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Review

Technological Advances in Mechanical Recycling Innovations and Corresponding Impacts on the Circular Economy of Plastics

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Abstract: The impact of plastic pollution on the world and its inhabitants is yet to be fully measured. Significant quantities of microplastics and nanoplastics have been found in human organs, and many diseases have been traced to their presence. Even human placentas have been found to contain microplastics. This study examines the recycling landscape, advanced reprocessing techniques, and technical challenges in this industry. It points out the top recyclable types of plastics (such as high-density polyethylene, polyethylene terephthalate, and thermoplastic elastomers) by analyzing their different recycling capacities globally. It highlights the most advisable recycling techniques by identifying those most successful, least environmentally damaging, and easiest. Mechanical recycling is arguably the easiest and most common recycling technique. This study examines mechanical reprocessing technologies for construction materials, composite boards, additive manufacturing, and other applications. It also points out prevailing setbacks of these approaches and analyzes different solutions. Promising recycling processes are suggested for further investigation.

Keywords: circular economy; mixed plastics; recycling; plastic; waste management

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1. Introduction

The damages caused by plastic pollution cannot be overemphasized. Microplastics have even been discovered in the placenta of newborns [1]. Scientists identified polypropylene and materials used in coatings, adhesives, and personal care products among these placenta deposits. Apart from the roughly interminable life span of plastics, the primary challenge is that due to their manufacturing processes and constituents, they are durable and flow in a linear economy. The summary of the relevant acronyms for this study are highlighted in the abbreviations below:

ABS	Acrylonitrile butadiene styrene
BOPET	Biaxially-oriented polyethylene terephthalate
CPET	Crystalline polyethylene terephthalate
DRAM	Distributed Recycling for Additive Manufacturing
Epoxy	Polyepoxide
FFF	Fused Filament Fabrication
HDPE	High-density polyethylene
LDPE	Low-density polyethylene
LLDPE	Linear low-density polyethylene
MDPE	Medium-density polyethylene

PA	Polyamide
PBAT	Polybutylene adipate terephthalate
PBS	Polybutylene succinate
PBT	Polybutylene terephthalate
PC	Polycarbonate
PCL	Polycaprolactone
PEEK	Polyetheretherketone
PEI	Polyetherimide
PET	Polyethylene terephthalate
PHA	Polyhydroxyalkanoates
PLA	Polylatic acid
PMMA	Polymethyl methacrylate
POM	Polyoxymethylene
PP	Polypropylene
PS	Polystyrene
PTFE	Polytetrafluoroethylene
PTT	Polytrimethylene terephthalate
PU	Polyurethane
PVC	Polyvinyl chloride
PVDC	Polyvinylidene chloride
rPET	Recycled PET
SLS	Selective Laser Sintering
TPE	Thermoplastic elastomer
ULDPE	Ultra low-density polyethylene
VLDPE	Very low-density polyethylene

Plastic packaging (i.e., film wrap, bags, and other packaging materials) is the predominant form of plastic waste globally. In Europe, 39.9% of plastic is employed in packaging [2]. In the US, LDPE, LLDPE, plastic films, and bags are the most abundant waste categories. These packaging materials are ubiquitous as shown in Table 1. Almost all packaging from flimsy films to durable medicine blister wraps is made of plastic, as it is cheaper and easier to use plastics for several applications. Table 1 shows the packaging applications of prevailing plastics.

Table 1. Plastic packaging applications (adapted from [3–5]).

Plastic	Packaging Application
PET	Food-grade containers, bakery trays, peanut butter jars, snack food wrappers, produce containers, soft drink bottles, single-use water bottles, carbonated drink bottles, miscellaneous bottles, bags, jars, tubs, detergent containers, and cleaning containers.
CPET	Plastic food trays, oven-proof plastic wrap, microwavable dinners, microwaveable storage containers, and ready-to-eat meal containers.
BOPET	Faux-foil packaging and microwavable meals' protective film.
PP	Yogurt, cream, cheese, and VSP containers; baby bottles; straws; ready-to-eat meal packs; microwavable kitchenware; salad dressing bottles; margarine tubs; shampoo bottles; straws; margarine tubs; soup packs; syrup containers; and clouded plastic containers.
LDPE	Plastic bags, grocery bags, squeezable food bottles, flexible lids, bread, and frozen food wrappers.
LLDPE	Bottle caps and shrink wraps.
ULDPE	Cheese, meat, and coffee packaging.
MDPE	Baked goods, wash bottles, dispensing bottles, heavy-duty produce bags, and flexible plastic sheets.
HDPE	Polyethylene film; water, juice, and milk bottles; margarine and butter tubs; retail bags; trash bags; grocery bags; cereal box liners; detergent, shampoo, and laundry detergent bottles; and motor oil bottles.
PC	Polycarbonate medical packaging, sterilizable baby bottles, and reusable water bottles.
PVC	Food and beverage tubing, mints and gum blister packaging, clear food packaging, cling wrap, meat wrappers, cooking oil bottles, plastic squeeze bottles, detergent and window cleaner bottles, and peanut butter jars.
PVDC	Food and medicine packaging.
Polyamide/nylon	Microwave and conventional cooking packaging applications.
PS	Hot beverage cups, meat trays, sCD cases, disposable cutlery (Styrofoam), food containers, take-home boxes, and egg cartons.

Humankind is working hard to shift to a circular economy by introducing scalable recycling. A circular economy calls for judicious allocation of the world's resources, including plastic raw materials, and revalorization of the products in circulation. The circular economy thrives on reuse, reduction, and recycling economies. An illustration of the linear and circular economies is shown in Figure 1 [2].

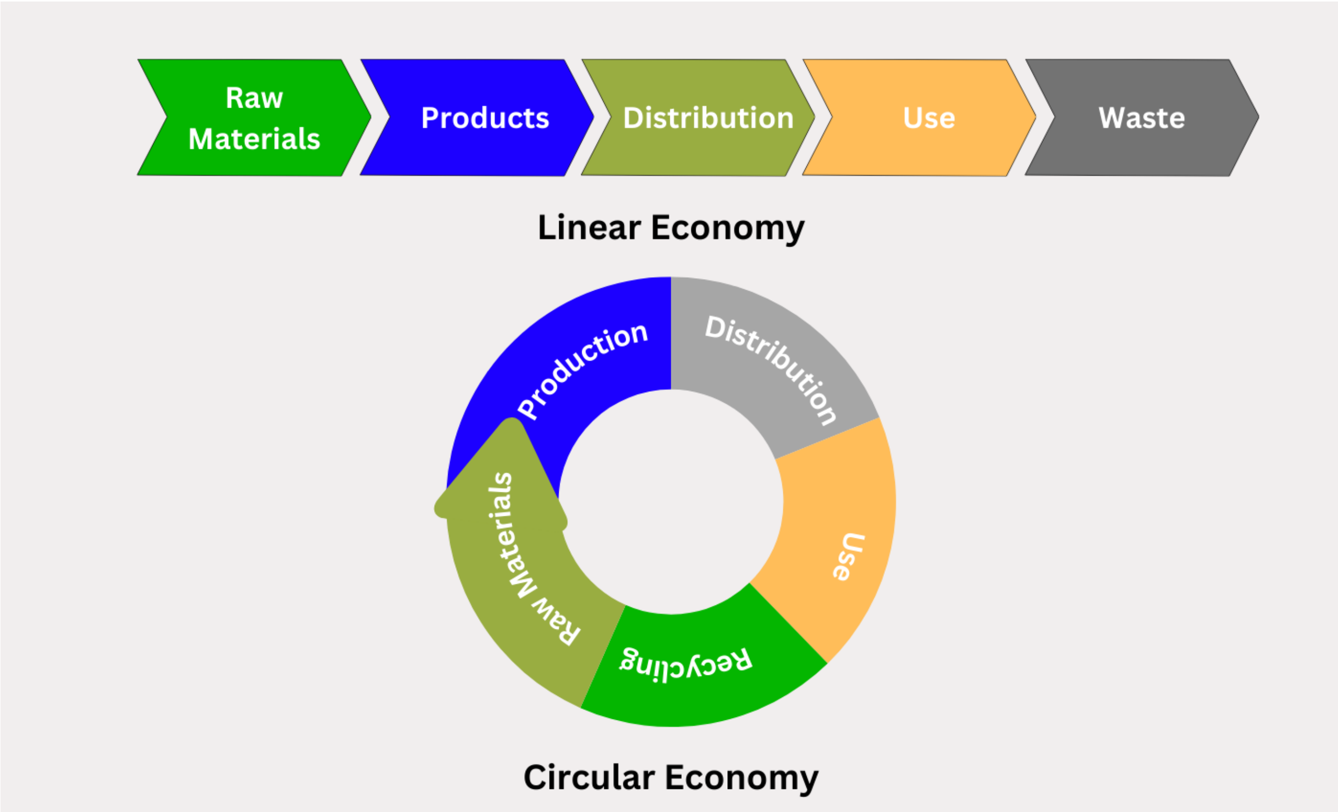

Figure 1. Illustration of the linear economy and circular economy [2].

