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Review

Accelerating Plastic Circularity: A Critical Assessment of the Pathways and Processes to Circular Plastics

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Abstract: Achieving plastic circularity is imperative to using plastics without adverse effects. Today, only 9% of global plastic waste is recycled, signifying the need for more substantial advancements to accelerate our progress toward achieving plastic circularity. This article contributes to our collective efforts to accelerate plastic circularity by critically assessing the state-of-the-art, gaps, and outlook of the pathways and processes to circular plastics. It employs qualitative methods to derive new insights that empower scholars and practitioners to prescribe effective strategies to shape the future of plastic circularity and its research agenda. This article concludes that today's circularity pathways for plastics are not economically viable, significantly hindering their scalability and widespread adoption. It further validates that focusing on the product design and effectiveness of the available collection and sorting systems can considerably improve our progress in achieving plastic circularity.

Keywords: plastics; life cycle; circular plastics; circular economy; circularity; sustainability; mechanical recycling; chemical recycling; sustainable development

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1. Introduction

Plastics play an integral role in shaping our modern society and are ubiquitous in our daily lives [1–3]. Their superior material characteristics, performance, and low production cost make them desirable for vast consumer and industrial applications [4,5]. Correspondingly, plastic consumption has quadrupled in the past 30 years [6–8] and is estimated to double within 20 years [9–11]. As plastics' popularity have soared, they have become better, cheaper, and more accessible to mass consumers [12–14]. Eventually, an inflection point was reached wherein consumers questioned the need to clean and wash durable products, such as porcelain tableware, if there were single-use plastic alternatives they could dispose of after use. Consumers favored the latter, ultimately shaping the "throwaway living" culture we are familiar with today.

The "throwaway living" culture follows an unsustainable linear (take, make, waste) economic model that depletes our natural resources and contributes to global greenhouse gas emissions and pollution [7,8,15–17]. Today, plastics are discarded faster than adequately managed [18]. Nearly two-thirds of plastic waste comes from plastics under five years old, and only 9% are recycled globally [6]. Thus, these social behaviors are responsible for the ongoing global plastic crisis in which the full extent of its impact is still undetermined [6,8,19,20]. Evidence suggests its persistence in our anthroposphere has devasting environmental, economic, and societal implications. Lately, microplastics have even been discovered in our biological system, posing an immediate threat to our health [8,20–22].

In response, society has swiftly demonized plastics and attempts to limit their use have been initiated [23–25]. Today, interventions to limit plastic use include a ban on single-use plastics, extended producer responsibility policies, and an environmental tax on virgin plastic products. Despite the introduction of these interventions, modeling suggests that none of these state-of-the-art interventions combined can peak plastic consumption by 2050. At best, this integrated approach can only limit global plastic consumption to around 1.25 times the 2019 figures [11].

This is because plastics have substantial advantages over their competitors, making them difficult to be replaced entirely [26,27]. For example, plastics are lighter and more durable than their alternatives, making them a superior medium for transporting goods. If repeatedly and responsibly used, plastic-based products may even embody a lower carbon footprint than their alternatives [28]. Plastics also have a high secondary production value, estimated to be USD 120 billion annually, as they can be recycled or converted into fuels after their first use [6,10,29]. Additionally, the successful recovery and treatment of used plastics may save the industry and its insurers USD 20 billion over the following eight years [30].

Therefore, while we empathize with society's reaction to this issue and understand the rationale behind their response, these reasons have led us to firmly believe that limiting the use of plastics should not constitute a long-term solution. Instead, we trust that a more appropriate and robust long-term solution entails the safe, responsible, and circular use of plastics. Our position aligns with leading authorities and scholars recommending plastic circularity to solve the ongoing plastic crisis. Increasing evidence suggests that it could mitigate the adverse effects of adopting plastics while allowing them to continue contributing to society's betterment. For example, the United Nations Environment Programme (UNEP) [8] believes that achieving plastic circularity could eradicate plastic pollution. Additionally, they posit that it could reduce a quarter of greenhouse gas emissions across the global plastic life cycle, save governments USD 70 billion over 20 years and create 700,000 additional jobs due to system changes. At the recently concluded G7 Hiroshima Summit in 2023, the G7 Climate, Energy, and Environment Ministers unanimously acknowledged in their communiqué that achieving plastic circularity is urgent and imperative [31].

2. Methods

For these reasons, scholars must actively investigate and contribute their insights on how society can accelerate our transition to achieve plastic circularity. Most scholars attempt to answer this research question today with quantitative methods, such as life cycle assessments (LCA), where they evaluate the environmental impacts of plastics within well-defined system boundaries and parameters [32,33]. While LCA-based contributions are comprehensive, their most significant limitation is that their derived results and insights only apply to the investigated material and their corresponding parameters. Thus, they do not represent the entire plastic ecosystem. Moreover, our review indicates that even with the same types of plastics and assessment scope, there is the possibility of significant variations in results, leading to vastly different insights [34].

