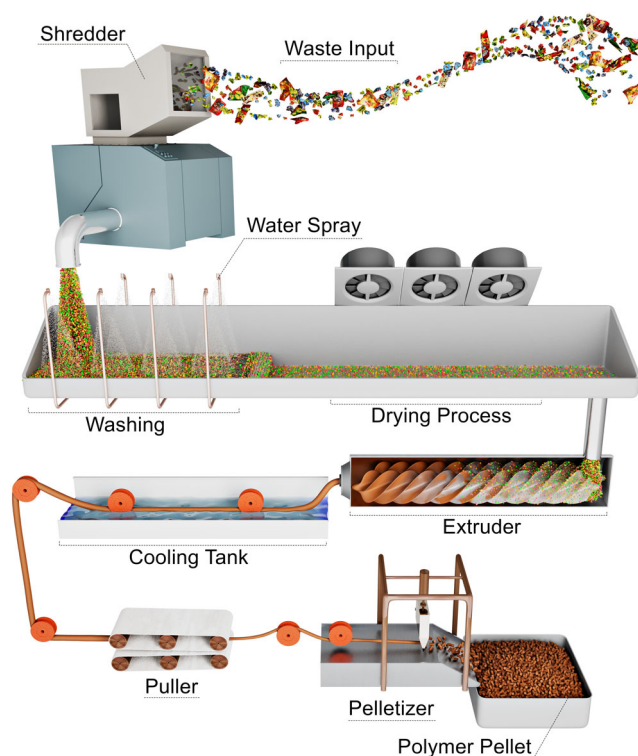


FIGURE 4 Schematic representation of processing plastic waste into pellets using a melt extruder following washing and thorough drying.

The limitations of recycled plastic in terms of physical and mechanical properties hinder its potential application. Additives, including fillers, stabilizers, antioxidants, plasticizers, pigments, and flame retardants, are introduced during extrusion to enhance and customize the properties of the recycled product.^[5] Recycled plastics are typically used to produce lower-quality products and face economic competition with those made from virgin plastics.^[38]

3.8 | Chemical recycling

Chemical recycling of plastic waste is a process that involves breaking down polymers into their constituent components, including valuable chemicals and gases, which can be repurposed as fuel or used for re-polymerization.^[99] As a result, chemical recycling offers a practical approach to recycle plastic waste. Researchers have explored various methods for breaking down polymers into valuable components. Chemical recycling encompasses processes like pyrolysis, solvolysis, and gasification. These processes are categorized based on the solvents or methods used: solvolysis^[100] (in the presence of solvents), pyrolysis^[16] (in the absence of air), and

gasification^[101–103] (in a controlled environment), which will be explained in the following sections.

3.8.1 | Pyrolysis

Pyrolysis is a thermo-chemical process in which a polymer is heated in the absence of oxygen. Consequently, the higher molecular weight polymer chains are broken down into smaller molecules, resulting in products in three different phases: a liquid phase (e.g., oil and tar, 10–45 wt.%), a solid phase (char, 5–25 wt.%), and a gas phase.^[104] The two main differences between pyrolysis and combustion are that pyrolysis occurs at lower processing temperatures and does not require oxygen, thus producing fewer air pollutants. Pyrolysis is a sustainable method that addresses both the reduction of plastic waste and the growing energy needs by converting waste into valuable products. The product yields depend on several factors, such as reaction temperature, heating rate, residence time, and reactor type.^[105] The flexibility of pyrolysis techniques is a crucial advantage, allowing for generating a combination of products in different states (solid, liquid, and gas) by simply adjusting temperature and heating rate parameters.^[106] The pyrolysis process for recycling plastic waste offers several benefits, including (i) operational, (ii) environmental, and (iii) economical. Typically, pyrolysis generates residual by-products like char, which can be used as a fuel or raw material for additional petrochemical applications. It is an eco-friendly and sustainable substitute for land-fill disposal, reducing greenhouse gas emissions.^[107] From an economic perspective, pyrolysis stands out as a preferred approach.

Moreover, pyrolysis is extensively applied to recycle PE, PS, and PP. Ahmad et al. achieved a 98.66% total conversion of PP at 300°C, yielding 69.82% liquid, 28.84% gases, and 1.34% residue. They observed a strong correlation between temperature variation and product yield.^[108] Eletta et al. conducted thermal pyrolysis of LDPE across a temperature range of 203–400°C, resulting in a product comprising 78.4% liquid, 17.2% solid residue, and 4.4% organic gases. The obtained hydrocarbons were primarily aliphatic and ranged from C10 to C27.^[109] Pyrolysis of PET and PVC encounters challenges due to the formation of undesirable chlorinated by-products, corrosion issues, downstream catalyst fouling, and potential blockages within pyrolysis plants.^[110] An effective strategy to mitigate these challenges involves employing dechlorinating agents. Snow et al. employed the 1:1 ratio of acidic β -zeolite with alkali hydrotalcite to absorb the produced chlorinated hydrocarbons. The synergistic effect of β -zeolite with alkali hydrotalcite resulted in a

dechlorination efficiency of 38%, while the individual efficiency was 12% and 6%, respectively.^[111]

The type of reactor is another essential factor in pyrolysis. Batch reactors, for instance, are among the reactors with high conversion value, owing to their closed system without any input or output. Semi-batch reactors, on the other hand, are capable of adding reactants or removing products while conversion is taking place. However, the products are not uniform from batch to batch, and char removal is challenging due to the long solid residence time. Thus, the appropriate choice of input and the optimal process conditions are the prerequisites for the successful application of pyrolysis.^[112] Nonetheless, the pyrolysis process entails specific shortages such as high reaction temperatures, prolonged reaction durations, and difficulties in accurately forecasting the distribution of the final products.^[107]

Catalytic pyrolysis

Compared to conventional pyrolysis (thermal pyrolysis), catalysts are used in catalytic pyrolysis, resulting in lower temperatures and shorter reaction times. The typical catalytic pyrolysis process is shown in Figure 5. Using catalysts can lead to particular reactions and inhibit side reactions, increasing the conversion yield.^[113] In catalytic pyrolysis, two types of catalysts are utilized and classified based on their phase. Homogeneous catalysts share the

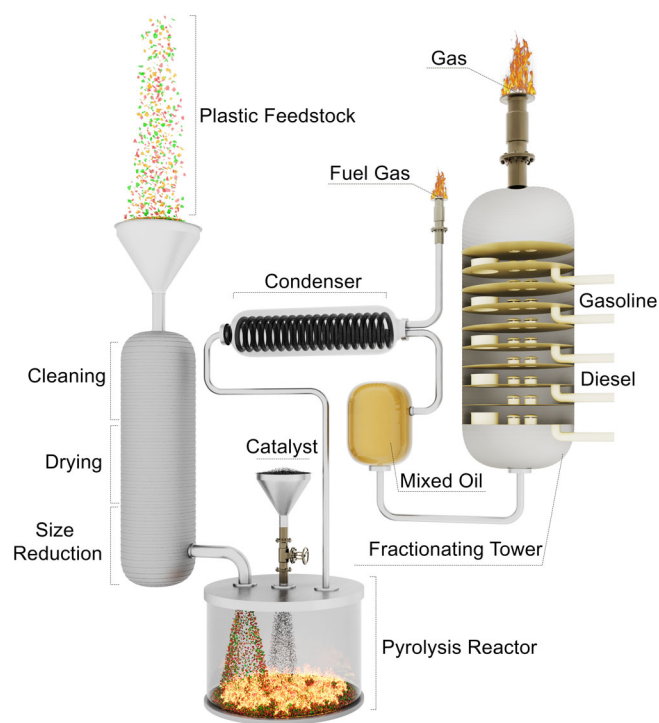


FIGURE 5 Schematic diagram illustrating the catalytic pyrolysis setup utilized for the conversion of plastic waste into value-added chemicals.

same phase as the reaction medium, while heterogeneous catalysts are in a different phase. Lewis acids, like AlCl_3 , and metal tetrachloroaluminates melts are commonly used as homogenous catalysts.

In contrast, heterogeneous catalysts comprise nanostructured zeolites, aluminium-pillared clay, mesostructured catalysts, and superacid solids.^[114] Moreover, heterogeneous catalysts are preferred for their ease of separation and recovery from the reaction medium.^[115] Utilizing alumina-supported or rare earth metal-based catalysts in recycling polyolefins increases reaction rates and yields more significant amounts of desirable isoalkenes and aromatics for biofuel production.^[116] These catalysts offer an efficient approach to breaking C—C bonds, allowing the metathesis of long polymeric chains into shorter alkene chains.^[117] Despite their cost, these catalysts are of great interest due to their reusability and high output.

Additionally, porous inorganic materials such as ZSM-5, zeolites, Y-zeolites, and MCM-41 are predominantly employed in polymer decomposition.^[118] These catalysts offer an extensive surface area, facilitating the effective breakdown of polyolefins into shorter-chain compounds. For instance, Pyo et al. investigated the catalytic pyrolysis of PP using a Ga-loaded HZSM-5 catalyst.^[119] Fuentes et al. employed a Ruthenium-based catalyst for PET recycling, demonstrating superior performance compared to conventional catalysts.^[120] Zhang et al. employed $\text{Pt}/\text{Al}_2\text{O}_3$ as a catalyst for recycling PE via hydrogenolysis, which performs better than metal oxides.^[121]

Recently, researchers have investigated biocatalysts for polymer recycling.^[122] Furthermore, 90 microorganisms, including bacteria and fungi, have been identified as capable of degrading petroleum-based plastics.^[123] The process of biodegradation can depolymerize the polymer into valuable chemicals and even lead to the degradation of plastics into CO_2 and H_2O . However, the biodegradation of non-hydrolyzable plastics is still challenging because of their non-hydrolyzable backbone structure.

3.8.2 | Solvolysis

Solvolysis represents a solvent-dependent technique wherein individual plastics undergo a chemical reaction or depolymerization process, ultimately yielding monomers. These depolymerization methods encompass alcoholysis, hydrolysis, acidolysis, and aminolysis. The above processes are named after the solvents or medium used, such as methanolysis^[100] (in methanol), glycolysis^[124] (in ethylene glycol), aminolysis^[125] (in ethanolamine), acidolysis (in acid), and hydrolysis^[126] (in water). Solvolysis is

generally well-suited for step-growth thermoplastics and thermosets like polyurethanes, polyesters, and polyamides.^[127] A well-known example of solvolysis is the solvolytic reactions of PET, such as hydrolysis, methanolysis, and glycolysis. PET hydrolysis can occur under high temperature and pressure conditions in neutral, alkaline, or acidic environments, producing terephthalic acid (TPA) and ethylene glycol (EG).

Furthermore, the choice of a catalyst affects the yield, reaction temperature, and reaction time. Alkaline catalysts, such as sodium bicarbonate, sodium hydroxide, and potassium hydroxide, increase yield while reducing char production. Conversely, acidic catalysts like phosphoric or sulphuric acid lower the liquefaction temperature and reduce residence time.^[128] The drawbacks of hydrolysis include the low purity of TPA and the relatively slow process due to the weak nucleophilic properties of water.^[16]

Another method is methanolysis, where PET is subjected to methanol along with a catalyst at elevated temperatures ranging from 180 to 280°C and pressures between 20 and 40 atm. This process predominantly yields dimethyl terephthalate (DMT) and EG.^[129] The significant disadvantages of methanolysis are its production cost and high corrosiveness.

Glycolysis is a widely employed method in industries that depolymerizes PET waste. It is mainly used for high-quality, uncontaminated PET scrap, but its effectiveness diminishes with lower-quality copolymers. In glycolysis, PET is depolymerized using EG or propylene glycol (PG) at a temperature range of 180–240°C and ambient pressure in the presence of a catalyst. Glycolysis yields a mixture of bis-2-hydroxypropyl terephthalate (BHET), dimer, and oligomers.^[130] The high reaction temperature, prolonged reaction time, and low purity of BHET hinder the applicability of glycolysis.^[131] The detailed description is given in Section 4.