Some experts view advanced plastic recycling as a failure. In some regions, mechanical recycling accounts for 22% of recycled plastic, chemical (feedstock) recycling for 3%, and energy recovery (combustion) for 60% [6]. This data has been used to create the illustration in Figure 2.

Many advanced technology recycling plants that promise to drastically reduce plastic waste are never commercially viable and generate even more environmentally damaging chemicals [7]. Pyrolysis, the most successful advanced recycling technique (in terms of the quality of regenerated polymers) only recovers 20% of the waste feedstock, burning 70%. It is sometimes seen as a "form of incineration" [8], and it has been declared that toxic emissions and chemicals from the most advanced plastic recycling technologies could cause new environmental and climate issues. Some experts from the Natural Resources Defense Council (NRDC) provide data from the United States Environmental Protection Agency (EPA) to point out that instead of reducing pollutants in the environment, chemical recycling releases a great amount of hazardous chemicals [8]. These pollutants are primary causes of several health conditions.

However, as shown in Figure 3, some industries are closer to circular claims than others. Based on 2018 data, out of the 1.3 million tons of construction plastic waste in Europe, 45% was reused as energy sources, and 25% was mechanically recycled. Figure 3 is a visualization created from the data collated by EuRIC AISBL [2] on the distribution of prevailing recovery techniques by polymer type in the construction sector [2].

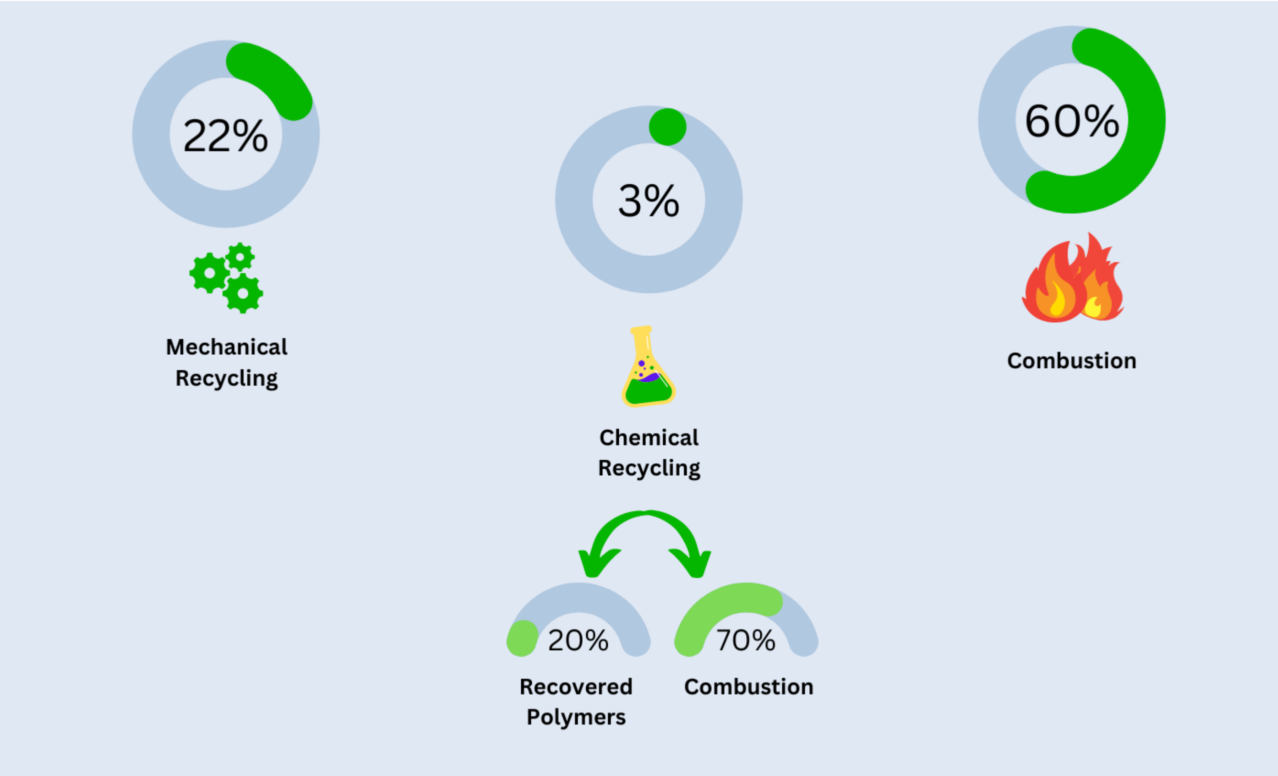

Figure 2. Breakdown of plastic recovery efforts in 2019 [6,8].

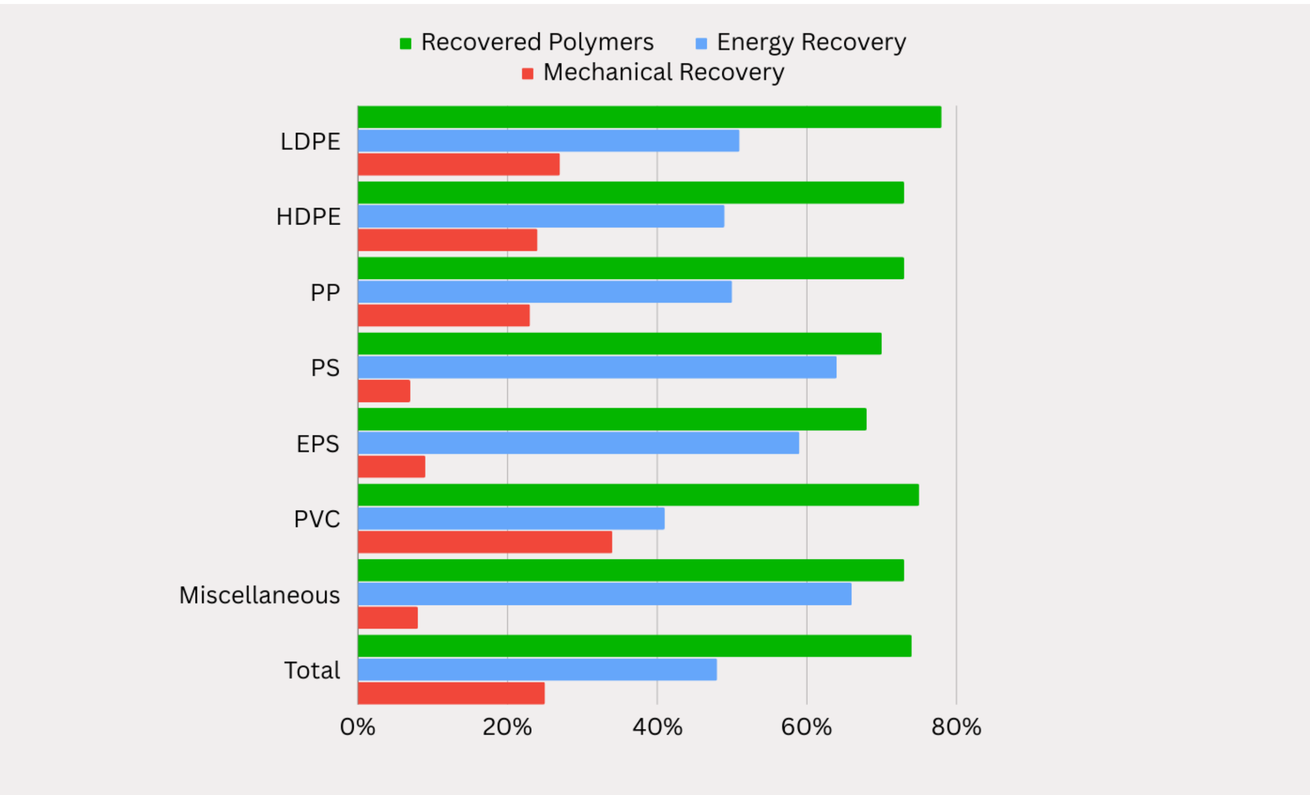

Figure 3. Breakdown of the percentage of material and energy recovered from plastics used in the construction industry (adapted from data collated by EuRIC AISBL [2]).

Structure and Approach

This paper begins by establishing the recyclability of various plastics and recycling challenges. Section 3 introduces reprocessing technologies and focuses on mechanical recycling (arguably the easiest approach). In Section 4, mechanical recycling innovations are investigated. To focus on recent technology, excluding technologies based on assumptions that might have been disproved with time, Section 4 focuses on innovations presented since 2019, highlighting a few from 2018. This provides an overarching exposition of mechanical recycling possibilities. Finally, Section 5 presents conclusions from the study.

2. Recyclability

Despite the urgency of the pollution problem, plastic pollution continues to gain momentum for several reasons. The most popular cause of this rise is increased plastic production. While production has dropped in some regions, there has been a net increase globally [2]. There are more salient reasons. The global plastic recycling campaign is filled with misinformation. Only 9% of plastic waste was recycled in 2019, and most plastic waste is completely unrecyclable despite current technological advancements [7]. Many brands promoting circularity messaging merely transfer their waste challenges to the Global South [9]. There is so much ignorance surrounding plastic recycling. Firstly, many consumers think all plastics are recyclable. Figure 4 shows the material-specific proportion of some reprocessed plastics.