Therefore, an extensive qualitative review of plastics' circularity pathways is more appropriate for answering the same research question. By holistically assessing each circularity pathway's state of the art, we can better identify critical areas for improvement, leading scholars and practitioners to (a) prescribe effective interventions that accelerate our transition and (b) shape the future of plastic circularity and its research agenda.

2.1. Determining Plastics' Circularity Pathways

For this review, we first identified all the circularity pathways for end-of-life (EOL) plastics today. Figure 1 visualizes the flow of plastics and their complex interactions with the biosphere throughout their lifecycle. At the end of their useful life, plastics are primarily dumped in landfills or incinerated. Undergoing either process deters these plastics from circulating back into the ecosystem after their first use cycle, leading them to exit the value retention loop.

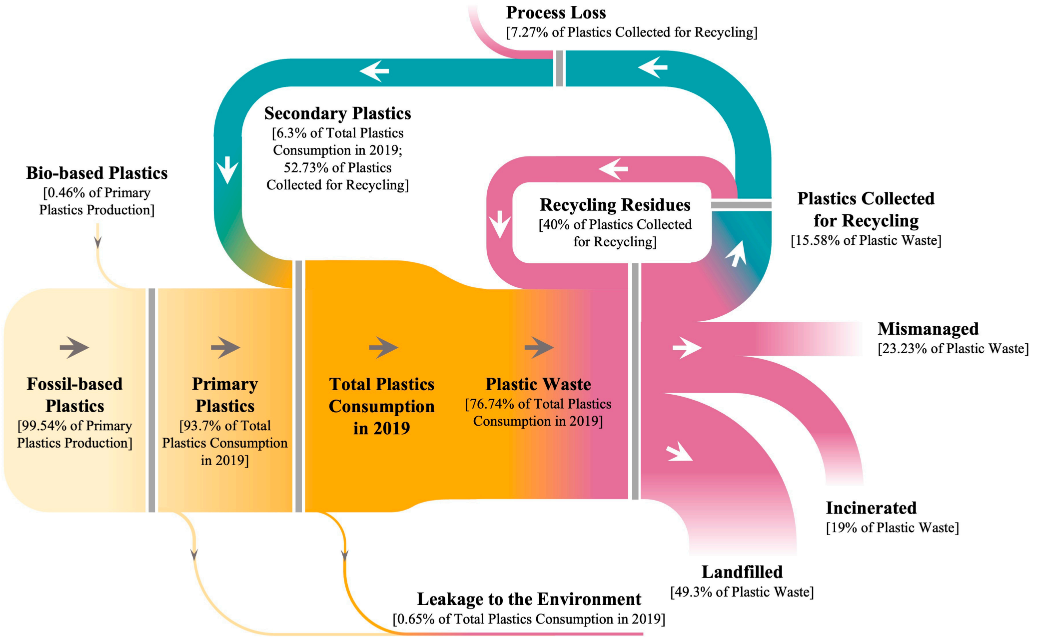

Figure 1. Material flows of plastics [6].

As circularity builds upon value retention loops, disposal in landfills or incineration is not recognized as a circularity pathway for plastics by UNEP [35]. Therefore, amongst the prevalent plastic waste treatment methods today, only recycling plastic waste is accepted as a circular pathway, as the recyclates can be channeled back into producing new plastics [36].

2.2. Assessing the State of Plastics Recycling

After determining plastics recycling as the only circular pathway, we critically assessed different plastics recycling technologies' state-of-the-art, gaps, and outlooks. These technologies can be grouped into mechanical or chemical recycling [37–39]. Figure 2 presents an overview of these technologies.