3.8.3 | Gasification

Gasification is a thermochemical method for transforming plastic waste into synthetic gas, also known as syn gas. Syn gas comprises various gases like H_2 , CO , CO_2 , CH_4 , and small quantities of acetylene (C_2H_2) and ethylene (C_2H_4), as shown in Figure 6.^[132] Among thermochemical methods, gasification is highly preferred because it generates H_2 gas, which is ideal for combustion. Moreover, product yield depends on the physical condition of plastics and the operating condition of gasification, such as temperature, feed concentration, time, and catalyst loading.^[133] Unlike the pyrolysis process, which occurs in the absence of oxygen, the gasification process requires an oxidizing agent, typically a combination of steam and pure

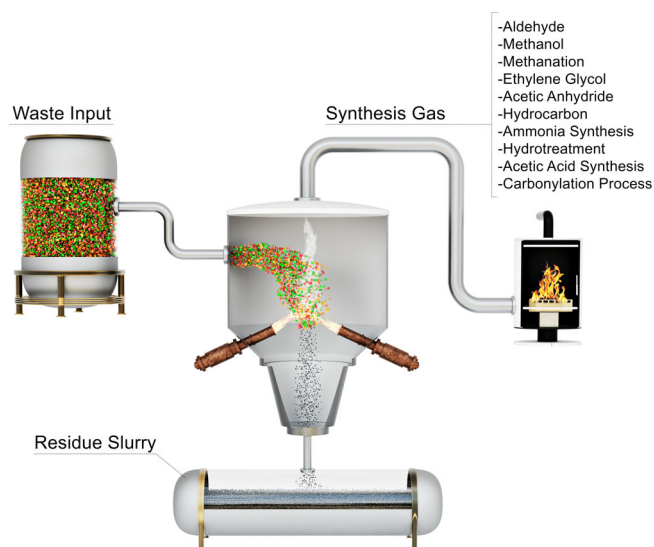


FIGURE 6 Representation of the gasification process using different gases and the resulting products.

oxygen or just air.^[134] Air is commonly used as a gasification agent in standard gasification processes to reduce costs and simplify operations. However, fuel gases were diluted with the inert N_2 in the air, reducing their calorific value. Thus, steam was introduced in a stoichiometric ratio to reduce N_2 content.^[107]

When air is used as the oxidizing agent, the temperature typically ranges from 800 to 1000°C, but it may reach up to 1500°C in the presence of oxygen. Bai et al. studied the supercritical water gasification of high-impact PS (HIPS) plastic at reaction temperatures ranging from 500 to 800°C and pressures between 22 and 25 MPa. Under the optimal gasification reaction conditions, they achieved a plastic carbon conversion rate of 94.48 wt.%.^[135] Zhao et al. studied the degradation of PS in supercritical water/ CO_2 .^[136] They found that at 400°C, the liquid product primarily comprised ethylbenzene, styrene, alpha-methylstyrene, and 2-phenylnaphthalene. As the reaction temperature reached 600°C, it predominantly contained naphthalene, biphenyl, fluorene, and anthracene. Saebea et al. transformed plastic waste (PE and PP) into syn gas through steam gasification.^[137] A significant amount of H_2 was produced from the LDPE and pine sawdust blend using a Ni-CaO-C catalyst.^[138]

A notable advantage of gasification is its ability to attain optimal conversion efficiency without fine sorting, accommodating a diverse range of materials. However, the expenses associated with the sophisticated equipment and advanced technology pose significant economic challenges, impeding widespread adoption and implementation. Therefore, continued research and development are necessary to enhance the cost-effectiveness and scalability of gasification technology.

3.9 | Energy recovery

As the name suggests, energy is generated through heat, steam, and electricity by burning plastic waste, as shown in Figure 7. Advancements in incinerator technologies have led to increased global use due to their high efficiency. Energy recovery is the last practical method of waste treatment when plastic recycling is restricted due to certain economic constraints and the degradation of chemical and mechanical properties. Moreover, it reduces the volume of waste, and the residue can be easily dumped in the landfill. This technique will be helpful in countries with limited land for landfill applications.^[139] As plastics are derived from crude oil, they possess a high calorific value when incinerated, as indicated in Table 3. Hence, they are a suitable alternative to energy sources.

The combustion of plastic waste causes various environmental hazards, such as the emission of carbon dioxide, NO_x and SO_x gases, VOCs, smoke, particulate-bound heavy metals, PAHs, polychlorinated dibenzofurans (PCDFs), and dioxins.^[107,142] Therefore, technological advancements in incineration techniques are necessary to address the issue of polluting gases and comply with strict regional legislation.^[143]

4 | RECYCLING OF PET WASTE

PET is a widely used thermoplastic polymer, ranking as the fourth most produced polymer globally and accounting for approximately 18% of all polymers manufactured.^[139] Its exceptional mechanical, thermal, and optical properties make it ideal for applications in beverage bottles, magnetic tapes, X-ray films, and textiles. PET is industrially obtained via either a polycondensation reaction involving EG and TPA or a transesterification reaction between DMT and EG.^[144]

PET can be recycled through various recycling methods, including mechanical, chemical, and incineration. PET is recycled via the mechanical recycling process involving sorting, washing, shredding, and reprocessing into new products.^[145] Mechanical recycling of PET offers advantages over chemical recycling, such as lower investment costs, process simplicity, feedstock flexibility, and minimal environmental impact.^[146] However, diversity within PET waste and the presence of contaminants lead to the production of inferior grey or yellowish products, restricting their suitability for desired traditional applications.^[147,148] Additionally, mechanical recycling decreases the mechanical properties of non-virgin materials with each reuse cycle. La Mantia and Vinci investigated PET recycling through multiple extrusions under both dry and humid conditions. They observed a rapid

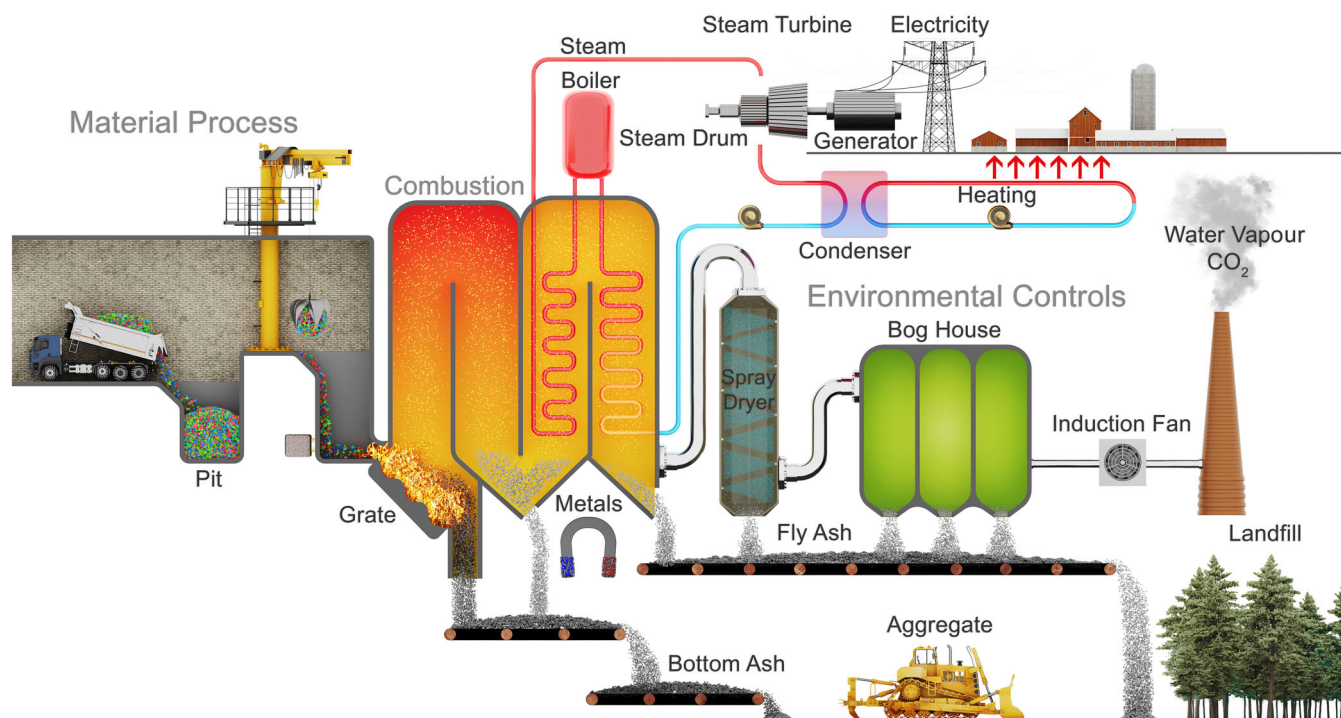


FIGURE 7 Schematic diagram depicting the incineration process.

TABLE 3 Calorific values for different polymers.

Item	Calorific value (MJ kg ⁻¹)	References
HDPE	47.39	Miranda et al. ^[140]
Jet fuel, JP-4	46.60	Kittle ^[141]
PP	46.50	Al-Salem et al. ^[107]
Gas oil	45.20	Al-Salem et al. ^[107]
LDPE	43.34	Miranda et al. ^[140]
Heavy oil	42.50	Al-Salem et al. ^[107]
Petroleum	42.30	Al-Salem et al. ^[107]
PS	41.90	Al-Salem et al. ^[107]
Coal, anthracite	32.80	Kittle ^[141]
Household plastic waste	31.80	Al-Salem et al. ^[107]
Polycarbonate bisphenol A (PC)	31.53	Kittle ^[141]
PET	24.13	Kittle ^[141]
PVC	21.00	Miranda et al. ^[140]
Wood, dry, average	20.00	Kittle ^[141]
Paper, Newsprint	19.70	Kittle ^[141]

Abbreviations: HDPE, high-density polyethylene; LDPE, low-density polyethylene; PET, polyethylene terephthalate; PP, polypropylene; PS, polystyrene; PVC, polyvinyl chloride.

fourfold reduction in the elongation at break value after subjecting the material to mechanical recycling for five cycles.^[149] The significant decline in material

properties can be attributed to the degradation of polymer chains through thermo-oxidative, hydrolytic, and thermo-mechanical degradations.^[18,149] López et al. also observed a decrease in the strain-at-break value from 42% to 0.7% after the fifth cycle of mechanical extrusion of PET.^[150]

Given the limitations of mechanical recycling mentioned earlier, there is significant exploration into chemical recycling methods to convert PET into higher-value products.^[150–153] Pyrolysis, for example, can be used to generate precursor monomers of PET and specific side products. Dhahak et al. conducted PET recycling using slow pyrolysis in a horizontal tubular reactor across various temperatures from 410 to 480°C. They obtained an 11 wt.% yield of benzoic acid at 430°C.^[154] Kenny et al. transformed PET into biodegradable plastic polyhydroxyalkanoate (PHA) through pyrolysis at 450°C. This process resulted in TPA and various oligomers. The TPA produced served as the primary material for bacterial synthesis of PHA.^[155]

In another study, PET was pyrolyzed in a bubble fluidized reactor at 420–450°C. This process resulted in gaseous products (16–18 wt.%) and benzoic acid, monovinyl terephthalate, divinyl terephthalate, vinyl benzoate, and benzene.^[156] PET was repurposed into synthetic graphite via thermal pyrolysis at 900°C in a tube furnace under inert conditions. Later, microwave-assisted liquid-phase exfoliation was utilized to produce graphene sheets from the obtained graphite.^[157] Significant advancements have been achieved in lab-scale pyrolysis, but its application at

an industrial scale for depolymerizing PET into monomers and valuable products is still limited. This limitation is primarily due to the production of diverse liquid and gaseous by-products during pyrolysis, which diminishes process efficiency and necessitates expensive separation techniques.^[158]

Catalysts have addressed thermal pyrolysis drawbacks like product selectivity, economic viability, high temperature, extended reaction time, and low yield. Claudino and Ariza obtained 80% benzene as the primary product in the oil phase and ethanol, acetone, and acetaldehyde in the aqueous phase by employing $\text{Ca}(\text{OH})_2$ as a catalyst for pyrolyzing PET waste at 430–440°C, with a residence time of 30 min.^[159] In another study, PET was successfully depolymerized under 1 atm of H_2 into TPA and ethylene using a supported single-site C/MoO_2 catalyst.^[160] Diaz-Silvarrey et al. recovered benzoic acid via catalytic pyrolysis of PET using sulphated zirconia.^[161] The impact of various metal oxides like $\text{Ca}(\text{OH})_2$, NiO , Fe_2O_3 , or TiO_2 on PET pyrolysis was investigated by Yoshioka et al. At 700°C, they derived benzene through the decarboxylation of TPA, catalyzed by $\text{Ca}(\text{OH})_2$.^[162] Ben and co-workers conducted PET pyrolysis using a ZSM-5 zeolite and NiCl_2 catalyst from 450 to 600°C under N_2 atmosphere. Their

results demonstrated that the introduction of a zeolitic catalyst significantly decreased the wax yield from 67.70 to 23 wt.%, while the gas yield increased from 20 wt.% to over 50 wt.%. Additionally, the presence of ZSM-5 led to relatively lower oxygen content in a waxy product, suggesting its potential to facilitate deoxygenation during PET pyrolysis.^[163] In another investigation, PET extracted from carpet waste was transformed into benzene-rich oils through thermal, catalytic, and catalytic steam pyrolysis methods.^[164] ZSM-5 and CaO served as the catalysts during PET's catalytic degradation. They noted that ZSM-5 facilitated polymerization reactions, resulting in polyaromatic hydrocarbons, while CaO generated undesirable ketones. Additionally, this study achieved high percentages of benzene with high purity.

Solvolysis represents a viable alternative for the chemical recycling of PET, involving PET reactions with various reagents such as glycols, alcohols, amines, and water to produce monomers or value-added chemicals. Existing commercial methods for the chemical recycling of PET include glycolysis, hydrolysis, methanolysis, and aminolysis, as depicted in Figure 8.^[16,153,165,166] Typically, depolymerization reactions yield products such as TPA, DMT, BHET, EG, and oligomers.