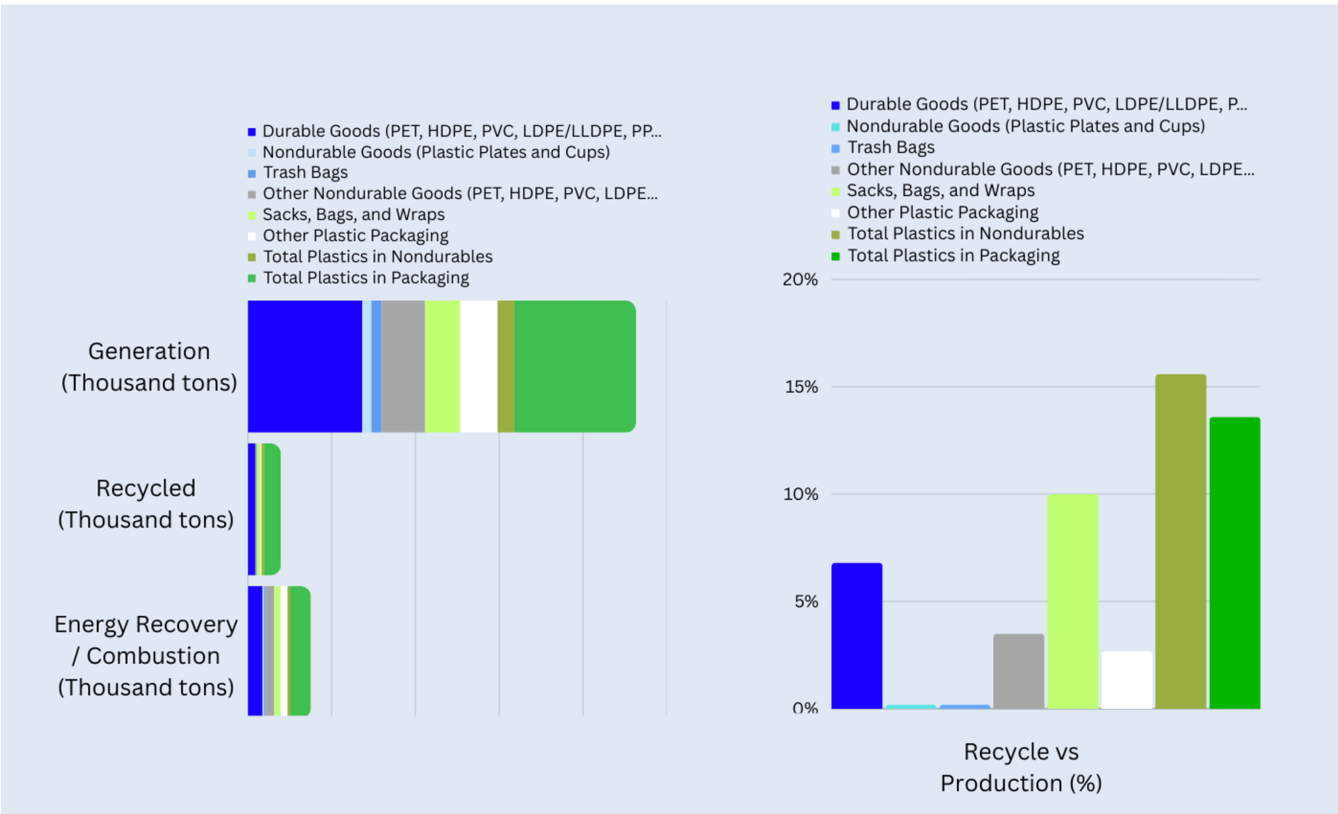

Figure 4. Breakdown of the recycling capacities of different kinds of plastics and plastic products (adapted from data collated by Greenpeace USA [7]).

In Europe, 26–52% of plastic packaging is recycled. Over 50% of European countries recycle more than 40% of their plastic packaging: Czechia, the Netherlands, and Spain recycle almost 50% [2]. In 2018, the US EPA published material-specific recycling statistics. A total of 8.7% of post-consumer plastics were recycled: 26.7% miscellaneous resins, 18.5% PET, 8.9% HDPE, 4.3% LDPE/LLDPE, 0.9% PS, 0.6% PP, 0% PLA, and 0% PVC. A total of 13.6% of plastic packaging and containers were recycled: 0% miscellaneous resins, 25.4% PET, 14.8% HDPE, 9.9% LDPE/LLDPE, 3.6% PS, 2.7% PP, 0% PLA, and 0% PVC [7]. Recycling PET is simple. In all, 52% of PET is recycled in Europe [10]. Coca-Cola, ALPLA, and the National Association for PET Container Resources (NAPCOR) declare that 30% of plastic PET bottles in recycling plants still end up as waste due to contamination and losses. About 1.8 million tons of flakes are recycled from bottles. Only 31% of recycled flakes from bottles are reused in bottle pellets; 69% are channeled into other PET products [11].

HDPE is wholly recyclable. It is recycled via shredding, melting, and pelletization [12]. Due to its soft composition, only 10–15% of HDPE is recycled in Europe. Only a small amount of recycled HDPE is used for food-grade applications. LDPE is flexible and less brittle than other plastics when recycled. It is reused for mechanical applications as it has superior qualities to other recycled plastics. PP is very rigid and has a high melting point. It is generally recycled into pellets. While very recyclable, PP suffers because it is difficult to collect and contains undesirable additives. PS recycling is limited due to the polymer's low density, which makes it unsuitable for conventional recycling. PVC is rigid and strong. When recycled, it is reused as plumbing and construction materials. Nylon recycling is becoming more popular. Due to its desirable mechanical qualities, Polyamide (nylon) is mostly employed in engineering applications. It can be recycled at lower temperatures than most polymers. Acrylonitrile Butadiene Styrene (ABS), the rigid, shiny polymer used in planes and Legos, can be recycled and repurposed through injection molding. Although the process is simple, most ABS waste is not recycled [13,14]. Thermoplastic elastomer (TPE), a popular 3D printing material used in phone cases, can be recycled mechanically or chemically. TPE can be shredded and compression molded. Again, it is a simple process that is not widely practiced [15]. Miscellaneous polymers such as bioplastics are rarely recycled because they are unsuitable for conventional methods.

Plastics are commonly identified with resin identification codes. This nomenclature, obtained from ASTM D7611, has numbers that generally include hashtags or inverted commas. For example, polyethylene terephthalate is identified as PET #1 or '1' (polyethylene terephthalate (PETE)) [4]. Recycled plastics #3–7, polyvinyl chloride PVC #3, low-density polyethylene LDPE #4, polypropylene PP #5, expanded polystyrene EPS #6, polystyrene PS #6, and other multi-material plastics #7, are not favorably received by the industry. Recycling plants, including California Material Recovering Facility (MRF) and the City of Knoxville, Tennessee, complain they are forced to dispose of these kinds of recycled materials because of the lack of end-market demand. PP #5 has high toxicity levels, barring its conversion to food-grade material. Recycled PP#5 cannot compete with newly produced materials because PP#5 manufacturing plants ramp up production and reduce the price since the polymer is a byproduct of gasoline. Also, plastics #3–7 cannot be collected on a large scale to justify investment in sorting initiatives [7]. This brings us to the next point. Some consumers do not know that plastic waste must be separated before it is recycled. By 2025 and 2029, European member states are mandated to increase the collection rate of beverage bottles by 77% and 90% [16]. Reusable plastic bags can be recycled but are challenging to collect. About $4.2 to $5.9 billion of US residents' taxes and contributions go to the collection of recyclable waste from curbside bins. An overview of the accessibility of municipal plastic collection in a US state is displayed in Figure 5.

Furthermore, the more attractive plastic packaging is, the more difficult recycling becomes. Colored plastics are more difficult to recycle than clear plastic. Chemical contaminants and additives make plastics virtually impossible to recycle. According to Canada's National Observer, some toxic materials frequently employed to give plastics desirable properties release endocrine disruptors and cancerous compounds during recycling [17]. According to the Ellen MacArthur Foundation's New Plastic Economy (EMF NPE) initiative, most plastic packaging is not recyclable. To be recyclable, plastic must have a 30% reprocessing rate. Polymers, such as PET #1 and HDPE #2, have rates of 20.9% and 10.3%, while the threshold for many other plastics is less than 5%. Only PET #1 and HDPE #2 can be said to be recyclable [7]. The reprocessing rate is obtained by the ratio of the reprocessing capacity to the waste produced. For example, the US produced approximately 11.5 billion lbs. of PET waste in 2019 (assuming a 4% growth rate), and its recycling plants can handle 2.4 billion lbs. Therefore, the reprocessing rate is 2.4:11.5 or 20.9% [7]. Another popular misconception is the degradability of bioplastics. Unfortunately, bioplastics and bio-based plastics are not necessarily biodegradable. The action of microorganisms can degrade biodegradable and compostable plastics. Some fossil-based plastics are biodegradable, such as PBAT and PCL. Bio-based biodegradable plastics include PLA, PHA, and PBS. Bio-based PE, PET, PTT, PBT, PVC, and PU are not biodegradable [2].