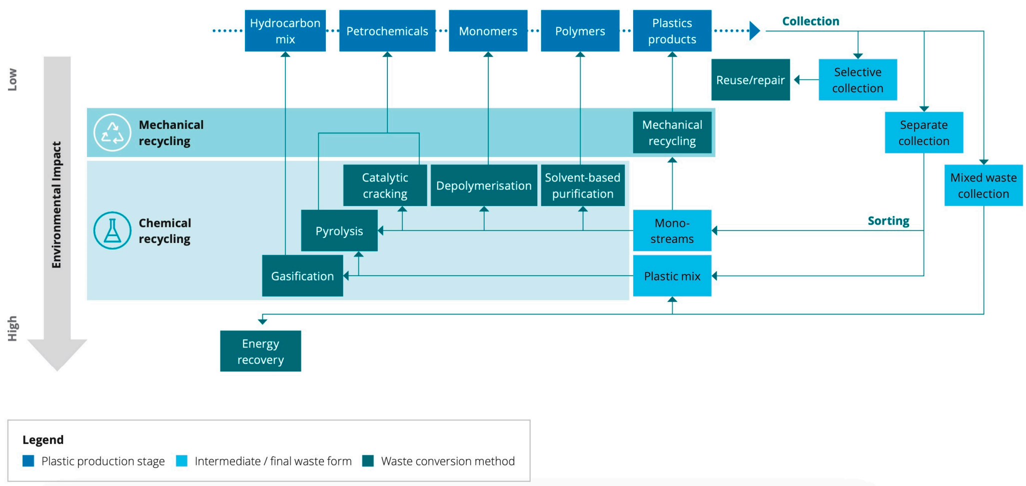

Figure 2. Plastics recycling technologies and their environmental impact. Reproduced with permission from Kerkhoven, et al. [40]: "Pathways towards circular plastics: Point of view", published by Deloitte Belgium, 2021.

Our assessment used refereed articles, books, policy papers, and other authoritative works across scholarly and nonscholarly platforms such as Scopus, Web of Science, and Google Scholar. We also examined news articles and press releases to identify the latest progress and developments that have yet to be covered in the academic literature. For relevancy, we prioritized works published from 2018 onwards and in English. We also prioritized highly cited articles. We present our findings in Section 3 (Mechanical Recycling) and Section 4 (Chemical Recycling). Additionally, we summarize the gaps observed from our review and discuss scholars' recommendations for addressing them in Section 5.

3. Mechanical Recycling

Mechanical recycling refers to processing plastic waste (or recyclates) into secondary raw materials or products without significantly changing the material's chemical structure [39]. Mechanical recycling optimizes the utilization of plastic resources [41,42] and has the least negative environmental impacts compared to other waste treatment methods [40,43]. It is also simple and straightforward to execute and effective in treating most thermoplastics [38,44,45]. Clear polyethylene terephthalate (PET), low-density polyethylene (LDPE), high-density polyethylene (HDPE), and polypropylene (PP) are the primary feedstocks and products of mechanical recycling globally [37,43]. Modern mechanical recycling is the most established and widespread plastic recycling method, accounting for almost all recycled [43,46–48]. Figure 3 illustrates a modern mechanical recycling process for plastic waste.

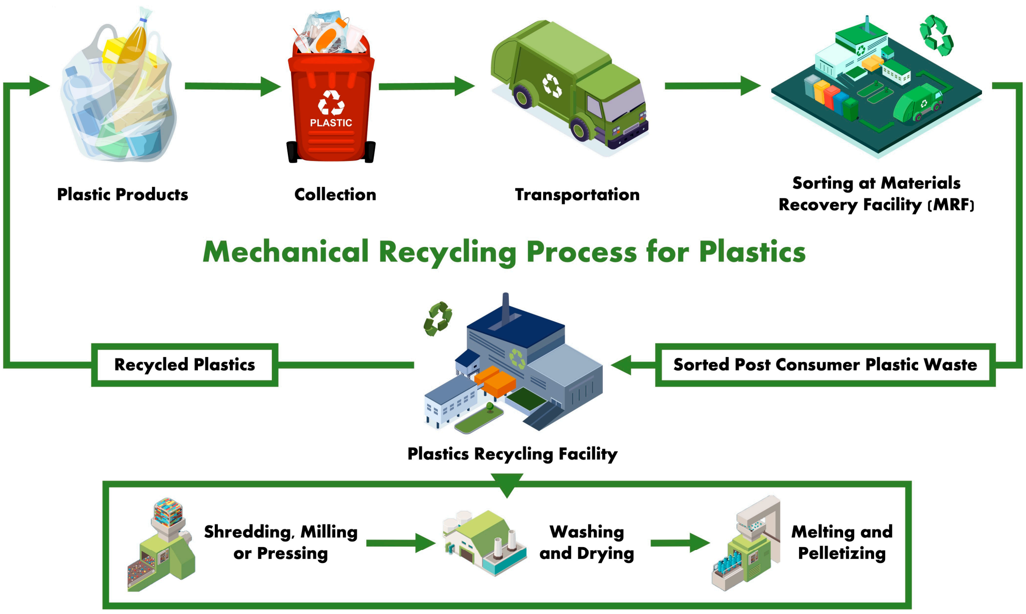

Figure 3. Modern mechanical recycling process for plastic waste.