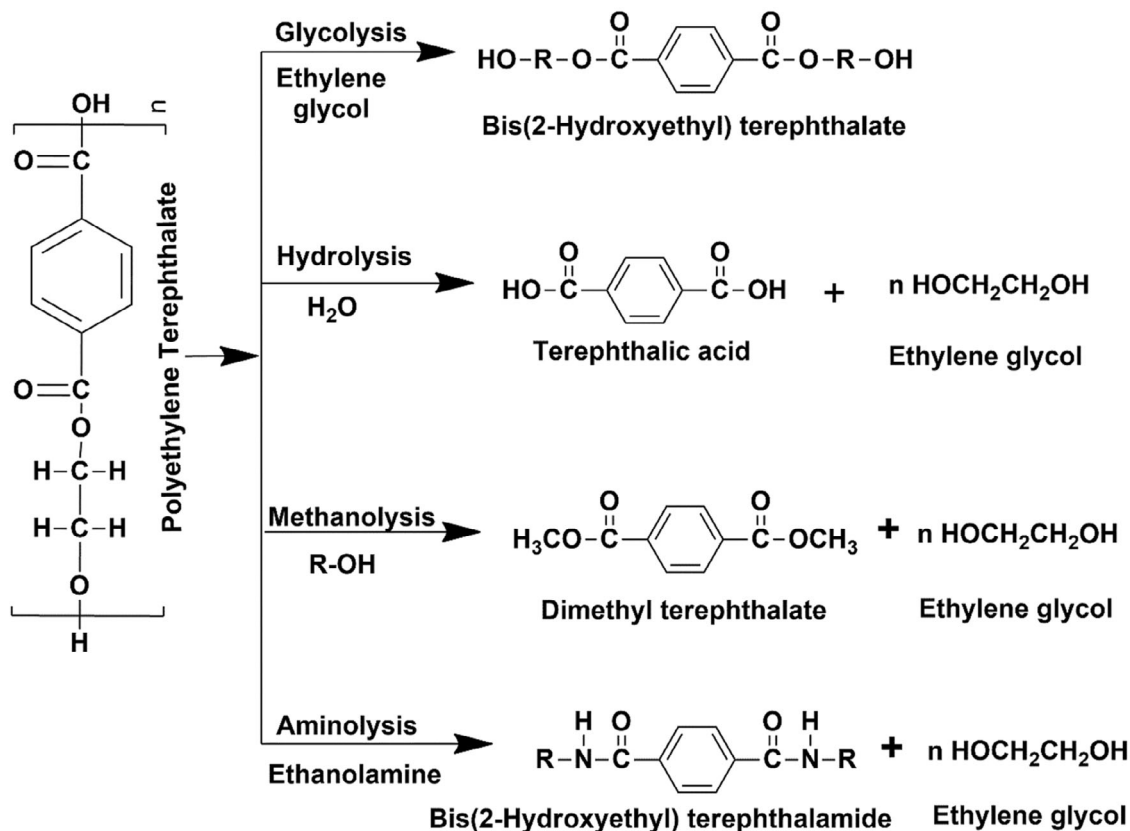


FIGURE 8 Chemical depolymerization of polyethylene terephthalate (PET) into different chemicals.

Glycolysis is a standard and most straightforward method for depolymerizing PET. The chemical recycling of PET through glycolysis was initially discovered by McDowell et al. in 1965.^[167] Several prominent companies, like DuPont, Goodyear, Shell Polyester, Zimmer, Eastman Kodak, and so forth, have utilized this process for PET depolymerization.^[16,168] PET undergoes depolymerization through glycols in this method, forming adaptable monomers, oligomers, or polyols for various applications. Glycolysis is generally subjected to high-quality PET feed.^[168] During glycolysis, PET undergoes transesterification with an excess of glycol, typically at temperatures ranging from 180 to 250°C, facilitating the formation of BHET.^[147] Usually, glycolysis involves the use of EG,^[169] diethylene glycol (DEG),^[170] PG,^[171] butylene glycol (BG), and dipropyleneglycol (DPG).^[172,173] Chen et al. observed a slow glycolysis rate without a catalyst and incomplete depolymerization of PET to BHET.^[169] Moreover, they found that the reaction parameters such as temperature, catalyst concentration, glycolysis time, and glycol concentration significantly influence the PET glycolysis process. The sequence of significance for these reaction conditions can be arranged as catalyst concentration > glycolysis temperature > glycolysis time.^[174–176] Pingale et al. reported an increased glycolysis reaction rate with the use of metal-based catalysts.^[177] Baliga and Wong studied the catalytic efficiency of four metal acetates (Pb, Zn, Co, and Mn) on the glycolysis of PET waste bottles. They found that Zn-acetate outshined all other catalysts in terms of depolymerization products.^[178] Farahat and Nikles investigated the effect of changing the glycol concentration while keeping other reaction parameters (time, temperature, and catalyst concentration) constant. They noted significant differences in the functionality and molecular weights of the obtained glycolysis products.^[179,180]

Another practical approach for breaking down PET is hydrolysis, a method in which PET interacts with water in an acidic, alkaline, or neutral environment, giving rise to its complete depolymerization into monomers (TPA and EG). There is a growing interest in this method owing to advancements in the direct synthesis of PET from EG and TPA, which removes the necessity for methanol in the synthesis process.^[166] However, inevitable drawbacks associated with this method include a long reaction time, elevated temperature (200–250°C), and high pressure (1.4–2 MPa). The depolymerization of PET can be achieved through (a) acid hydrolysis and (b) alkaline hydrolysis (as shown in Figure 9).^[146]

Acid hydrolysis is commonly conducted using sulphuric acid, although other concentrated mineral acids like nitric acid can also be used.^[146,181] Achilias and Karayannidis demonstrated acid hydrolysis of PET at various temperatures and solution concentrations, ranging from

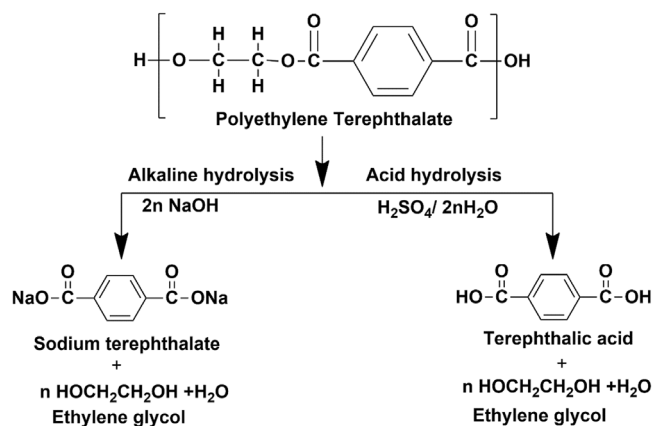


FIGURE 9 The hydrolysis reaction of polyethylene terephthalate (PET) waste under different pH conditions.

30 to 90°C.^[182] Mehrabzadeh et al. also investigated the influence of parameters like PET particle size, reaction temperature, time, and acid concentration on the decomposition and reaction yield.^[183] Yoshioka et al. developed a process for depolymerization of PET waste bottles using 7–13 M nitric acid at 70–100°C for 72 h. The EG obtained was simultaneously oxidized into oxalic acid, which possesses greater commercial value than both TPA and EG.^[181] Despite its high yields, a significant challenge of acid hydrolysis is the separation of EG from the acidic solution. Moreover, the use of highly corrosive acids at an industrial scale poses economic, process, and environmental challenges.

The standard method for the alkaline hydrolysis of PET typically involves using an aqueous solution of NaOH or KOH at a concentration ranging from 4 to 20 wt.%.^[166,168] Mainly, the reaction produces EG and disodium or dipotassium terephthalate salt, achieving close to 100% PET conversion. Typically, the reaction runs for 3–5 h at 210–250°C under a maintained pressure of 1.4–2 MPa.^[184] A major drawback of this method is the presence of impurities in the recycled PET, which can constitute up to 40% by weight.^[146] Yoshioka et al. demonstrated that simultaneous hydrolysis and oxidation in the presence of concentrated NaOH during the recycling of PET flakes effectively yields TPA and oxalic acid.^[185] In another study, Polk et al. employed a phase transfer catalyst (triethylmethylammonium bromide) in the alkaline hydrolysis reaction at lower temperatures (70–95°C), achieving 99.6% high-purity TA with up to a 93% yield.^[186] Kosmidis et al. conducted a thorough analysis of the kinetics involved in the depolymerization reaction of PET flakes, utilizing a phase transfer catalyst to facilitate the process.^[187]

Neutral hydrolysis is performed in high-pressure autoclaves at temperatures of 200–300°C and pressures of

1–4 MPa, utilizing either hot water or steam.^[166] This method yields low-purity TPA and has a relatively slow reaction rate due to the weak nucleophilic nature of water.^[166] Moreover, excessive water (water: PET ratio 5:1) is required for the complete depolymerization of PET.^[188]

As previously mentioned, during the methanolysis process, PET is broken down into its constituent components using methanol at elevated temperatures and pressures, resulting in DMT and EG as the primary products. Typically, PET flakes undergo methanolysis at temperatures ranging from 180 to 280°C and under pressures of 2–4 MPa.^[168,189] The DMT and EG produced can be used to resynthesize PET via a transesterification process. Usually, methanolysis is applied to various materials like discarded bottles, fibre waste, leftover films, and plant waste. Transesterification catalysts like magnesium acetate, zinc acetate, lead dioxide, and cobalt acetate are often used to catalyze the reaction, with zinc acetate being the most commonly employed catalyst.^[144] The reaction product of methanolysis includes a mixture of alcohols, glycols, and phthalate derivatives in addition to DMT. The costly separation and refinement of reaction products represent a significant drawback of methanolysis.^[144]

Aminolysis, a newly developed technique for the depolymerization of PET waste, has not yet been adopted in commercial PET recycling. Additionally, the amines' expensive and often toxic nature hinders its large-scale application. In general, aminolysis is conducted using primary amines (methylamine, ethylamine, and ethanolamine) within a temperature ranging from 20 to 100°C, resulting in the production of mono- and di-amines of TPA and EG.^[144] Shukla and Harad conducted aminolysis on PET waste fibres and bottles using an excess of ethanolamine, with various catalysts such as sodium acetate, glacial acetic acid, and potassium sulphate. This reaction results in bis (2-hydroxyethyl)terephthalamide formation, achieving a high purity yield of 91% across all catalysts.^[190] Complete degradation of PET into diamide was achieved through 45-day reactions in different amines.^[191] Teotia et al. achieved the conversion of PET into low molecular weight oligomers at ambient temperature and pressure by using four different amines (ethylenediamine, ethanolamine, methylamine, and butylamine).^[192]

5 | RECYCLING OF POLYOLEFINS WASTE (PP, HDPE, AND LDPE)

Plastics derived from polyolefins, that is, PP, HDPE, and LDPE, are essential commodity materials synthesized from fossil hydrocarbons through addition

polymerization. These plastics are extensively used in various applications, including packaging materials, grocery bags, toys, containers, pipes, industrial wrappings, and films.^[20] Consequently, polyolefins waste significantly contributes to post-consumer plastic waste. Depending on the level of contamination and the quality of the polyolefin waste materials, mechanical and chemical recycling, as well as energy recovery processes, can be employed.

5.1 | LDPE

PE is commonly produced through polymerization of ethylene (C₂H₄), often aided by a Ziegler–Natta or metallocene catalyst.^[193] LDPE is synthesized through free radical polymerization, using organic peroxide or oxygen as initiators, in autoclave or tubular reactors under high pressures and temperatures.^[194] The branched architecture of LDPE inhibits the close packing of polymer chains, leading to a lower density and reduced crystallinity. Consequently, LDPE exhibits high ductility and flexibility. LDPE is generally utilized in applications where resistance to high temperatures or structural strength is not required, including plastic bags, stretch film, and squeezable bottles.^[195] The significant volume of discarded LDPE has resulted in severe environmental concerns.^[196,197] Jin et al. studied the effect of extensive mechanical recycling on the properties of LDPE.^[198] Rheological measurements revealed simultaneous chain scission and crosslinking in LDPE. However, one mechanism appears to dominate over the other during specific extrusions. For instance, during the initial extrusion, chain scission slightly outweighs crosslinking, whereas crosslinking becomes predominant from the second extrusion cycle.

Figure 10A illustrates the storage modulus (G') and loss modulus (G'') across various extrusion cycles as a function of angular frequency at 240°C. The reduction in the angular frequency at which G' and G'' intersect suggests that the relaxation time of the polymeric chains increases with each extrusion cycle. This observation supports the hypothesis that crosslinking occurs during the extrusion process. The authors observed a substantial decrease in the melt flow index (MFI) of pristine LDPE, dropping by two orders of magnitude from 2.25 g/10 min to an unprocessable MFI of 0.02 g/10 min following extrusion cycles, as depicted in Figure 10B.