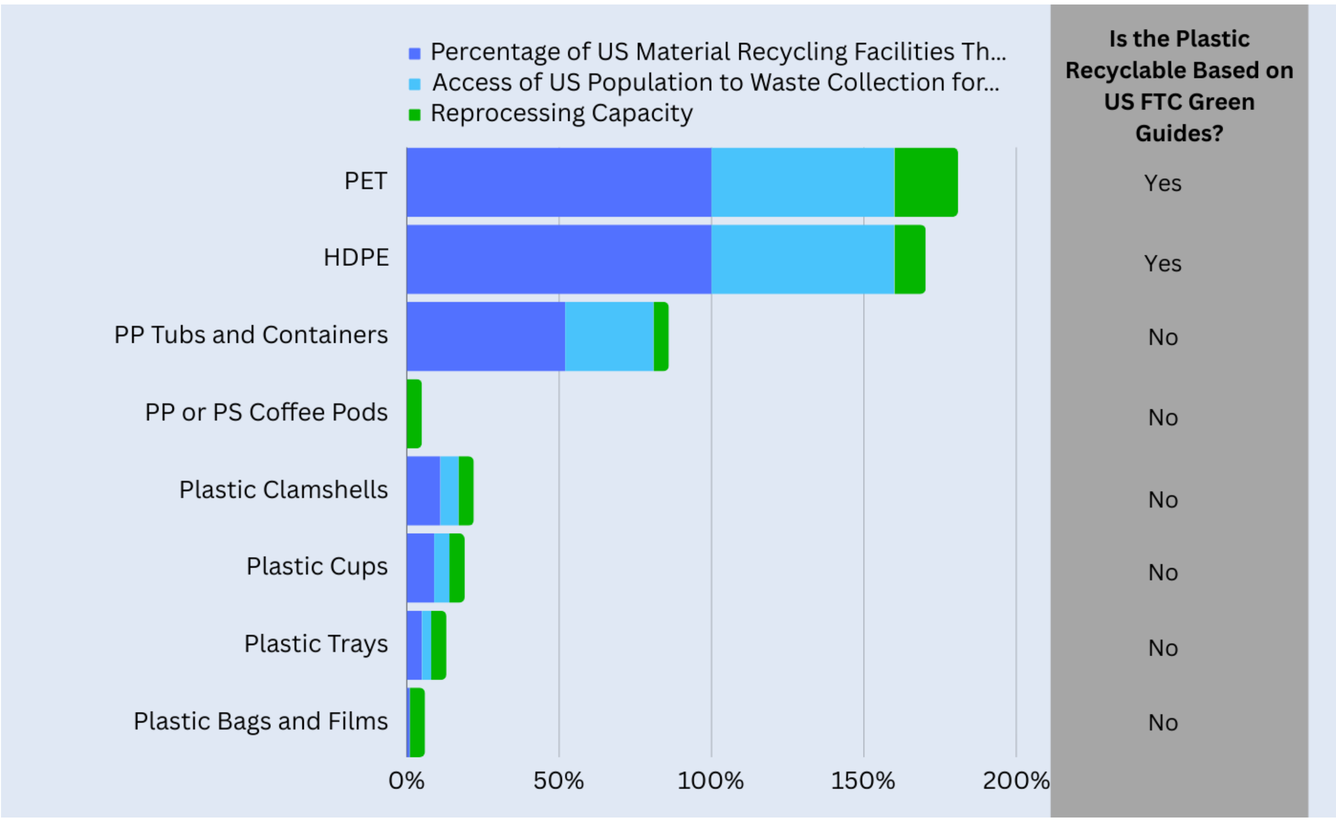

Figure 5. US residents' access to municipal plastic collection schemes (adapted from data collated by Greenpeace USA [7]).

In summary, for conventional plastics the hierarchy of recyclability is PET, HDPE, LDPE/LLDPE, PS, and PP in descending order. Unfortunately, thanks to the PET recycling capacity worldwide and other factors, landfills are dominated by largely unrecyclable plastic. More recycling initiatives should focus on other types of plastics. TPE and ABS are relatively easy to recycle. Although they might not be repurposed for food-grade applications, most of these plastics can be recycled using simple processes to reproduce the same materials [3]. For plastic recycling schemes to be successful, the public must have no misconceptions about recycling. Buttressing the Recycling Partnership's advice that a $17 billion investment of $1.2 billion per year should be channeled toward recycling education and outreach strategies, more educational initiatives have been established to keep the public informed of the nuances in the recycling space [7].

3. Reprocessing Technologies

Due to their distinct constituents, all plastics cannot be recycled using the same methods. Reprocessing technologies are subdivided into primary, secondary, tertiary, and quaternary (i.e., ASTM 1–4). Primary recycling covers the recovery of waste from plastic factories. Secondary includes reprocessing post-consumer plastic (mostly into lower-grade products) [18]. Tertiary recycling is the chemical recovery (via depolymerization) of solids, liquids, and gases contained in used plastics. Quaternary recycling recovers energy (usually via combustion). Mechanical recycling is one of the most common ways to recycle plastics, especially PET [17]. This section will briefly outline these methods and dive deeper into mechanical recycling, which is the focus of this study.

3.1. Overview

There are various types of recycling: biological, thermal, chemical, and mechanical. Biological recycling is used for biodegradable polymers. Biodegradable plastics are increasingly being sought after. These biopolymers are recovered and are degraded via the action of aerobic composting, fungi, and bacteria, and the action of other microorganisms. Biological recycling also permits the recovery of carbon as biomass for energy applications [18]. Conversely, repurposing plastic waste not only solves the waste problem but also reduces the demand for natural resources (such as wood, limestone, and sand) [19].

Chemical recycling includes several processes that depolymerize plastics into monomers or feedstock. This output can be reused to build new polymers or convert plastics to other chemical products [16]. Unfortunately, most chemical recycling processes are still struggling at the pre-commercial stages. Not all polymers are reversibly polymerized. PET, a condensation polymer, is reversibly polymerized. In recent years, innovative chemical and hybrid processes have been developed to improve the quality and quantity of recycling throughput, improve the sustainability and scalability of the process, and reduce cost [17]. Chemical recycling includes solvent-based and chemical depolymerization (methanolysis, glycolysis, hydrolysis, and pyrolysis) [7,16] and thermal depolymerization (pyrolysis and gasification) [18].

- In glycolysis, a polymer is doped with glycol under controlled conditions. In the case of PET, mono-ethylene glycol (MEG) can be added for the polymer to generate bis 2-hydroxyethyl terephthalate (BHET) [16].
- Hydrolysis is achieved by mixing a polymer with an alkaline, acidic, or neutral solution under specific conditions. In the case of PET, this produces PTA/MEG [17].
- Methanolysis is achieved by mixing a polymer with methanol under controlled conditions of 270–300 °C and 0.1–15 MPa [18]. This generates ethylene glycol monomers (MEG) and dimethyl terephthalate (DMT) [16].
- Alcoholysis is depolymerization in alcohol. PU is depolymerized this way. The process produces carbamates and polyhydroxyl alcohols but no carbon dioxide [18].
- Pyrolysis is gaining attention as an optimal approach for recovering chemical feedstock from end-of-life plastics [6]. Several advantages of pyrolysis make it a desirable approach. Pyrolysis encourages the adhesion of different chemical bonds in inert gaseous conditions, producing smaller molecules. Pyrolysis-based recycling technologies are suitable for "hard-to-recycle" waste, including PET and PUs. Nevertheless, some plastics cause corrosion and produce undesirable compounds when subjected to pyrolysis [20]. Polyvinyl chloride- and bromine-coated plastics corrode the recycling vessel and generate halogenated compounds [6]. Incinerating PVC releases a heavy-duty pollutant, dioxin [21]. Unfortunately, a more significant percentage of plastic waste fed into the pyrolysis process is used as high-temperature fuel (burned), and only small quantities of waste are recycled. Brightmark Energy rated that in its Ashely pyrolysis plant, 70% of the waste feedstock would be burned, 10% would become waste char, and only 20% would be recovered as unrefined pyrolysis oil [7].
- Gasification converts feedstock to syngas. Polymers are oxidized and treated with carbon dioxide, methane, and other light hydrocarbons like carbon dioxide, water, and methane under high temperatures. In some gasification processes, polymers are thermo-cracked in a liquefaction step into gas fractions with varying solubilities. Non-condensable gases are recovered as fuel.
- Hydrocracking converts long-chain hydrocarbons to small molecules like kerosene and gasoline. Polymers are mixed with hydrogen under high pressure in the presence of catalysts [22].

3.2. Mechanical Recycling

Mechanical recycling is a prevailing technique. It is only suitable for specific polymers (thermoplastics). Split fibers during the shredding process damage some mechanical properties of the output [17]. The process starts with collection and sorting. After waste is collected, specific plastics are separated, leaving behind hazardous materials and debris. The collected pieces are ground, and resulting flakes are further separated based on color, resins, and other physical properties. Next, the chips are washed and sorted if desired. Finally, the flakes are melted and homogenized into pellets. This process is illustrated in Figure 6 [2].

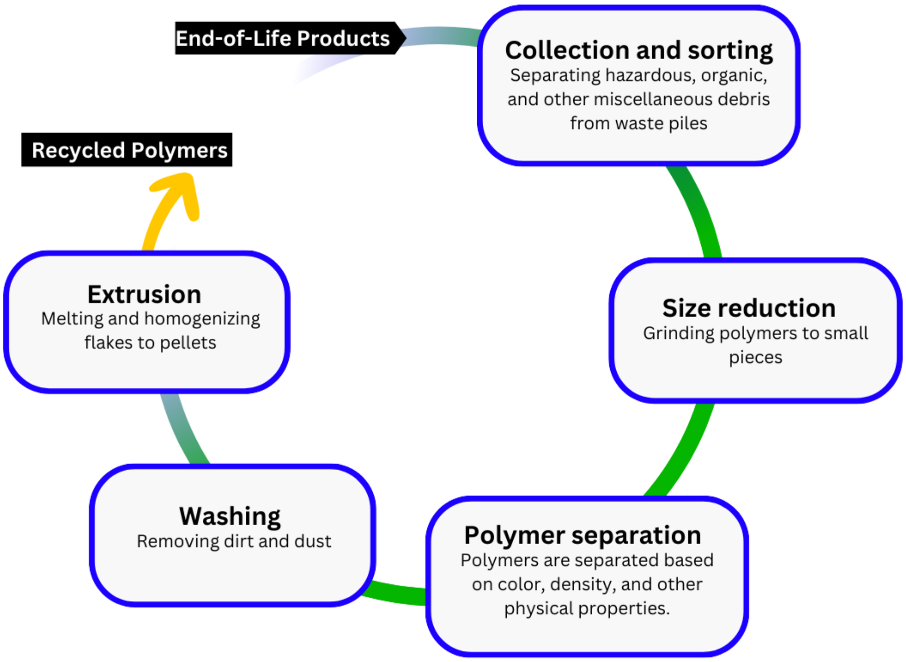

Figure 6. An illustration of the mechanical recycling process adapted from [2].

3.3. Mechanical Recycling Challenges

Mechanical recycling is a resource-consuming process: a lot of energy is expended in processing, sorting is labor intensive, and cleaning predominantly releases wastes requiring further treatment [18]. The following processes must be improved to address the failure of mechanical recycling:

- Reduce and eliminate toxic materials and additives in plastic production [23]. For example, the Coca-Cola Company replaced its signature Sprite green PET bottle with clear plastic as colorants and additives reduce the recyclability of plastic. Japanese beverage firms voluntarily switched to clear PET in 1992, and South Korea banned colored PET in 2020 [7].
- Make plastic easier to collect. The US uses drop-off facilities and curbside pickup services, and European member countries run door-to-door collection and deposit return schemes. Some collection schemes are more effective than others [16].
- Improve sorting technologies, reduce sorting time, and make the process less labor-intensive. Research and development (R&D) efforts are increasingly channeled toward boosting the accuracy and speed of automated sorting systems. Better artificial intelligence (AI) algorithms are developed to resemble and enhance the decisions of manual sorters.
- Discover more environmentally friendly ways to reprocess waste. Recycling processes are very harmful to plant operators. Also, washing mechanically recycled waste releases microplastics into the environment [7]. Recycling plant workers and communities are exposed to toxins [24,25]. Residents around recycling plants have reported plastic films and dust blankets over their properties. Plastics' high inflammability also

makes recycling plants fire hazards. In August 2022, people living close to an MRF in Dallas, Texas sued Poly America for the health impacts of the recycling fire that burned for 23 h [7]. The Last Beach Cleanup team runs a project that reports plastic recycling fires globally [26]. these data are presented in Figure 7.

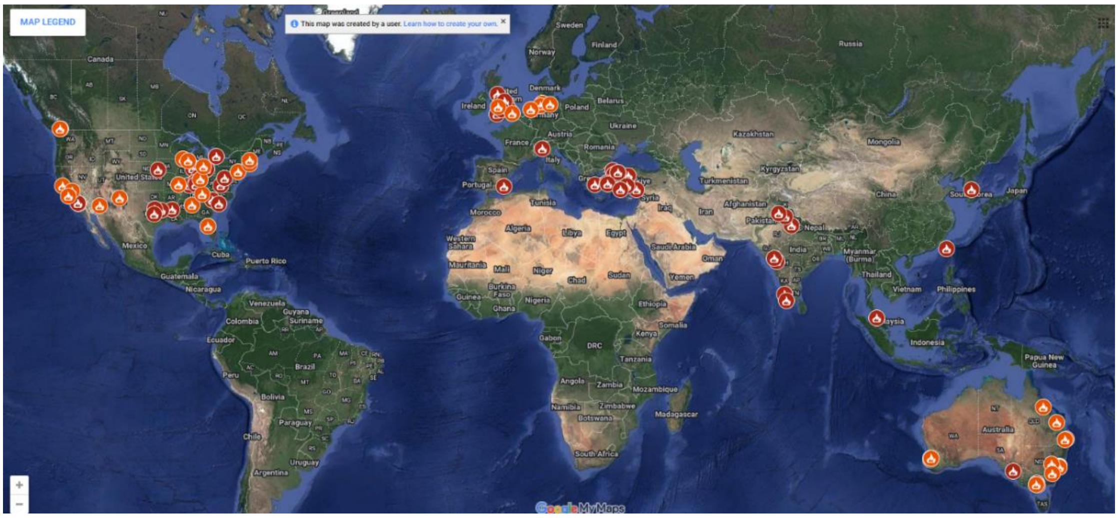

Figure 7. Map showing the location of fires caused by recycling efforts [26].

4. Mechanical Recycling Innovations

The aforementioned challenges in mechanical recycling call for urgent solutions. This section examines innovations in the mechanical recycling space. It addresses technological limitations and identifies solutions that make mechanical recycling a sustainable and viable option. Several patented and open-source technologies have been developed to improve mechanical plastic recycling. Plastic recycling technologies are being churned out by the thousands every year. The number of plastic recycling and treatment patents granted annually continues to increase. In fact, researchers are competing to invent scalable commercial technologies that will generate clear recycled plastic. The race to produce cost-effective recycling technologies to meet the ever-growing demand for low-carbon products fuels this interest. Chinese companies have the highest number of patent filings in this space due to the strict single-use plastic laws in the country. The second highest number of patents are filed by Indian researchers. Researchers have developed improved devices covering every stage of the recycling process: grinding, granulation, extrusion, additive manufacturing, and so on [27]. Some recycling initiatives, like Trashy Bags, use fairly simple methods [28]. Trashy Bags makes affordable consumer goods out of sewn plastic waste. The company gathers 180,000 plastic sachets or 275.143 kg of waste every month and washes and sun dries them. They are straightened and sewn into sheets. These sheets are used to make bags, stationary purses, caps, and so on [28]. Other solutions are more complicated.

4.1. Additive Manufacturing

One attractive recycling destination is 3D printing. However, virgin materials are predominantly used for products made from Selective Laser Sintering (SLS) or Fused Filament Fabrication (FFF) processes [29]. Recovered plastic products made from additive manufacturing usually require moderate- to high-quality properties. Thus, several additives and heat treatments are applied to improve the recyclate. Studies have addressed all aspects of the additive manufacturing process. Some focus on optimizing 3D printing filaments, and others on capturing processing commercially available recycled filaments. Producing filament begins with sorting, cleaning, and extruding. The pellets produced are melted, cooled, and extruded into filaments using a die [29]. Companies like Ultrafuse, Verbatim, and Madesolid sell PET filaments [30]. Distributed Recycling for Additive Manufacturing

(DRAM) is a consumer-friendly process for producing consumer goods at affordable prices. Initially, DRAM technologies focused on recyclebots that extruded 3D-printing filaments from waste [30].

Guinaldo [29] patented a way to recycle vacuum bags to make 3D-printing filaments. The bags were cut into thin films and rolled. They were mixed with virgin fibers extruded and pelletized in the presence of an anti-hydrolysis additive. The pellets were melted, cooled, and processed through a die into filaments. This invention provided a way to recycle vacuum bags, which has been little explored [29]. Mixing plastics with fiber, wood, and carbon-reinforced polymers is beneficial in 3D-printing applications [31,32]. Lupisan et al. [33] produced monofilaments for FFF and manufacturing processes. Contaminated waste was sorted and ground to uniform flakes, washed in a closed-loop cycle, and dried. The flakes were extruded and melt-filtered if desired. Another patent describes a way to produce a 3D-printing material made of regenerated and recovered plastic, grapheme, biochar, foaming agent, lubricant, toughening agent, chain extender, compatibilizer, and fiber. It produced a PP composite with improved properties [34]. Little et al. [30] proposed a mechanism for using post-consumer PET flakes for DRAM feedstock without converting the rPET to filaments. The process uses Fused Particle Fabrication (FPF) and Fused Granular Fabrication (FGF). Industrial PET flakes have also been recycled using FPF and FGF processes [35]. For additive manufacturing applications, plastic wastes are generally heated and converted to pellets or filaments [29]. These recycling innovations significantly address recapturing plastics that elude regular reprocessing facilities [29].