The reduction in MFI in consecutive extrusion cycles indicates a decrease in the flowability of molten LDPE. This outcome is ascribed to crosslinking or molecular enlargement. Additionally, an increase in reprocessing cycles leads to a reduction in the number average

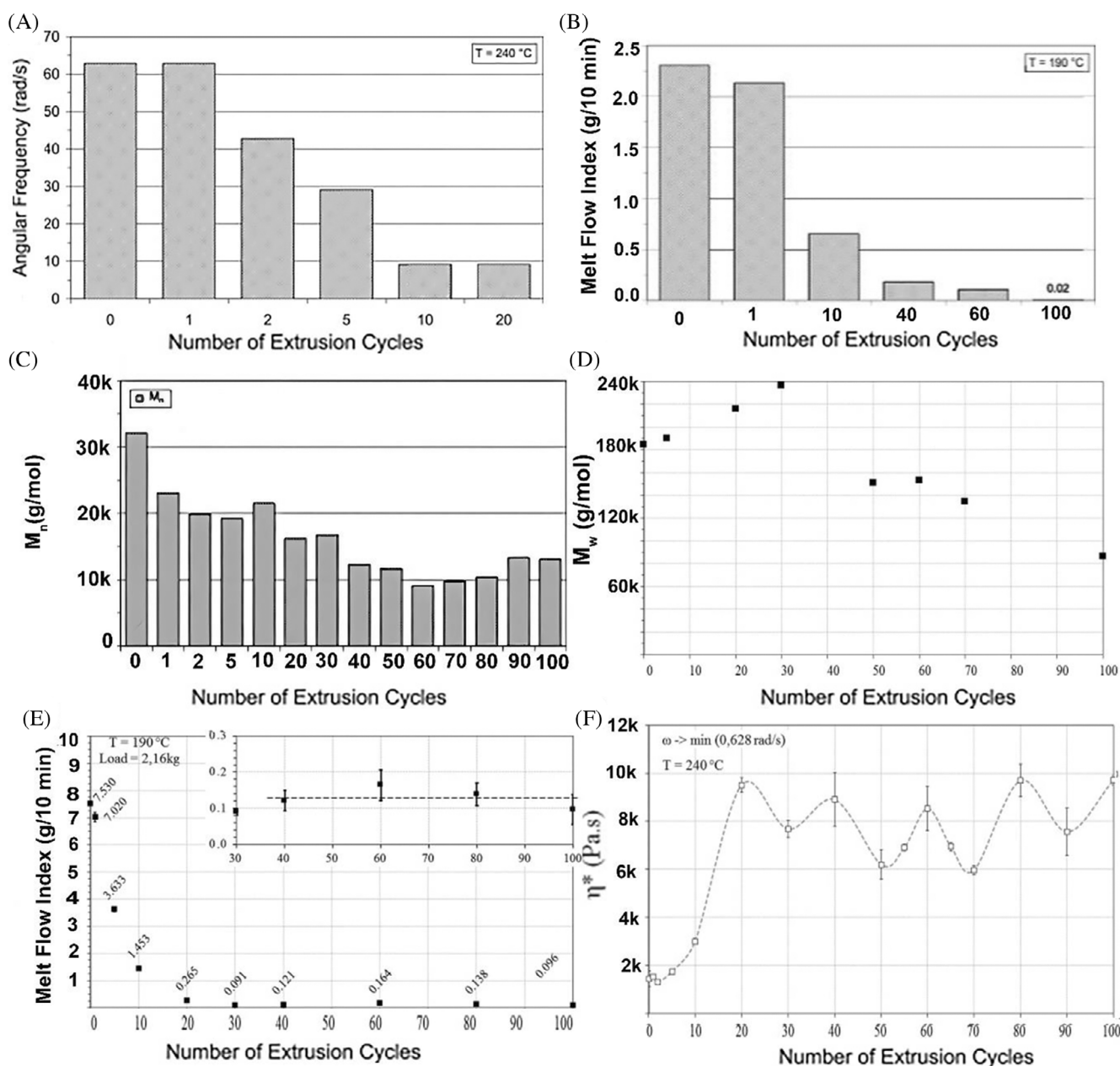


FIGURE 10 Effect of the number of extrusion cycles on various processing parameters of low-density polyethylene (LDPE) and high-density polyethylene (HDPE): (A) angular frequency at which G' and G'' intersect, (B) melt flow index (MFI), and (C) M_n for LDPE. (D) Variation of M_w , (E) MFI, and (F) complex viscosity for HDPE as a function of the number of extrusion cycles. (A–C) Adapted from Jin et al.^[198] Copyright 2012, with permission from Elsevier. (D–F) Adapted from Oblak et al.^[98] Copyright 2015, with permission from Elsevier.

molecular weight (M_n) (Figure 10C), suggesting an increase in low molecular weight materials. This confirms the occurrence of chain scission during consecutive extrusion cycles. However, the authors noted that after the 50th extrusion cycle, molecular weight distribution (MWD) parameters showed a significant decline. This deterioration in MWD parameters is likely indicative of more extensive polymer degradation. This degradation results in reduced crystallinity and increased creep compliance. Therefore, these results provide further evidence

that chain scission, molecular enlargement, and crosslinking occur concurrently during mechanical recycling. The authors concluded that LDPE retains its processability and long-term mechanical properties for up to 40 extrusion cycles due to the competing mechanisms of chain scission and crosslinking.

As mechanical recycling becomes impractical, pyrolysis emerges as an optimal method for recovery due to the structural variability and relative chemical inertness of PE. Both catalytic and thermal pyrolysis methods are

utilized for LDPE recycling. During thermal pyrolysis, long polymer chains undergo thermal degradation, breaking down into shorter molecular fragments at elevated temperatures. This process demands a significant amount of energy and typically occurs at elevated temperatures. Yan et al. applied a modified Coats–Redfern method to carry out LDPE pyrolysis, yielding 21.30 wt.% (C_6 – C_{12}) gasoline and 67.76 wt.% (C_{13} – C_{22}) diesel in the cracked product at 460°C.^[199] In another investigation, the thermal pyrolysis of LDPE resulted in liquid products comprising 78.4% hydrocarbons (C_{10} – C_{27}), 17.2% solid residue, and 4.4% organic gases at temperatures ranging from 201 to 400°C.^[109] The presence of waxes and other by-products negatively impacts the quality of the end products. Employing catalysts in the pyrolysis process can enhance the selectivity for specific hydrocarbons in the final product. Wong et al. explored the effect of the ZSM-5 catalyst on productivity and selectivity. They achieved a 98.6% conversion of LDPE and a 99.5% liquid yield, with product selectivity optimized by adjusting various reaction parameters such as temperature, feed rate, solvent quantity, and catalyst amount.^[200]

In a different investigation, HZSM-5 underwent modification with P and Zn to enhance the selectivity of monocyclic aromatic hydrocarbons (MAHs). The reaction occurred in a two-stage fixed bed reactor at 500°C using Zn-P/HZSM-5 catalysts, resulting in a liquid yield of 55.9 wt.%. Notably, a high selectivity of 87.44% was achieved for MAHs, with benzene, toluene, ethylbenzene, and xylene content reaching 75.16%.^[201] Janajreh et al. utilized a gasification process to recycle PE, PP, PS, and a mixture of these plastics. They observed that air gasification resulted in efficiencies of 59.03%, 62.73%, and 73.13% for PE, PP, and PS, respectively. However, the gasification of a mixture containing PE, PP, and PS in the same study showed an increase in efficiency, reaching up to 89%.^[132]

5.2 | HDPE

Globally, HDPE is a common component of the waste stream and typically ranks among the top plastics, alongside PET, PP, and LDPE. Its precise position varies regionally based on usage and recycling patterns. HDPE exhibits higher tensile strength, melting point, chemical resistance, stiffness, and opacity than its LDPE counterpart. The reason behind improved properties is ascribed to the lower branching and long linear chains in the structure of HDPE.^[202] Various types of HDPE can be prepared by varying the molecular weights through heterogeneous catalyst polymerization. Due to its high tensile strength and stiffness, HDPE is commonly utilized

for manufacturing toys, containers, and reusable bottles.

As discussed in Section 3.7.1 on mechanical recycling, the mechanical properties and processability of recycled plastics are notably influenced by elevated shear force, temperature, and the presence of oxygen. Nevertheless, the degradation mechanisms of LDPE, HDPE, and PP in mechanical recycling differ despite all belonging to the polyolefin group.^[203] Loultcheva et al. recycled HDPE containers and observed that rheological and mechanical properties heavily rely on reprocessing conditions, including residence time, temperature, and applied stress. They found that by employing apparatus with short residence times, mechanical and rheological properties stay nearly unchanged even after five extrusion cycles, resembling virgin material.^[204] Similarly, an intriguing investigation on the impact of extrusion cycles on HDPE degradation behaviour (at 240°C) during 100 consecutive extrusion cycles was conducted by Oblak et al.^[98] In the initial 30 cycles, HDPE exhibited a reduction in chain mobility as a result of dominant chain branching, as evidenced by a notable increase in weight average molecular weight (M_w), complex viscosity (η^*), and a significant decrease in MFI, as shown in Figure 10D–F. M_w is sensitive to large molecules, so its increase during the first 30 extrusion cycles supports the assumption of a branching process (Figure 10D).

As seen in Figure 10E, MFI experienced a substantial decrease in the first 30 extrusion cycles, followed by a moderate rise in the subsequent cycles. The MFI index reveals changes in material flowability, signifying variations in the polymer's molecular structure. The decrease in MFI indicates reduced chain mobility, suggesting that a chain branching process occurs throughout the first 30 reprocessing cycles. In addition, the study examined the dependence of complex viscosity on the extrusion cycle at a low frequency of 0.628 rad/s for HDPE (Figure 10F). The complex viscosity increased by 600% from the initial value by the 20th cycle, likely due to reduced chain mobility caused by chain branching. As shown in Figure 10F, after the 20th reprocessing cycle, the complex viscosity fluctuated between 400% and 700%. The authors observed a decrease in complex viscosity values at high frequencies, indicating chain scission. Furthermore, the literature indicates that chain scission and chain branching occur simultaneously during the degradation of HDPE in processing.^[204,205]

According to these findings, it was concluded that during the initial 30 extrusion cycles, the molecular structure is primarily dominated by chain branching. However, after the 30th cycle, chain scission started to prevail over chain branching. Following the 60th cycle, the branched molecular chains commence cross-linking, an

effect that intensifies with increasing extrusion cycles. Nevertheless, up to the 100th extrusion cycle, chain scission remains the dominant process despite nearly a quarter of the material undergoing cross-linking. Consequently, the polymer's degradation during extrusion restricts its potential for further mechanical recycling.

Recently, the influence of mechanical recycling on HDPE's molecular structure and rheological properties has been assessed by Zhang et al.^[206] Variations in the molecular structure and rheological behaviour were analyzed using various techniques. The results showed that the first four recycling cycles did not significantly affect the rheological properties, but a substantial increase in zero-shear viscosity by a factor of 6 occurred between cycles 4 and 8. However, there was an 11% decrease in the M_w between the 1st and 16th extrusion cycles. The analysis of rheological behaviours suggested the occurrence of possible chain branching in the recycled HDPE material. The study excluded the possibility of extensive crosslinking (gelation) based on mass recovery results from size exclusion chromatography. NMR spectra revealed randomly branched structures, which were attributed to thermo-mechanical degradation. These findings suggest that the formation of random branching, including short-chain branching, contributes to the observed increase in viscosity. However, further research is necessary to fully understand the degradation mechanisms and their effects on the mechanical properties of recycled HDPE. This could facilitate the development of appropriate additives to maintain the material's properties and applications.

Different additives have been incorporated during processing to enhance mechanical properties and minimize degradation, leading to improved mechanical strength of the recycled polymer. These additives include stabilizers, which prevent degradation during mechanical recycling. These stabilizers effectively neutralize the free radicals produced during reprocessing, safeguarding against degradation caused by environmental factors such as UV radiation and free oxygen. Phenol-based compounds are predominantly favoured as stabilizers in the mechanical recycling of polyolefins. Researchers have also utilized a blend of phenolic and phosphate-based antioxidants during recycling to extend the shelf life of recycled plastics.^[207] Recently, researchers have begun exploring bio-based stabilizers like caffeine, curcumin, ascorbic acid, and others as alternatives to inorganic or phenolic compounds.^[208,209] These substances are introduced into the polymer during melt processing, often called reactive extrusion. This method is commonly used for functionalization, post-polymerization reactions, cross-linking, and grafting.^[210]

Various chemical recycling methods have been employed to recycle HDPE. Pyrolysis emerged as an excellent option to chemically recycle HDPE due to the absence of aromatics and other heteroatoms in its structure, making it ideal for achieving high liquid yields. Pyrolyzing HDPE plastic bags at temperatures of 420–440°C, followed by distillation, yielded a significant amount of liquid hydrocarbon. After adding antioxidants, the resulting product met all ASTM D975 and EN590 fuel specifications, except for density in the case of EN590.^[211]

In another study, the thermal decomposition of HDPE waste produced products consisting of gasoline, kerosene, and diesel-grade hydrocarbons.^[212] Additionally, they observed that longer residence times led to higher pyrolysis oil yields. Bäckström et al. recycled HDPE using a microwave-assisted hydrolysis process in the presence of nitric acid, producing three dicarboxylic acids: adipic, succinic, and glutaric acid. These dicarboxylic acids were subsequently used to synthesize plasticizers. When these plasticizers were mixed with PLA, a decrease in the glass transition temperature (T_g) was observed.^[213] Undri et al. recycled HDPE waste utilizing microwave-assisted pyrolysis (MAP). The resulting liquid fraction comprised linear alkanes and 1-alkenes, with minimal production of branched, cyclic, or aromatic hydrocarbons.^[214] Typically, effective pyrolysis of HDPE is achieved at temperatures above 500°C. However, at temperatures exceeding 800°C, there is an increased tendency to produce gaseous products.^[215] According to the literature, the utilization of catalysts during thermal pyrolysis lowers the reaction temperature, improves product selectivity, and facilitates the upcycling of waxes at reduced temperatures.^[216–219]

5.3 | PP

PP is synthesized from propylene (C_3H_6) monomer using different catalysts like Ziegler–Natta or metallocene catalysts. It is typically grouped as a polyolefin polymer with HDPE and LDPE and shares characteristics of being semi-crystalline and non-polar.^[220] It finds applications across the automotive, textile, and consumer goods industries because of its low density, light weight, exceptional heat resistance, and strong durability.^[195] PP is the most abundant plastic in municipal solid waste, with its quantity increasing each year. The presence of a methyl group in the polymeric backbone of PP inhibits chain rotation, thereby increasing its strength but reducing its flexibility.^[221] PP can be recycled through mechanical recycling like other polyolefins.^[222,223]

Furthermore, rapid degradation of the PP backbone occurs after repeated heating cycles. PP undergoes more severe thermal degradation than PE due to the vulnerability of the tertiary carbon atom in the PP backbone to thermo-oxidative and photo-oxidative degradation.^[224] Da Costa et al. investigated PP degradation comprehensively through multiple extrusion cycles.^[225] They examined the impact of repetitive extrusion cycles by assessing factors such as the degree of crystallinity (χ_c), heat of fusion (ΔH_m), and melting temperature (T_m), as well as mechanical properties, including tensile and impact strength. They observed that the χ_c increases with higher die temperatures and a greater number of extrusion cycles. Notably, a significant increase in χ_c was detected at 270°C after 19 extrusion cycles. Specifically, the crystallinity rose from 42.4% at 240°C after the 5th cycle to 51.1% at 270°C after the 19th cycle (as shown in Figure 11A).

The increase in χ_c values with rising temperature and the number of extrusion cycles is due to heat, thermo-oxidative, or mechanical degradation, which releases entangled macromolecular segments, allowing further crystallization. This degradation reduces the concentration of entanglements and tie chain segments that initially hinder crystallization in PP. As a result, the strained or entangled macromolecular sections are freed, enabling their rearrangement and promoting additional crystallization (χ_c increases). Correspondingly, the increase in crystalline fraction is accompanied by a rise in ΔH_m , as confirmed in Figure 11B. Furthermore, a decrease in T_m values was noted with an increase in die temperature or extrusion cycles, as illustrated in Figure 11C. This was attributed to the well-known fact that T_m of PP generally decreases with a reduction in molar mass^[226] and with an increase in stereo-irregularities.^[227] Thus, continuous thermo-mechanical degradation of PP leads to a broader molecular weight distribution and a consequent reduction in melting temperature (T_m).^[225,228]

A similar trend was reported by this research group in the correlation between rheological data and multiple extrusion cycles. To this end, they investigated how PP degrades when subjected to multiple extrusions at various temperatures by examining changes in its rheological properties. The study found that repeated extrusion processes, especially at higher temperatures, significantly altered PP's rheological behaviour. This was evident from a marked increase in the MFI, indicating a rapid rise in chain scissions and a consequent drop in molecular weight. This resulted in a noticeable decrease in the complex viscosity and elasticity of the molten polymer. The variation of complex viscosity versus frequency for PP extruded at two different die zone temperatures, 240 and

270°C, is shown in Figure 11D,E. This research highlighted the importance of controlling extrusion parameters to minimize the loss of properties in recycled PP.^[228] Usually, stabilizers are incorporated during PP extrusion to mitigate radical attacks on the polymer chains.^[210,229] In a one-step stabilization process of the recycled polymer, researchers introduced electron-donating groups (EDG), such as maleic anhydride, onto the primary polymer chains. These groups internally stabilize the polymer by scavenging free radicals, thus preventing chain scission during polymer processing akin to traditional stabilizers.^[230]

Enabling the mechanical recycling of polyolefins while preserving their mechanical properties can be achieved by substituting some covalent bonds or cross-linking in the polymeric material with dynamic bonds. This replacement of a few covalent bonds with dynamic bonds can enhance properties such as toughness, additive properties, and recyclability.^[233] Kar et al. modified polyolefins with maleic anhydride and combined them with epoxy anhydride to introduce a dynamic bond, resulting in a mechanically stable recyclable PE or PP composite. This method ensures consistent mechanical properties even after three recycling cycles, with the samples demonstrating both weldability and self-healing capabilities, as shown in Figure 11F.^[231] In another study, Dey et al.^[232] investigated the influence of varying dynamic networks on the mechanical characteristics of post-consumer recycled PP (PCR-PP). They incorporated maleated castor oil (MCO) into PCR-PP and facilitated its reaction with epoxidized castor oil (ECO) and silyl ether to create dynamic bonds during the recycling of PCR-PP waste. The recycled PP maintained a comparable Young's modulus even after five cycles of recycling, as shown in Figure 11G,H. This method is significant as it could prolong the lifespan of recycled plastics, reducing the need for chemical recycling.

Chemical recycling has emerged as a viable method for converting PP waste into valuable products, offering an alternative to conventional recycling approaches. Sogancioglu et al. pyrolyzed the PP waste at temperatures ranging from 300 to 700°C. At 700°C, the pyrolysis conversion ratio peaked at 97.83%. The resulting char at 700°C exhibited an increased aromatic structure compared to chars obtained at lower temperatures.^[234] Parku et al. examined the impact of heating rate and vacuum conditions at four distinct temperatures (450, 488, 525, and 600°C) using both slow (15°C/min) and fast (approximately 180°C/min) heating rates. Promising yields of condensable products were obtained at 488 and 525°C under both atmospheric and vacuum conditions, with total yields ranging from 81 to 93 wt.%.^[235]

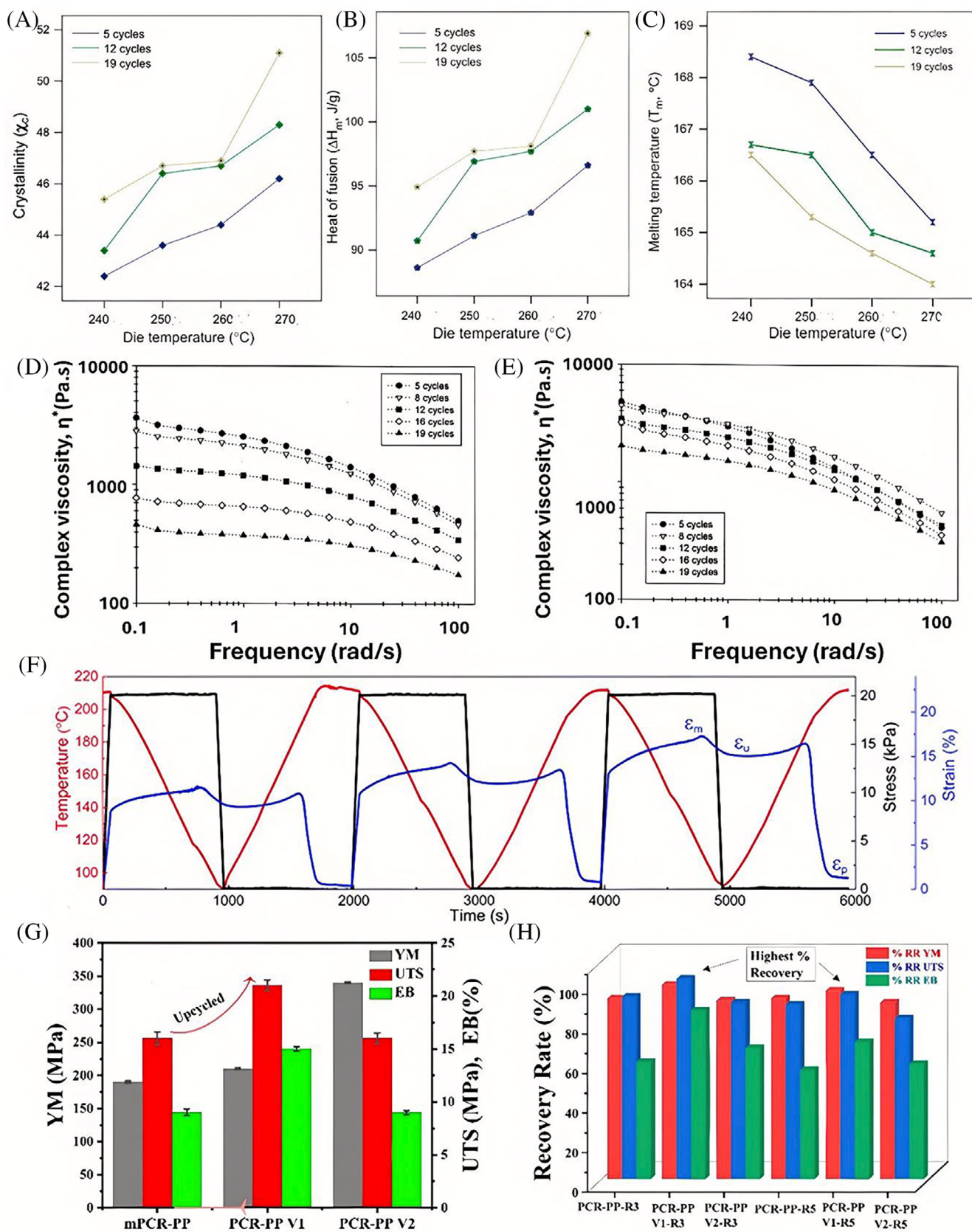


FIGURE 11 Legend on next page.

FIGURE 11 Variation of (A) crystallinity (χ_c), (B) heat of fusion (ΔH_m), and (C) melting temperature (T_m) with die temperature and extrusion cycles. (D) Complex viscosity of polypropylene (PP) extruded with die zone temperature 240°C and (E) 270°C. (F) Three thermomechanical cycles showing the repeatability of fixity and recovery of PP vitrimer, (G) alterations in Young's modulus (YM), ultimate tensile strength, and elongation at break for recycled vitrimer. (H) Recovery rate (%) after the 3rd and 5th recycling cycles for the recycled vitrimer samples. (A–C) Adapted from da Costa et al.^[225] Copyright 2007, with permission from Elsevier. (D–E) Adapted from da Costa et al.^[228] Copyright 2005, with permission from Elsevier (F) Adapted with permission.^[231] Copyright 2020, The Royal Society of Chemistry. (G–H) Adapted with permission.^[232] Copyright 2023, American Chemical Society.

Catalytic pyrolysis has been utilized in the recycling of PP waste, with recent advancements aimed at overcoming catalyst deactivation caused by coke deposition and improving fuel and hydrogen production. For instance, NiO was incorporated into UiO-66(Ce), resulting in a NiO@CeO₂ catalyst with a highly porous structure. This catalyst demonstrated higher hydrogen yield and reduced coke production, thereby extending the catalyst's lifespan. With a 15% loading of NiO/CeO₂, the catalyst exhibited no carbon deposition and achieved a notably high hydrogen production of 90.3 mmol/g. Additionally, using a 30NiO@CeO₂ catalyst, the maximum hydrogen yield reached 137.0 mmol H₂/g LDPE without any coke formation.^[236] Kassargy et al. conducted catalytic pyrolysis of PE and PP with USY zeolite catalyst (in ratio 1:10) at 450°C. The resulting liquid fractions of PP and PE were abundant in C₅–C₁₁ and C₁₀–C₁₃ compounds, respectively.^[237]

PP has been recycled using another chemical recycling technique known as liquefaction, which typically produces short alkyne chains. Ahamed Kameel et al.^[238] conducted liquefaction of recycled polypropylene (rPP) using n-hexane in a batch reactor, achieving a peak oil yield of 88.2% at 400°C. The resultant oil primarily contained alkanes, alkenes, and alcohols, with carbon numbers ranging from C₆ to C₂₀. Seshasayee et al. explored the hydrothermal liquefaction (HTL) of PP waste and found that it requires lower temperatures compared to pyrolysis.^[239] They achieved a maximum bio-oil yield of 32% by liquefying PP at 425°C for 30 min.

Bai et al. investigated the gasification of PP waste using supercritical water gasification technology.^[102] They employed response surface methodology (RSM) to determine the optimal gasification conditions. The study used a quartz tube reactor within a temperature range of 500–800°C for durations of 2–60 min under a pressure of 23 MPa, with feedstock concentrations ranging from 5% to 25% by weight. They found that higher gasification temperatures favour the process while increasing feedstock concentration beyond optimal levels led to insufficient gasification. Additionally, the use of seawater was shown to facilitate the gasification of marine microplastic waste. Results indicated that aromatization of fragments occurs readily in the initial stage of gasification, leading

to tar formation. The application of catalysts significantly lowered the gasification temperature and enhanced the production of desired products.

Wu et al. investigated the use of mixed metal alloys, primarily nickel-based (Ni-Al, Ni-Mg-Al, and Ni-Cu-Al), as catalysts for the gasification of PP waste.^[240] Their study demonstrated that incorporating magnesium into the Ni-Al catalyst led to a higher amount of reacted water and improved catalyst performance in terms of reducing coke generation. However, it did not significantly enhance hydrogen production. The literature indicates that noble metal catalysts, specifically those based on rhodium (Rh) and ruthenium (Ru), exhibit higher efficiency and reduced coke formation compared to nickel (Ni)-based catalysts in steam reforming applications.^[241] Park et al. carried out a reaction in a continuous reactor in the presence of a Ru-based catalyst.^[242] They found that at 673 K with a 5 wt.% catalyst, the residue was significantly reduced, leading to higher fuel generation.