Unfortunately, each reprocessing cycle depletes the plastic's properties. After five cycles, the waste must be supported with virgin feedstock. PET also struggles with uncontrollable water absorption and crystallization [30]. ABS and PLA are the most used filaments. HDPE, PP, PU, LDPE, PC, LLDPE, PS, and other popular plastics were difficult to convert to filaments using this process [36–38]. There is a need for better ways to granulate PET for 3D-printing feedstock [30]. The nozzle height, tube size, and fan specifications of 3D printers need improvement. Some plastics cool while in the nozzle and PET has wrapage and shrinkage issues, which has led many commercial PET filament companies to stop production. rPET's wrappage and shrinkage issues limit its use in additive manufacturing. The common steps involved in reprocessing plastics for additive manufacturing applications is shown in Figure 8.

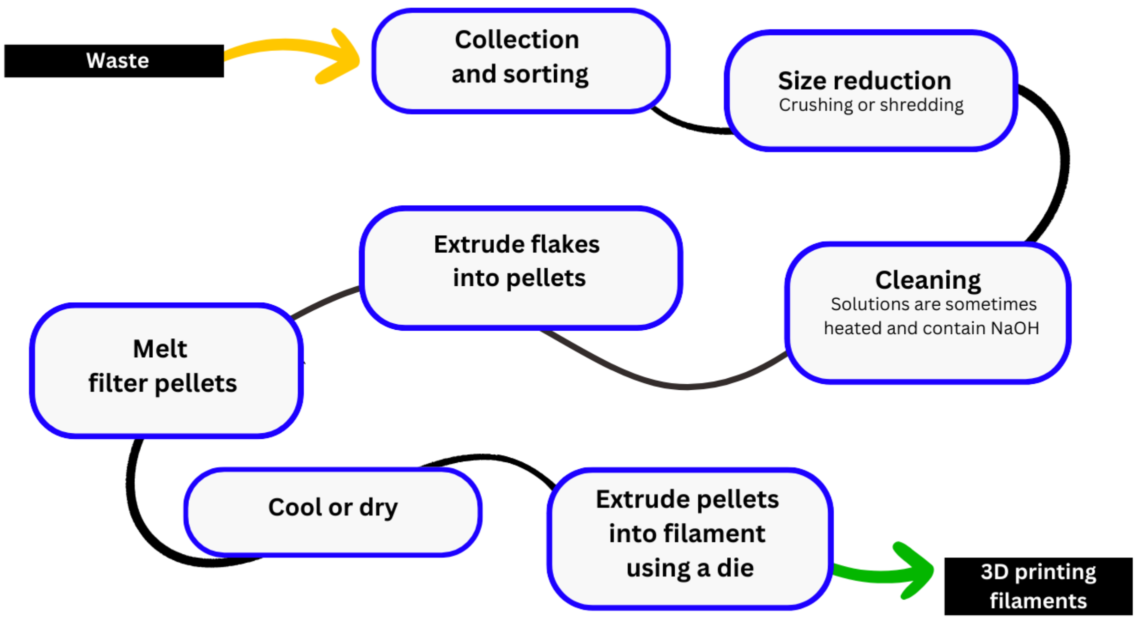

Figure 8. An illustration of the common steps taken in reprocessing plastics for additive manufacturing applications adapted from [29,30,35].

4.2. Bricks, Blocks, and Similar Construction Materials

The construction industry consumes a lot of waste plastic because (i) it is one of the largest industries across the globe, (ii) plastics are very durable, and (iii) many construction applications are not specific about virgin materials [19]. Since a majority of recycled plastics cannot be reused for food-grade applications, they make suitable aggregates in construction materials. Concrete production from waste plastic has enjoyed a lot of attention. Some studies and products have used recycled PET, PU [39], HDPE [40,41], PC [42], PS [42], nylon [43], LDPE [44], high-impact polystyrene (HIPS) [45], PVC [46], polyolefin [47], and butadiene [48] as binders and aggregates in brick, blocks, and similar construction materials. Plastic waste shows improved qualities when mixed with glass [49], ceramics [44], metals [44], and asphalt [50].

Due to the low adhesion of plastic to cement, heat is often used to mix plastic waste with cement. Yang and Gong [51] developed a concrete production process involving waste concrete and PET plastics. The concrete and plastic were crushed, screened, and heated in a heating tank. The soil particles were separated from liquid plastic using a metal screen. Concrete, plastic, and fine aggregates were mixed with Portland cement, a water-reducing substance, an air-entraining compound, and a silane coupler and stirred, adding water at intervals. The result was a durable impermeable concrete [51]. Another patented process uses waste polyethylene granules or polyolefin [47]. The waste polyolefin was melted, extruded, and crushed into cylindrically shaped pieces of 2 mm and 2–5 mm diameter. Then, 10 wt.% waste pieces were mixed with 12–25 wt.% Portland cement, 5–40 wt.% rough aggregate, 37–57 wt.% smooth aggregate, 5–12 wt.% water, and 0.1–2.0 wt.% superplasticizer. This created lightweight concrete with low thermal conductivity and high compressive strength [47].

Neplast makes pavement blocks out of plastic waste. The blocks are not very selective; they consume sorted plastic waste, except PVC [28]. The company collects plastic waste, sorts it, and shreds it into small threads. The blocks made of 70% waste and 30% sand leverage the polymers' inherent resilience and binding elasticity. Pore volume and grain size varied with variation in sand. These blocks are more robust than their cement counterparts [28]. Neplast plastic blocks support a maximum load and compress of 36.77 kN and 460 kN whereas concrete pavers support 19.64 kN and 340 kN. Aneke and Shabangu [52] mixed 80–60% of PET and 20–40% of foundry sand to make bricks. The strength of the bricks with different proportions of plastic was 85% greater than their clay counterparts. The bricks with a 70:30 (plastic waste to sand) ratio showed maximum compressive and tensile strengths of 38.14 MPa and 9.51 MPa [52]. A ratio of 60–40 had the next best tensile strength, and 80:20 was the next best compressive strength. Another study mixed recycled crushed glass and PET in ratios of 80:20, 70:30, and 60:40, boosting compressive and tensile strengths by 54.85% and 70.15% higher than clay counterparts. The average compressive strength and tensile strength obtained were 42.01 MPa and 9.89 MPa, respectively, and the average water absorption value was only 2.7%. Because of their high hydrophobic properties, both types of masonry bricks made from foundry sand and crushed glass do not need water for construction, are more resistant to chemical attack, and are less deformable under strain stress as compared to burnt clay bricks [49]. Shredded PET has also spiked the compressive strength of compacted earth blocks by 244% [53]. Some studies have taken another route, simply filling PET bottles with sand, recycled aggregates [54], and cement mixtures [42]. However, the abrasive resistance of plastic pavers is significantly less than clay and concrete paving blocks [55].

Waste plastics have also found applications in other construction materials like asphalt and composite rubber used in various building materials. Coe [48] produced a composite construction material from waste rubber and an auxiliary polymer. The auxiliary polymers were butadiene and functionalized styrene-butadiene. Vulcanized waste tires were mixed with organometallic compounds to make an aqueous slurry, which was delaminated in an electromechanical reactor. This invention provided a better way to break down waste tires into their original constituents. Li [50] presented a method for producing enhanced asphalt from waste tires. The waste tires were crushed and mixed with white carbon black, SBS asphalt modifier, pentaerythritol phosphate, zinc oxide, and a solubilizing agent and then powdered. Then, 50–60 wt.% of matrix asphalt was mixed with the powder, heated to 170–180 °C, stirred at 300–500 r/min, swelled into a liquid state, cooled, and inoculated. The process improved the elasticity, tensile strength, stability, aging resistance, and cohesive qualities of the asphalt blend. Styrene-butadiene-styrene (SBS) and other asphalt additives with these properties are expensive. Similar inventions using rubber powder-produced blends have poor compatibility, as asphalt does not readily mix with rubber [50].

Overall, plastic bricks and blocks have low water absorption and maintain their durability when soaked [49,52,55]. Introducing plastics into bricks, blocks, and similar construction materials has increased sound blocking [43], thermal insulation [39,42], and impermeability [47,50]. Lightweight concrete translates to smaller-sized structural columns, walls, beams, and foundations and therefore smaller seismic loads [47]. Using plastic waste in concrete mixtures reduces the compressive, tensile, and flexural strength of concrete due to concrete's poor adhesion to plastic [56]. Moderate to low additions of average plastics (i.e., plastics unadulterated with additives) into construction materials increase strengths up to a point. Beyond this, higher proportions of plastics in structural mixtures deteriorate strength, create huge voids, weaken adhesion, and lower workability [56]. Treated recycled plastics show better results. Table 2 presents an overview of these developments.

Table 2. The application of mechanically recycled plastic in construction materials.