In summary, polyolefin plastics such as PP, HDPE, and LDPE are extensively utilized across various industries, significantly contributing to post-consumer plastic waste. Effective management of these materials relies on mechanical and chemical recycling processes. Mechanical recycling, however, encounters challenges related to material degradation, which drives the investigation into chemical recycling methods such as pyrolysis, solvolysis, and gasification. While these chemical methods offer promising solutions, they also present limitations, highlighting the necessity for further research to develop more efficient and economically viable recycling technologies for polyolefins at scale.

6 | RECYCLING OF PS AND PS-BASED THERMOPLASTIC WASTE

PS is a thermoplastic polymer widely utilized in disposable food containers, packaging materials, and consumer goods. Despite its versatility and wide-ranging applications, recycling PS poses notable challenges due to its low density and limited demand for recycled PS. In recent years, researchers have explored various techniques to improve the efficiency and effectiveness of mechanical

recycling for PS. These include modifying the process conditions, incorporating additives or fillers, and investigating the impact of recycled material properties on the final product. One approach to PS recycling is mechanical recycling, which involves breaking down the polymer into smaller particles or pellets and using them as raw material for new products.

Seyedi et al.^[243] investigated the use of bottlebrush-modified graphene oxide (GO-P) as a compatibilizer to enhance the mechanical properties of recycled PS/PP blends. They compared the effectiveness of this approach with virgin GO (GO-V) and examined the impact of mixing order on blend properties, including morphology, rheology, and mechanical properties. The study revealed that modified GO-P was located at the interphase of PS/PP blends, while GO-V migrated into the PS phase (Figure 12A).

Incorporating GO-V into the PS phase before blending improved strength and strain by 9% and 16%, respectively, compared to the unmodified PS/PP blend. Conversely, pre-mixing GO-P with PS resulted in a 16% decrease in flexural modulus and a 21% increase in flexural strain compared to the unmodified PS/PP blends (Figure 12B). These results highlight the potential of GO-P as a promising compatibilizer for the mechanical recycling of PS/PP blends.

In another effort, Hamad et al.^[244] investigated the impact of recycling on a polymer blend comprising PLA and PS. They subjected the blends to multiple extrusion and injection moulding cycles and then evaluated the rheological and mechanical properties of the recycled materials. The findings revealed that the recycled materials exhibited lower viscosity and inferior mechanical properties compared to the original blend, with the level of degradation commensurate with the number of recycling cycles. Each processing cycle led to a significant decrease in the apparent viscosity of the blend by a factor of 0.15–0.3. After four consecutive processing cycles, Young's modulus decreased by a factor of 0.26. The authors proposed that these changes in properties were attributed to thermal and mechanical degradation, as well as reductions in molecular weight and chain length during the recycling process.^[244]

In similar research, the effect of repeated processing cycles on the mechanical and rheological properties of the high-impact polystyrene (HIPS)/clay nanocomposites and the distribution of the clay nanoparticles within the matrix were evaluated.^[245] Since recycled plastics suffer from inferior physical and structural properties, the authors incorporated clay nanoparticles (Cloisite 20A) into HIPS to address these issues. Intercalated morphology was first observed during the initial cycle of melt-processing via extrusion and remained

consistent across subsequent cycles. There was a slight dependency of the MFI of neat HIPS and its nanocomposites to reprocessing cycles, in which this parameter fell and then rose, respectively (Figure 13A). They mentioned that reprocessing cycles could enhance the degree of exfoliation or intercalation of clays in the polymer matrix and promote a decrease in MFI. This can stem from restraining polymer chain movements as well as their diffusion into clay layers. They concluded that these outcomes emphasize the role of thermo-mechanical and thermo-oxidative degradation in flow behaviour.

They observed that the tensile strength of neat HIPS and its nanocomposites containing 3 wt.% clay slightly decreased during the second and third extrusion cycles but showed a marginal increase in subsequent cycles (Figure 13B).^[245]

Hirayama and Saron^[246] explored the impact of recycling on the properties of a polymer blend comprising recycled acrylonitrile-butadiene-styrene (rABS) and HIPS sourced from waste electrical and electronic equipment (WEEE). They emphasized the challenge of precise polymer separation from WEEE and examined the morphological and mechanical properties of blends with different HIPS and rABS weight ratios. On the basis of SEM images, they concluded that the phase inversion occurred upon increasing the loading of ABS from 25 to 75 wt.%. In this context, spherical domains in blends containing 25 wt.% ABS, with a diameter of approximately 3.0 μm , were transformed into elongated domains with a length of around 4.6 μm and a width of about 1.0 μm in the blend with 75 wt.% ABS (Figure 14A–C).

Remarkably, recycled blends exhibited improvement in mechanical properties, including tensile strength and Young's modulus, compared to virgin polymer-based blends. For instance, in a 50/50 weight ratio, Young's modulus for virgin blends was recorded at 1592 ± 44 MPa, while the recycled blend showed a higher modulus of 1703 ± 22 MPa. This enhancement was attributed to the degradation process in the rubber phase, which increased the blend's rigidity and, in turn, elevated both tensile strength and Young's modulus (see Figure 14D,E).

In another study, the same research group investigated the use of compatibilizers such as styrene-butadiene-styrene (SBS), named as C1, and styrene-ethylene-butylene-styrene/styrene-ethylene-butylene (SEBS/SEB), named as C2, to improve compatibility and mechanical properties in recycled blends of ABS and HIPS. These materials were sourced from WEEE. They melt-blended recycled ABS and HIPS in various weight ratios with the mentioned compatibilizers using a single-screw extruder at 190°C. They compared the resulting properties with those of virgin counterparts. The findings

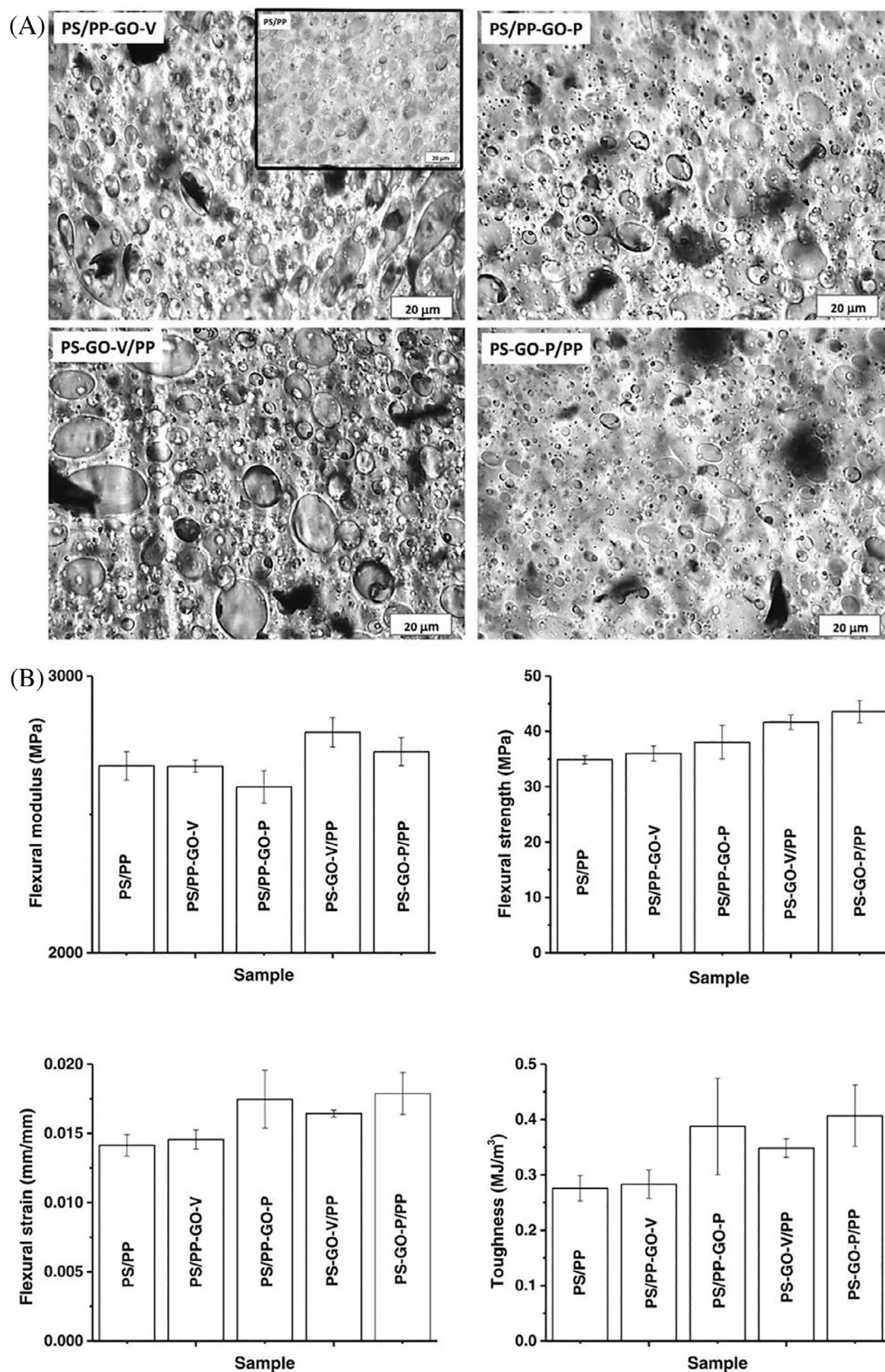


FIGURE 12 (A) Optical microscopy images (with inset image for polystyrene [PS]/polypropylene [PP] pristine blend) and (B) mechanical properties of PP/PS blends containing virgin GO (GO-V) and bottlebrush-modified graphene oxide (GO-P). Adapted with permission.^[243] Copyright 2022, American Chemical Society.

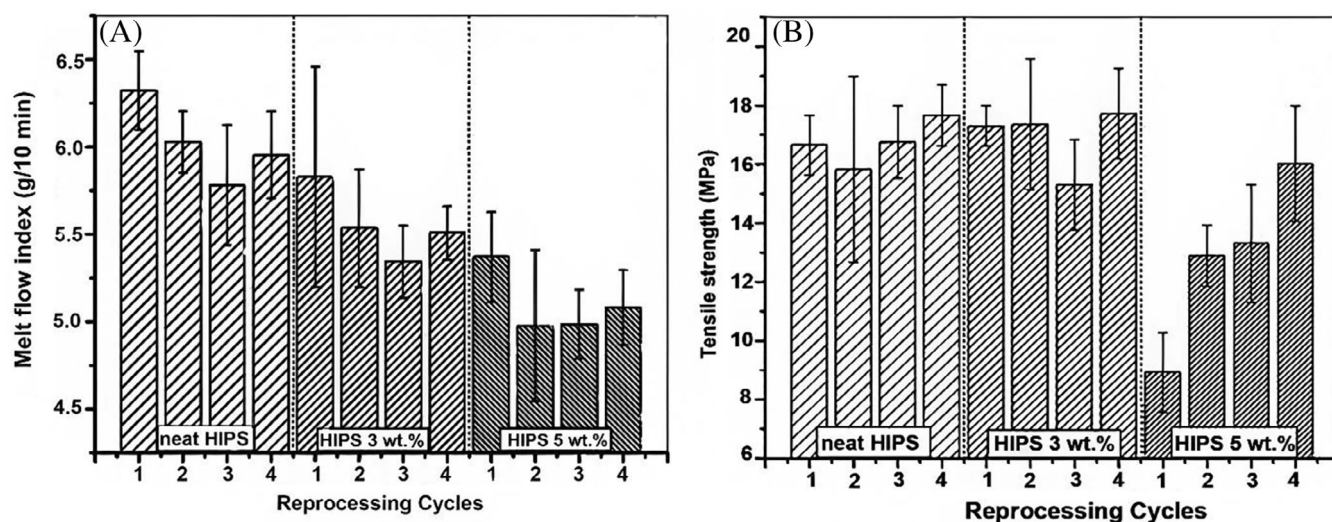


FIGURE 13 Influence of reprocessing and clay content on (a) melt flow index (MFI) and (b) tensile strength of pure high-impact polystyrene (HIPS) and its nanocomposites with 3 and 5 wt.% of Cloisite 20A. Adapted with permission.^[245] Copyright 2007, Elsevier.

revealed that the addition of compatibilizers reduced Young's modulus for both virgin and recycled polymers, signifying a decrease in the hardness of ABS and HIPS. However, both compatibilizers significantly improved the elongation at break for both virgin and recycled polymers by over 50% (see Figure 15A,B). As can be observed in the SEM images (Figure 15C,D), ABS/HIPS blends intrinsically featured a complex phase morphology containing PS, styrene-acrylonitrile copolymer (SAN), and polybutadiene (PB). In this regard, the size of SAN domains in the HrAr-C2 blend ($22 \mu\text{m}^2$) is twice their size in the HrAr-C1 blend ($11.22 \mu\text{m}^2$), confirming the higher elongation at break of the HrAr-C2 blend (Hr, Ar, C1 and C2 denote the rHIPS, rABS, SBS, and SEBS/SEB, respectively). Note that HrAr blends contained 75 wt.% HIPS and 25 wt.% ABS. As a result, compatibilizer C2 was more efficient than C1 for these blends. The authors proposed that incorporating virgin polymers and fine-tuning the compatibilizer loading could be a promising strategy for enhancing the mechanical properties of recycled ABS/HIPS blends derived from WEEE.^[247]

Another approach involved introducing additives or fillers to enhance the mechanical properties of recycled polystyrene (rPS). For instance, Poletto et al.^[248] examined the impact of incorporating wood flour (WF) and a coupling agent on the mechanical properties of rPS/WF composites. They prepared these composites with varying proportions of WF and the coupling agent, poly(styrene-co-maleic anhydride) oligomer (SMA). Subsequently, they conducted mechanical tests to assess the tensile, flexural, and impact strength. The results indicated that adding WF enhanced the mechanical properties of rPS/WF

composites. In contrast, the coupling agent had a diminishing effect. Notably, an increase in the filler content led to a continuous improvement in flexural modulus, attributed to the greater rigidity of wood fibres compared to the polymer matrix. For instance, the flexural modulus for recycled polymer was $3315 \pm 189 \text{ MPa}$, while the value for rPS containing 40 wt.% WF reached $5725 \pm 86 \text{ MPa}$. In contrast, both flexural strength and strain decreased significantly with higher filler loadings. The authors proposed that incorporating WF and a coupling agent could be a viable strategy to improve the mechanical properties of rPS from post-consumer plastic waste.

In a similar attempt, the morpho-structural and thermo-mechanical properties of rPP and rPS derived from mixed post-consumer plastic waste were examined by Rodríguez-Liébaña et al.^[249] The researchers created recycled materials through a blend of extrusion and injection moulding. Subsequently, they conducted extensive tests to assess their morpho-structural and thermo-mechanical characteristics. Remarkably, the recycling process had minimal impact on the material's structure. Additionally, the study showed that the recycled materials displayed thermo-mechanical properties similar to those of the virgin materials, with only minor reductions in certain aspects. Specifically, the tensile strength of the recycled materials was 4%–10% lower than that of the virgin materials, while the flexural strength was barely affected.

Another approach is chemical recycling, which involves breaking down the PS polymer into its constituent monomers, which can then be used as a raw material for new polymers. In this case, BaO powder was exploited as an effective catalyst at 623 K, which had about 90%

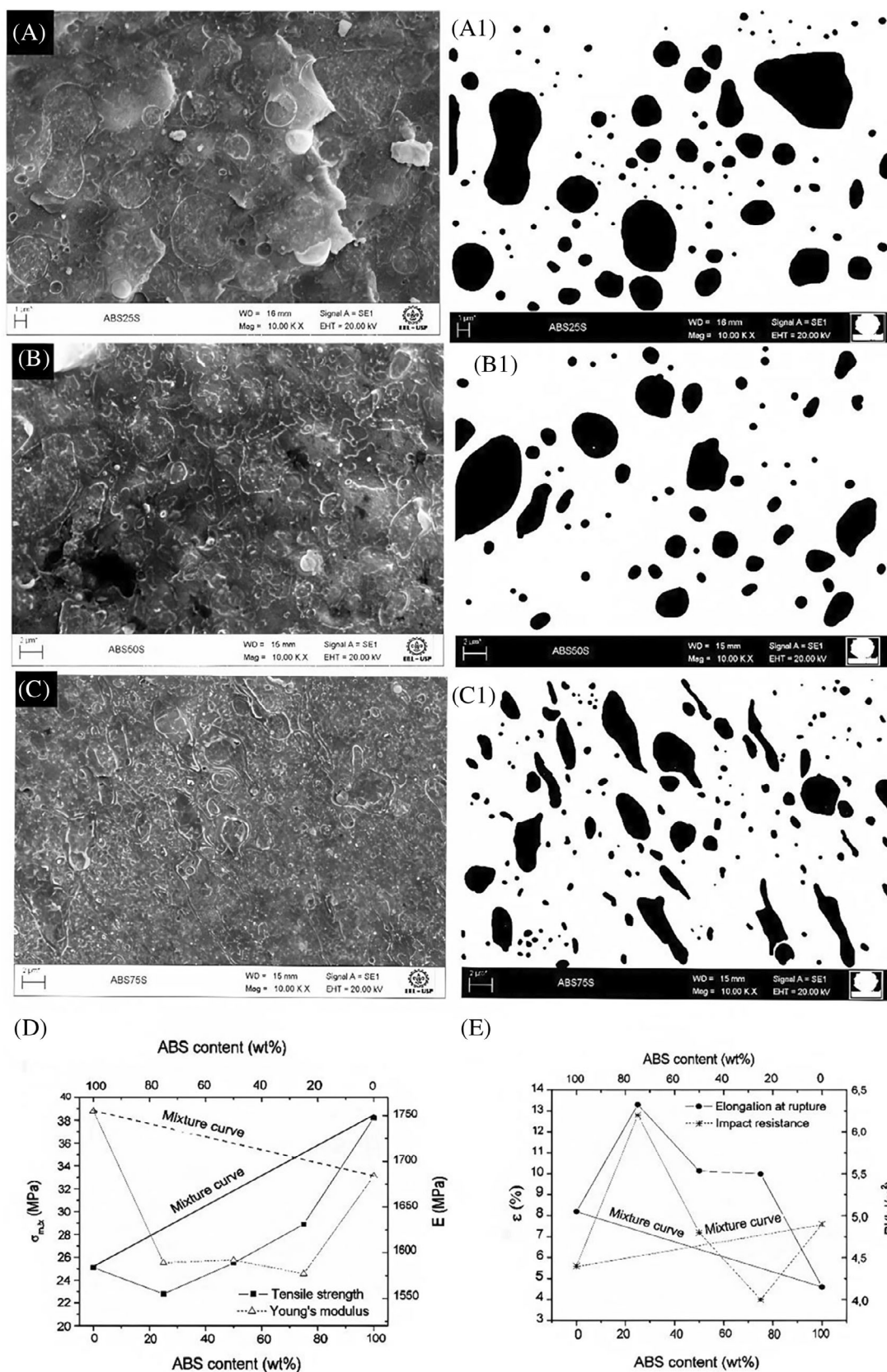


FIGURE 14 SEM images of the acrylonitrile-butadiene-styrene (ABS)/high-impact polystyrene (HIPS) virgin blends containing (A) 25 wt.%, (B) 50 wt.%, and (C) 75 wt.% ABS. Mechanical properties and the mixture curve for virgin blends containing (D) Young's modulus and tensile strength and (E) elongation at break and impact resistance. Adapted with permission.^[246] Copyright 2017, Elsevier.

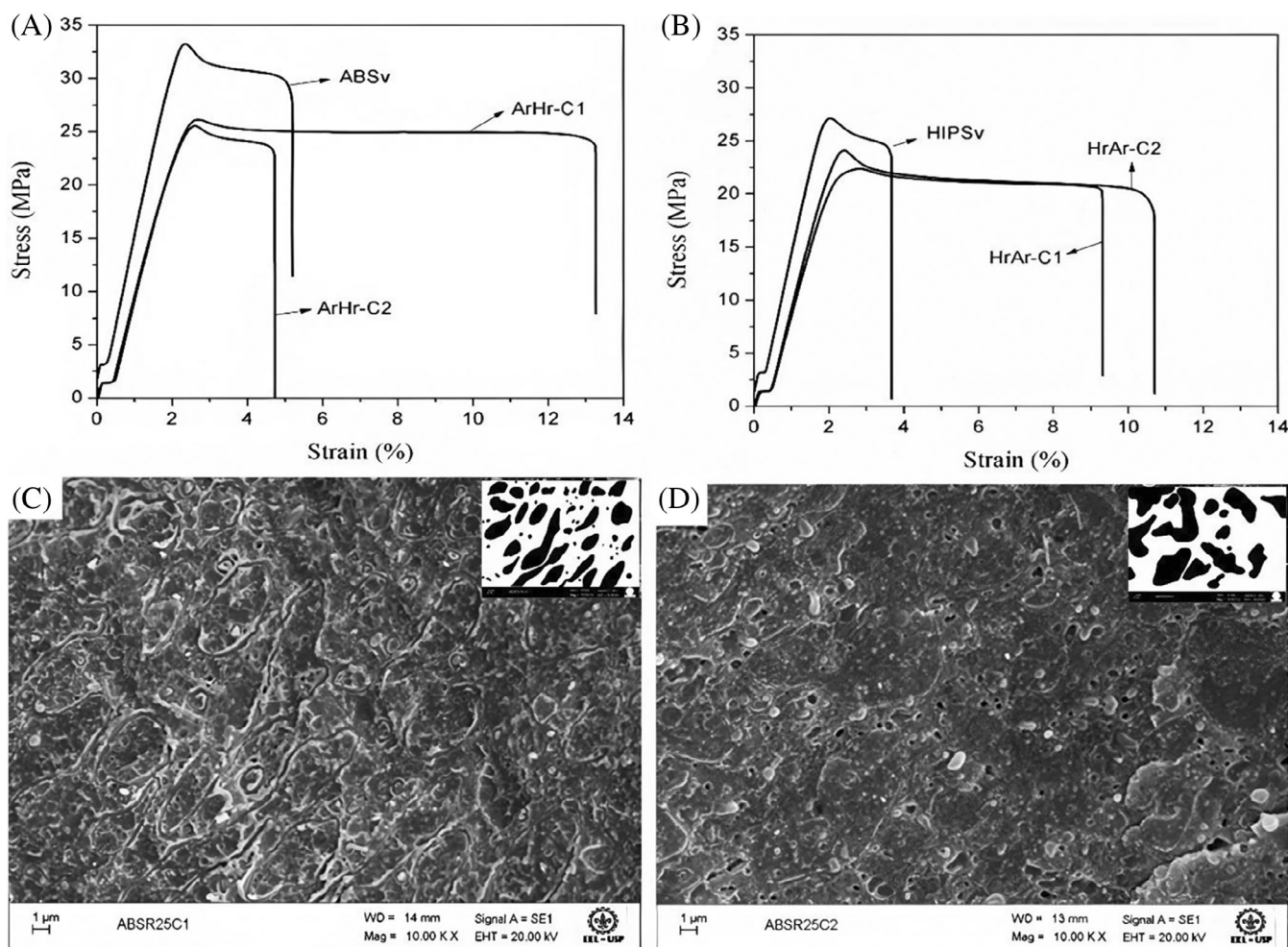


FIGURE 15 Stress–strain curves for recycled acrylonitrile-butadiene-styrene (ABS)/high-impact polystyrene (HIPS) (A) comprising 75 wt.% of ABS and (B) containing 75 wt.% of HIPS. SEM images of recycled ABS/HIPS blends with 75 wt.% of recycled HIPS and 25 wt.% of recycled ABS for (C) HrAr-C1 and (D) HrAr-C2. Adapted with permission.^[247] Copyright 2021, Wiley Periodicals LLC.

efficiency for converting PS into styrene.^[250] Huang et al.^[251] explored using an acid-catalyzed chemical recycling of PS to generate valuable chemicals such as formic acid, benzoic acid, and acetophenone under 1 bar of O₂. The study found that adding hydrogen peroxide improved the yield and purity of the recycled components significantly. Recently, Zeng et al. employed a novel methanol-assisted depolymerization method to extract alkylbenzenes from PS. In fact, they used methanol as a hydrogen source to assist PS depolymerization into alkylbenzenes. This method, termed PS-MAD, employed Ru nanoparticles on SiO₂ as a catalyst, achieving a high yield of liquid products that include valuable monocyclic aromatics and diphenyl alkanes. This process not only efficiently converts PS into useful chemical substances but also significantly reduces the environmental burden of plastic waste by offering an effective recycling pathway.^[252] In a very recent study, the influence of cellulose contamination on the recycling of PS into styrene

monomer via microwave pyrolysis was studied. It was found that pyrolyzing pure PS yielded a high amount of styrene. However, introducing cellulose led to the production of hydrogen. This hydrogen converted some styrene into ethylbenzene and other byproducts, reducing the styrene yield by 29%. Additionally, the presence of cellulose lowered the overall oil yield and increased char production, confirming the necessity of effectively separating contaminants from PS waste to optimize recycling processes.^[253]

Ma et al.^[254] developed a superhydrophobic textile coated with PS waste and SiO₂ nanoparticles. For this purpose, they dissolved PS waste sourced from used food containers and coffee cups in 5 mL of toluene and then combined it with SiO₂ nanoparticles. The prepared precursor was coated on the textile surface via a dip coating. The resulting textiles had a water contact angle of approximately 153°, indicating excellent superhydrophobicity. The resultant coating demonstrated

superhydrophobic and self-cleaning properties, facilitating the separation of the oil/water mixture through adsorption or filtration. The authors propose that their technique could be applied to create a variety of superhydrophobic products from plastic waste, offering a promising solution to the increasing issue of environmental plastic pollution.

Although recycling PS remains a complex challenge, advancements have been made in mechanical, chemical, and biodegradable methods to lessen its environmental impact. Further research is crucial to develop recycling technologies for PS that are not only more effective but also economically feasible for industrial applications.

7 | APPLICATION OF RECYCLED PLASTICS

Recycled plastics are widely employed in manufacturing industries to produce various materials. Utilizing recycled materials offers the dual benefits of cost reduction and waste reduction. Because of their high functionality, lightness, and affordability, polymers are a great and practical material to replace ceramics, wood, and metals.^[43] However, recycled polymers may contain or transport contaminants that restrict their use in food and cosmetic packaging applications. Indoor appliances and gadgets have been made using a sizable percentage of recycled plastic waste. Various parts of air conditioners, refrigerators, electronic gadgets, TV cabinets, and flow fans have been composed of recycled plastics.