Study	Plastic Waste	Reinforcements	Additives	Percentage of Plastic Waste by Weight	Product	Product Qualities
[28]	Mixed plastics including PVC	Sand	-	70%	Bricks	High compressive strength
[39]	PET and PU	-	Binder	20–80%	Bricks	Thermal insulation and good strength for non-load-bearing bricks
[40]	HDPE and PET	Clay	Binder	1–20%	Bricks	High thermal conductivity, high compressive strength, and low water absorption
[42]	PC, PS, and mixed thermoplastics	Portland cement and sand	Fly ash	0–10%	Bricks	Thermal insulation and high compressive strength
[45]	HIPS and LDPE	Cement, coarse aggregate, and sand	Binder	0–50%	Concrete	Lightweight durable concrete
[46]	HDPE, PP, and PVC	Natural aggregates and cement	Binder	25–75%	Concrete	Sound blocking, heat insulation, and high strength
[47]	Polyolefin and PE	Portland cement, rough aggregate, and smooth aggregate	Super-plasticizer	10%	Concrete	Lightweight concrete with low thermal conductivity and high compressive strength
[48]	Butadiene and functionalized styrene-butadiene	Asphalt	SBS asphalt modifier, solubilizing agent, and white carbon black	-	Construction material	Improved elasticity, tensile strength, stability, aging resistance, and cohesive qualities
[49]	PET	Glass	-	20–40%	Bricks	High compressive and tensile strength, low water absorption, and resistance to chemical damage
[51]	PET	Fine aggregates, concrete, and Portland cement	Silane coupler and air-entraining compound	-	Concrete	High durability and impermeability
[52]	PET	Foundry sand	Binder	30%	Bricks	High compressive and tensile strength and low water absorption
[53]	PET	Clay	-	1%	Compressed earth bricks	High compressive strength

4.3. Composite Boards

Manufacturing particle board is another good use for recycled plastics that are unsuitable for food-grade applications. These boards include wood-based [57], stone-based [58], metal-based, and copolymer boards. The fabrication of composite board usually involves sorting and shredding the plastic waste, crushing wood or another base material, and mixing the dry materials with couplers under heated conditions. Afterwards, the agglomerate passes through the extrusion machine [59]. Wood–polymer composites (WPC) predominantly consist of a mixture of natural fibers or flours and polymer(s). They have been made from PP [60], PE [59], HDPE, PET, and LDPE [61]. They are generally used in decking, furniture, cladding, fencing, and paneling [59]. Compatibilizing or coupling agents are used to improve interfacial bonding between polymers and incompatible materials or polymers [59]. These improve interfacial adhesion and evenly distribute dispersed phases. Wood flour and fibers are incompatible with plastics due to wood's hydrophilicity and the latter's hydrophobicity. Additives like couplers and compatibilizers also enhance the tensile and flexural strengths, impact resistance, density, water resistance, and stiffness of wood polymer composites [59]. These properties usually vary with the amount of time the composites are immersed in additive solutions. Maleic grafted polyolefins are common couplers for wood composites [59,60].

Tensile, impact, and flexural strength increase with up to 7% of specific couplers. A total of 7% of elastomer-related compatibilizers increase several desirable plastic qualities by more than 25% [59]. Lim [57] patented a process for recycling bio-artificial wood fiber powder, plastic powder, a dispersing substance, a crosslinking agent, a foaming agent, a reinforcing agent, and a processing aid. The waste powders were mixed and kneaded into pellets. This mixture was dried, melted, mixed with the additives mentioned, and extrusion-molded. This mold was supported by a cover material made of PVC resin, a UV blocker, and a stabilizer (used to form the wood patterns). Unlike conventional artificial wood products, this plastic-reinforced wood was suitable for complex construction demanding light materials [57]. Sahajwalla et al. [62] presented a technique for making a recycled board in which 50 wt.% plastic waste was shredded, mixed with less than 50 wt.% binder, molded, and hardened to form a layered composite board [62]. The product had a high melting point, water resistance, thermoplasticity, structural strength, ductility, and hardness.

Wicaksono et al. [61] used a combined recycled plastic and reclaimed wood waste using a melt–blending process. Mixtures with higher plastic concentration show lower water permeability, higher thermal stability, and better mechanical strength. Epoxy resin boards have also been made from waste plastic and recycled circuit boards. Song [58] prepared these boards from waste prepreg, stone, metal, wood, plastic, and printed circuit board powder. After cleaning the raw materials, they were crushed, ground, and mixed. After being converted to slabs in a paving machine, the mixture was sent to a press-forming machine, placed in an extruder, or heated in an internal mixer and press-formed. The adhesive comprised a toner, an antioxidant, a lubricant, an ultraviolet ray inhibitor, a carbide stabilizer, a reinforcing agent, and a coupling agent [58]. The waste prepregs reduced the need for adhesives due to high bonding strength.

Overall, using recovered plastics in composite boards is another good way of reclaiming mixed waste, as these boards are made of particles of different kinds of materials [59]. However, some composites like LDPE-based wood composites have better physical and mechanical qualities than others like PP-based composites [61]. Also, mixed polyolefins have lower properties than recycled PP- and PE-based composite boards [60]. Polymer composite boards have shown high melting points, water resistance, thermoplasticity, structural strength, ductility, and hardness, leveraging the environmental resistance and durability of plastics. Plastics also make wood composites easier to be molded into various shapes [60]. These boards usually require couplers to not only lessen the adhesives needed but also enhance the properties of the end products. The properties of wood polymer composites improve with up to 7% of compatibilizers. Beyond this threshold, the properties deteriorate [59]. Another challenge with recycled plastic composites is that they are frequently made of either recycled plastic or recycled base material (wood) but not both. Fully recycled plastic composite boards are more sustainable and should be encouraged. Table 3 presents an overview of these developments.

Table 3. The application of mechanically recycled plastic in composite boards.

Study	Plastic Waste	Reinforcements	Additives	Percentage of Plastic Waste by Weight	Product	Product Qualities
[57]	Bioplastics	Bio-artificial wood fiber powder	Stabilizer, UV blocker, crosslinking agent, foaming agent, and PVC resin	30–40%	Epoxy resin boards	High density construction material
[58]	PVC, PE, HDPE, and Printed circuit board	Waste prepreg, stone, metal, and wood	Toner, antioxidant, lubricant, UV blocker, carbide stabilizer, and coupling agent	<70%	Epoxy resin sheet	Improved hardness, low shrinkage rate, good water absorption, and high strength
[59]	PP, PET, and PE	Wood	Compatibilizer	-	Wood plastic composite (WPC)	Good impact strength, good mechanical strength, and moisture resistance
[60]	PP, PE, and mixed-polyolefins	Wood flour	Compatibilizer, UV stabilizers, and anti-oxidants	30 wt.% wood flour	Wood plastic composite (WPC)	High melting point, water resistance, thermoplasticity, structural strength, ductility, and hardness.
[61]	LDPE and PP	Sawdust and organic waste	-	30 wt.% wood flour	Wood plastic composite (WPC)	Low water permeability, high thermal stability, and good mechanical strength
[62]	Thermoplastic polymers (such as ABS, PP, HDPE, etc.)	Glass, metal, carbon, ceramics, graphite, etc.	Binder	50%	Composite board	High melting point, water resistance, thermoplasticity, structural strength, ductility, and hardness

4.4. Mixed Plastic

Sorting is among the biggest challenges of recycling. Processing mixed plastic waste is very difficult because many polymers are chemically incompatible and exhibit different mechanical properties [63]. Breaking down two-phase polymer blends is facilitated by an additive that reduces the interfacial tension and improves adhesion [64]. This additive can be a copolymer partly compatible with both compounds or a functionalized polymer that generates such a copolymer. The copolymer becomes a compatibilizer between the components [64]. Processing mixed polymers creates a fragile end product with very poor mechanical and aesthetic qualities. For example, heating polymers with different melting points can cause thermomechanical damage to components with lower melting points.

In one study, recycling PE and PET from plastic bags and bottles was improved with the addition of a compatibilizer. Flakes of the mixed waste in a 3:1 ratio (PET:PE) were mixed with a dried compatibilizer. The mixture was processed in a piston injection molding system for melting homogenization and injection [65]. The recyclates with compatibilizers had tensile strengths five times better than those without. However, using compatibilizers beyond 3 wt.% deteriorated the end product. Ostvik [66] produced mixed-waste powder comprising two of the following: polyester, PET, PE, PVC, PS, PA, ABS, PP, PC, PLA, PMMA, PEI, polyurethanes, melamine formaldehyde, maleimide/bismaleimide, PEEK, polyimide, PTFE, and epoxy. A mixture of solid waste with organic materials was heated, melted, cooled, and broken down into small particles. The particles were combined with a polymer to form a composite [66]. Richter [67] developed a tertiary recycling method for mixed plastic waste comprising PET, HDPE, PVC, LDPE, PP, PS, PC, PMMA, PA, PEI, POM, and ABS. The unwashed, unsorted, and unidentified waste of varying polymer weights and lengths was ground and dried to a material of quasi-uniform dimensions of 0.03–0.4 inches. A carrier containing fiberglass, aromatic bromine, sodium bicarbonate, tri antimony oxide, sodium chloride, and alluvium was also prepared and dried. The mixed waste was melt-blended at 150–800 °F, mixed with the carrier, dried to remove all moisture, and extruded to create a homogeneous and stable feedstock material. These processes reduce the need for sorting, which is one of the biggest challenges of recycling technologies. Many solutions to the sorting problem depend on relatively unscalable solutions, downgrade the feedstock, or leave existing plastic waste (by focusing on creating new materials like biodegradable polymers) [67]. More mixed plastic recycling technologies are needed. With technological advancements, someday, mixed plastic waste will be convertible to top-quality polymers.

4.5. Multilayered Recyclates

Multilayer plastics consist of an average of six layers of different materials [68]. They are commonly made with coextrusion and lamination processes. Coextruded recyclates are melted together, bonding each layer. Alternatively, a third polymer or adhesive, such as anhydride-modified polyethylene (PE), can be used. Multilayer polymers are recycled via compatibilization and delamination [68]. These processes require either the processing of all layers or their separation. After separation, selective dissolution or delamination can be used. Unfortunately, no commercially viable delamination treatment for industrial-scale recycling has been proposed as of 2020 [69]. Saperatec GmbH is developing a low-energy recycling process for mixed plastics (including PP/Aluminum, PE/Aluminum, and PE/PET). Another company is working on a PVC separation process that swells multilayer polymers in a heated solvent [70] to delaminate them.

Making multilayer recyclates is a way of using higher proportions of recycled materials to produce high-quality products [71]. Baldwin et al. [72] produced a multilayer polymer material consisting of polyethylene/polypropylene (foam) and a polyolefin layer. The polyolefin was shredded, agglomerated, and granulated to produce a crosslinked foam. After pulverizing the foam, 2.5–25 wt.% of this foam was combined with 75–97.5 wt.% polypropylene/polyethylene and a chemical foaming substance. The mixture was coextruded forming a three-layered material with the first and third layers free from polyolefin. This new material passed through ionizing radiation (under a 200–1500 kV electron beam) up to four times and was sent through another foaming process of heating the irradiated coextrusion with a salt [72]. Another patented technology produces a multilayer plastic bag with improved tensile strength from a waste synthetic resin [73]. The outer and inner layers were made of a mixture of biodegradable film and HDPE, LDPE, or LLDPE. The layers were extruded and blow-molded. Liao et al. [74] addressed the problem of low recycled PET material in reprocessing feedstock. Mechanically and chemically reprocessed waste polyesters were mixed, granulated, and melted. They were supported by an electrostatic adhesive containing a metal salt. The mixture was coextruded into a laminated polyester film with up to two recycled polyester film layers.

Other multilayer technologies have produced heat-sealable film, comprising a recycled layer and heat-sealable layer, and other layered films from a plurality of recycled materials and chemically enhanced materials [75–79]. Many processes involve producing polyester film from physically and chemically recycled polyester pellets. Then the chemically recycled pellets undergo an electrostatic process in the presence of a metal salt. The plurality of the physically and chemically recycled mixture was melt-extruded into a film. These technologies provide ways to recycle polymers (such as crosslinked polyolefin) that are not readily recyclable via other commercial processes [77–80]. Recovering these polymers using conventional processes depletes mechanical and surface properties making them unsuitable for many manufacturing processes [72].

4.6. Pellet Production

Preparing recycled high-quality plastics usually requires virgin feedstock and limited quantities of recycled polymers. This relies on the virgin plastic production market and questions the sustainability of the processes. Many inventors have developed ways of using more recycled feedstock and less virgin material [81]. Kleczek and Bayley [81] developed a process to recover polyethylene (such as LLDPE, MDPE, LDPE, HDPE, and VLDPE) from 20–80 wt.% recycled polyethylene and 80–20% polyethylene. Polyethylene was cleaned, melted, and pelletized. The recyclate was a 3–7-layer stretched or shrink film produced using a cast- or blown-film process. Liao et al. [82] presented an injection molding recycling composition for polyester pellets in which 50–80 wt.% virgin polyester was mixed with 20–50 wt.% modified recycled polyester pellets comprising 65–72 wt.% terephthalic acid, 28–31 wt.% ethylene glycol, and 0.1–5 wt.% isophthalic acid. The recycled polyester was obtained by crushing sorted PET waste. The waste was refined via flotation, melted, and pelletized with a single- or twin-screw extruder. The recycled polyester can also be obtained by chemically recycling PET waste using a depolymerizing agent. The patented invention reduces the proportion of virgin polyester pellets used in producing recycled PET products. The process also improves the crystallinity and fluidity of the recyclate [82]. In one process, PET was soaked, washed, chopped, heated to 90–100 °C, and cooked for 20–40 min [76]. The result was cooled into fragments, placed in a tetrahydrofuran-filled stirrer, and stirred for 60–120 min under 70–90 °C. The plastic fragments were separated from the tetrahydrofuran, washed, soaked in sodium hydroxide for 2–4 h, and washed in hydrochloric acid for 1–3 h. The chips were further washed with clear water and dried at 80–100 °C. These chips were melted at 160–200 °C and mixed with a modified elastomer, keeping the mixture at 180–210 °C. The mixture was combined with fillers heated to 150–180 °C; stirred for 20–40 min; mixed with a coupler, antioxidant, and lubricant; and melt-extruded into a composite polymer. This long process created a polymer with good mechanical properties made from a high percentage of recycled plastic [76].

Another patented technology describes a plastic grain-making device comprising a stirring, melting, cutting, and blanking chamber [83]. The device's mode of operation and materials reduce time spent granulating plastic waste and improve the quality of the output. Another process recovered plastic granules from fabrics [84]. Fabrics (including wool, viscose, silk, cotton, denim, rayon, linen, elastane, and polyester) were collected, pulverized into powder, pelletized with a binder, and extruded into plastic granules. The granules were additively manufactured, injection molded, injection stretch blown, or extruded. Previously, recycled PET products required more than 80% of virgin polyester pellets to maintain desirable properties. Currently, researchers work toward pellets with more recycled components. Nevertheless, some processes are long, with repeated crushing, mechanical purifying, purification, melting, and drying steps [75]. While multiple steps are needed to impart desirable qualities into recycled plastic, these processes can start reducing the sustainability of recycling if they are too long, consume too much energy, and release waste fluids into the environment. These processes need to be improved to increase the overall sustainability of dependent recycling end products.

4.7. Treatment

Impurities are generally removed by hand-sorting, flotation, alkaline/aqueous dissolutions, electrostatic separation, wind, density separation, power sorting, and cleaning [66,75]. These processes are all fallible. Hand-sorting is labor intensive, water cleaning produces contaminated wastewater (another pollution problem), and some other processes lead to poorly sorted waste [75]. Inventors have developed better technologies for the treatment of recycled plastics. Côté [85] provided a treatment process for waste thermoplastic polymers. Thermoplastic polymer or copolymer waste was dissolved in a specific solvent generating a combination of solids and liquids. This mixture was heated in the presence of an acid and filled into a supernatant of the plastic solution and solid waste residue. After separating the polymers from the residue, the recovered polymers were filtered and precipitated once or twice. The treated plastic passed through a flocculation step [85]. Another purification system patented by Layman et al. [86] refines polyethylene starting with heating the polymer to 80 °C to 280 °C at a pressure of 1.03–55.16 MPa in the presence of a liquid solvent. Recovering the polyethylene involves dissolution in two more solvents under different temperature and pressure conditions. More innovations like the filter washing bag by Guppyfriend are needed to reduce the release of microplastics into the environment during recycling [87].

5. Conclusions

The growing plastic waste problem poses a serious threat to humankind and the environment. Inventors and researchers fiercely attack this challenge, proposing open-source and patented technologies to achieve a circular economy. This study explored various technologies centered on mechanical recycling, as this kind of recycling has a lower environmental impact (fewer harmful byproducts, less energy used, and greater acceptability of various plastics). The study explores the technologies for producing construction materials, 3D-printing materials, furniture, pellet production, and multilayer materials. After establishing the plastic issue and challenges with recyclability, this paper picks mechanical recycling, a popular recycling technology, and investigates its possibilities and challenges. The paper points out prevailing setbacks in applying mechanical recycling and analyzes different solutions to these limitations. Then, the innovations are categorized based on the polymers recovered, the recovery technology used, and the complexity of the process. From this analysis, promising recycling processes are suggested for further investigation.

In summary, the study established:

- The suitability of repurposing different kinds and grades (including unsorted and unwashed) of plastic in various industries. PET, the most favored replacement, showed better results than other plastics without additives. The qualities of recovered plastic products vary based on the proportions of recycled materials to other materials and additives.
- Moderate and low percentages of (unadulterated) recycled plastic replacements across several industries, especially the construction sector, generally improve the quality of end products.
- Plastic replacements make end products lightweight, more resistant to chlorides, and more durable; reduce shrinkage, drying, permeability, and workability; and increase thermal and sound resistance.
- Inexpensively collected mixed-waste plastic finds several applications in concrete and composite board manufacturing.
- More mixed polymer recycling technologies are needed to reduce the resources used in sorting. Most mixed recycling technologies require compatibilizers, which improve mechanical properties when they do not exceed 3 wt.%. Thus, research should also improve compatibilizing substances.

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