Generally, thermoset waste is mechanically disintegrated for recycling and added as a filler to virgin thermoplastics and rubbers to manufacture automobile accessories, bathroom sinks, and door panels.^[255] Okamoto et al. devulcanized EPDM and blended it with PP, resulting in thermoplastic elastomer (TPE).^[256] The TPE derived from recycled plastic exhibited properties comparable to virgin TPE. Liu et al.^[257] demonstrated the suitability of such composites for structural applications. These TPE composites have also revealed potential use in various automotive products, including hoses for vacuum sensing and sunroof drains.^[258]

Numerous investigations have reported an increment in the performance of asphalt after the introduction of plastic waste. Specifically, the addition of plastic waste enhanced the rutting resistance,^[259] fatigue cracking,^[260] moisture resistance,^[261] and stability of asphalt mix,^[262,263] which led to improvement in the performance of the road pavement.^[264] Plastic waste has also been extensively used in construction products such as paving bricks, concrete mixes, and building blocks.^[265,266] The production of plastic timber is one of the intriguing applications of recycled

plastic. Polyolefins, such as HDPE, LDPE, and PP, are primarily used to manufacture plastic timber.^[267] The wood plastic composite (WPC) demonstrated high strength and specific stiffness, biological resistance, low density, and low moisture adsorption.^[43,268,269] The natural fibre recycled plastic bio-composite has gained significant importance in a variety of applications, including automotive, packaging, construction, and structural components.^[270,271]

In addition to its use in construction, plastic waste has been investigated as a potential material for electrodes in microbial fuel cells used for wastewater treatment^[272] and supercapacitors for charge storage.^[273] This is due to their chemical structure, which serves as a potential source for various forms of carbonaceous materials, including porous carbon, carbon dots, carbon nanotubes, and nanofibers.^[274,275]

By applying the thermal degradation methods described earlier, plastic waste can be broken down into olefins and low molecular weight hydrocarbons when heated at high temperatures in anaerobic conditions, resulting in the production of oil.^[276] The efficiency of this process depends on various factors such as sample purity, catalyst usage, temperature, and duration. The produced oil has potential applications as a fuel for internal combustion engines.^[277] Typically, polyolefins such as PP and PE are considered for fuel production via chemical recycling. The oil obtained from the pyrolysis of PE exhibits calorific values similar to diesel, while PP conversion yields oil with lower carbon content. Consequently, burning such fuel leads to higher CO, NO_x, and HC emissions than diesel.^[278] Thermal pyrolysis is particularly suitable for recycling multilayer structures containing different polymers like PP and PE. Plastic waste also finds applications in the textile industry, where it is transformed into long fibres through melt spinning. PET, in particular, exhibits a high drawing ratio, making it suitable for reuse in textile manufacturing. Other polymers like acrylics, elastane, nylon, and PP are also being investigated for textile applications.^[279] Recycled PET fibres are also utilized in membrane production via electrospinning, with applications ranging from oil–water separation to air and smoke filtration.^[280]

In an alternative approach, depolymerization transforms plastic waste into various valuable chemicals. For instance, PET can be converted into TPA and EG using different catalysts or microbial action, as discussed earlier.^[281] Moreover, plastic waste from common applications such as packaging, disposables, and food containers—primarily composed of PVC, PP, and PE—can be catalytically decomposed into C₂ (mainly ethene) fuel.^[282,283] During this process, the catalyst activates the long-chain polymer, which subsequently cracks into smaller chain molecules and oils.

Chong et al.^[284] studied the feasibility of using recycled HDPE as a feedstock for 3D printing, particularly in fused filament fabrication (FFF) printers. The study compared the properties of filaments made from recycled HDPE pellets with those made from ABS pellets, a common material for 3D printing. The results indicated that producing filaments from recycled HDPE pellets is viable, with favourable water rejection, consistent diameter, extrusion rate, and thermal stability comparable to ABS filaments. However, challenges such as warping and adhesion issues in 3D printing with HDPE were noted.

The study highlights the need for further optimization of the extrusion process, an investigation into mechanical and aging properties, and the development of standard characterization methods and databases to enhance the market competitiveness of waste-derived filaments.

In another research, filaments-based polymeric nanocomposite from 100% recycled solid PS, mixed with TiO₂ nanoparticles with mass concentrations up to 40 wt.% were reported by Sevastaki et al.^[285] The fabricated 3D-printed objects were used to produce 3D photocatalytic structures. The 3D-printed rPS/TiO₂ nanocomposites were effectively used as photocatalysts for the degradation of drug residues like acetaminophen. The 3D-printed TiO₂/PS nanocomposite samples exhibited promising photocatalytic properties, achieving a yield of almost 60% after three cycles of reuse in 200 ppm of APAP aqueous solution under UV-A irradiation. The research presented a novel, low-cost alternative method for producing large-scale photocatalysts that are appropriate for practical applications. This study was the first report of the production of photocatalytic devices out of fully recycled PS, with a TiO₂ loading as high as 40 wt.% via 3D printing.

Recently, 3D printing technology has been employed to fabricate value-added products based on recycled PS. For instance, 3D-printed filaments were developed based on rPS and rPP. The recycled polymer blend filament was made from post-consumer expanded polystyrene (EPS) and a single-use PP container with various ratios (Figure 16A).

The results showed that increasing the printing temperature led to enhanced interlayer adhesion and a decrease in the air gap. Based on mechanical properties, the 3D-printed specimens with recycled EPS/rPP filaments had a tensile strength of 32 MPa at a blend ratio of 50/50 and a printing temperature of 240°C. This value was comparable with the tensile strength of a specimen made of neat rPS at 34 MPa. The outcomes of mechanical properties, including the printed specimens' stress-strain curves and tensile strength, are presented in Figure 16B,C. Overall, this study emphasized the viability of using recycled EPS and rPP materials to produce 3D printing filaments with mechanical and rheological properties that compare favourably with those of virgin materials. These findings could contribute to

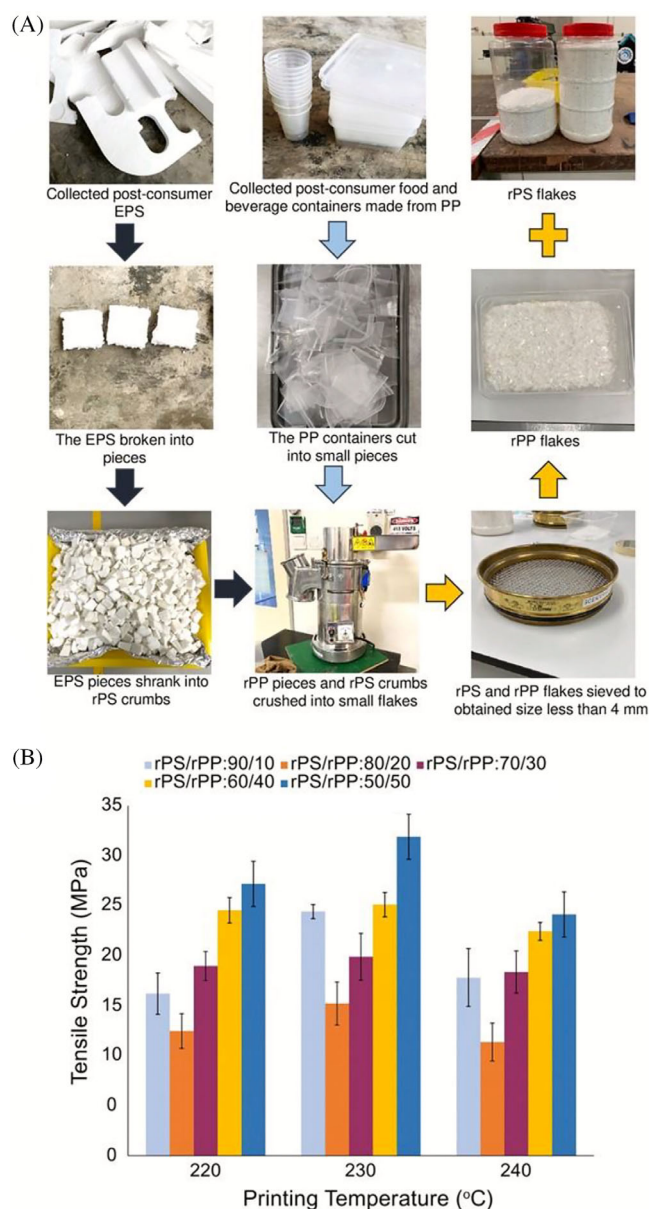


FIGURE 16 (A) Steps involved in preparing recycled polystyrene (rPS) and recycled polypropylene (rPP) from post-consumer expanded polystyrene (EPS) and polypropylene (PP) containers; (B) tensile strength of printed specimens using rPS/rPP blended filament with different blend ratios and printing temperatures. Adapted with permission.^[286] Copyright 2022, Society of Plastics Engineers.

developing more sustainable and efficient manufacturing processes in the 3D printing industry.^[286]

8 | SUMMARY, FUTURE PROSPECTS, AND CHALLENGES

This critical review evaluates the state-of-the-art mechanical and chemical recycling techniques for plastic waste

management. It emphasizes mechanical recycling as the primary and most economically viable method for processing packaging plastic waste, detailing key reprocessing steps such as sorting, decontamination, and separation while also addressing challenges like contamination. The review explores innovative solutions, such as effective disassembly strategies, to improve the quality of mechanically recycled products, which often suffer from quality degradation and price volatility. In response to these challenges, advancements in chemical recycling are highlighted. Although less prevalent, chemical recycling is an expanding approach that breaks down post-consumer polymers into smaller molecules for reuse as chemicals and fuels, offering performance akin to virgin materials. This method shows particular promise for contaminated and mixed waste, where traditional separation methods are impractical or cost-prohibitive.

The review underscores the significant potential of both recycling methods in fostering a circular economy, noting their synergistic effects. It also discusses the application of plastic waste across various sectors and industries. The review concludes by suggesting that energy recovery or landfill disposal may be viable options for managing residual plastic waste, given the current lack of adequate plastic substitutes in many sectors. While current advancements in plastic recycling technology are promising, significant challenges remain that must be overcome to enhance efficiency and scalability. The integration of emerging technologies, such as artificial intelligence for enhanced sorting and robotics for precise and faster processing, could revolutionize the recycling landscape. Economic barriers, particularly the cost competitiveness of recycled plastics against virgin plastics, continue to impede the adoption of recycled materials in mainstream applications. Financial incentives, supportive regulatory frameworks, and greater consumer awareness are critical to increasing the attractiveness of recycled plastics.

8.1 | Enhancing mechanical and chemical recycling methods

Future research should prioritize improving the mechanical properties of recycled plastics, which tend to deteriorate with each recycling cycle. Advanced compounding techniques and incorporating novel compatibilizers could enhance the interface between recycled polymer fractions, thereby improving the material properties of mechanically recycled plastics. Chemical recycling, though still in its early stages, presents a promising avenue for converting waste plastics back into their monomers, enabling a facile recycling process. However, further efforts are needed to

reduce the energy requirements and environmental impact of these processes to better align with sustainable development goals.

8.2 | Developing sustainable materials

The development of bio-based and biodegradable plastics should be accelerated to reduce reliance on fossil fuel-derived plastics. Research into new materials that can match the functionality of conventional plastics while offering enhanced end-of-life options, such as compostability and recyclability, is essential. Additionally, standardizing biodegradable plastics and implementing clear labelling could prevent contamination of recycling streams and promote more effective waste management practices.

8.3 | Legislative and global coordination

Enhanced legislative measures and global coordination are crucial for managing the transboundary movement of plastic waste. Policies that promote design for recycling and extended producer responsibility (EPR) can drive the market toward more sustainable practices. A global agreement on managing plastic waste, similar to the Paris Agreement on climate change, could effectively coordinate efforts and resources, significantly contributing to global plastic waste reduction.

8.4 | Addressing microplastic pollution

As microplastics pollution becomes an increasingly urgent environmental issue, more rigorous methods for tracking and mitigating their release are essential. Innovations in filtration technologies and the development of materials that are less prone to degrading into microplastics can help reduce environmental impacts. Additionally, ongoing research into the effects of microplastics on human health and ecosystems remains critical, requiring sustained attention and funding.

The journey toward a sustainable approach to plastic use and recycling is challenging but filled with opportunity. As technology advances and awareness grows, the potential to significantly reduce the environmental impact of plastics is within reach. The plastic recycling industry must leverage technological innovations, regulatory support, and global cooperation to increase recycling rates and develop sustainable materials that meet societal needs without compromising the environment or human health.

AUTHOR CONTRIBUTIONS

Pradeep Sambyal: Writing – original draft; conceptualization. **Parisa Najmi:** Writing – original draft; conceptualization. **Devansh Sharma:** Writing – original draft; conceptualization. **Ehsan Khoshbakhti:** Writing – original draft; conceptualization. **Abbas S. Milani:** Supervision; resources; funding acquisition; writing – review and editing. **Hadi Hosseini:** Conceptualization; writing – original draft; writing – review and editing; validation. **Mohammad Arjmand:** Conceptualization; writing – original draft; funding acquisition; writing – review and editing; supervision; validation; resources.

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DATA AVAILABILITY STATEMENT

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

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