A severe limitation of the mechanical recycling process is that its fragmentation and conversion stages may cause chain scission and thermo-oxidative reactions, which shortens the polymer chain length and degrades the recyclates' material properties, effectively downcycling it [36,49–53]. Subjecting recycled plastics to additional mechanical recycling cycles exponentially ages them and accelerates their degradation. For example, Schyns and Shaver [54] reported that subjecting recycled PP to five or more extra cycles causes chain scissions so severe that they limit its end-use applications. Therefore, mono-streams of clean films and rigid plastics such as pallets, boxes, bottles, and barrels are ideal feedstocks for mechanical recycling, as they can be directly processed into homogenous pellets using specialist recycling extruders [55].

3.1. Advances in Enhancing Properties of Mechanically Recycled Plastics

As recycled plastics tend to possess lower material performance than their virgin counterparts, they are often blended with virgin plastics in a 3:7 ratio to achieve good material properties for subsequent use. While this approach may reduce virgin plastic production globally, it does not prevent new virgin plastics from being introduced into our ecosystem, thus defeating the purpose of recycling plastic waste. To mitigate this phenomenon, targeted agents such as resins or additives may be introduced during the mechanical recycling process to modify certain aspects of recycled plastics and make them usable for higher-grade applications. For instance, inorganic fillers and surface-active agents can be added to improve the compatibility between fillers and plastics [56]. Elastomers, such as rubber, thermoplastic, and thermoset resin, can also be added to improve the recyclates' toughness [57]. Other known resins or additives may be added, including compatibilizers, antiaging agents, lubricants, stabilizers, plasticizers, and coloring agents [58,59].

Our review indicates that these manipulation techniques are gaining heightened interest and momentum in polymer composites research and have had recent notable advancements. For example, scholars have used fibers to enhance the overall performance of mechanically recycled thermoplastics to the point where it exceeds their original strength, modulus, and resistance values, making them suitable for use as engineering plastics and structural materials [60–63]. Additionally, scholars have actively experimented with polymer alloys and developed new composites such as wood–plastic composites that are resistant to moisture and corrosion [50,64]. While these manipulation techniques facilitate the valorization of otherwise inferior recycled plastics, scholars have expressed concern that these agents would accumulate with multiple recycling loops to a point where it disqualifies the recycled plastic from further mechanical recycling [53]. Therefore, the prospects of these targeted enhancements are promising if they do not contaminate and impede the recycled plastics from being further recycled.

3.2. Advances in Eliminating Contaminants and Impurities

Contaminants, even in small amounts, can reduce the output quality of recycled plastics and limit their end-use options, implicating their profitability and attractiveness to prospective buyers [52,65]. Naturally, as financial incentives are involved, applied research to eliminate contaminants and improve the quality and purity of recyclates has been gaining serious momentum. Contamination occurs while plastic waste is being sorted and prepared for recycling. There are no perfect sorting systems today. The state-of-the-art, combining manual labor with complementary sorting technologies, can achieve 99% sorting efficiency [19,66]. However, deploying and managing these best-in-class processes is costly, resulting in most mechanical recycling endeavors being unprofitable [67]. Yet, it remains in recycling facilities' best interest to maximize sorting effectiveness and efficiency, as suboptimal sorting performance can lead to increased contaminants that diminish plastics' recoverable value. As a result, scholars are actively investigating next-generation technologies that are practical, cost-effective, and capable of consistently achieving maximum sorting performance.

Precision waste-sorting robots are a recent technological advancement with high technology readiness levels and have been commercially deployed. For example, US-based AMP robotics has deployed at least 100 AI-powered precision waste-sorting robots since 2017. Their robots are designed for single-stream recycling systems and use computer vision to identify and sort objects. Compared to manual labor, they claimed that their robots are more accurate and twice as productive, thus reducing the unit cost of each sorting [68]. Another commercial player, the Netherlands-based company, Bollegraaf, uses near-infrared spectroscopy and height detection cameras to identify items based on their materials in 3D [69]. MIT, on the other hand, experimented with a soft Teflon hand embedded with tactile sensors to detect an object's size and stiffness with 85% and 63% accuracy when the object is stationary and on a conveyer belt, respectively [70].

Other notable technological advancements include tracer-based sorting (TBS) and digital watermarking (DW) technologies. These technologies shorten the time needed to accurately identify the recyclates by their chemical signature, thus allowing them to be rapidly sorted to their intended recycling route. For TBS, inorganic fluorescent pigments are embedded into the plastic substrate. These pigments do not alter the plastic's properties nor contaminate it, as only a tiny amount of these pigments—to the tune of 1–10 μg per cm^2 of printing ink or 1–10 ppm in the plastic substrate—are required to achieve reliable identification. Aesthetically, these pigments remain invisible to the naked eye and are only visible under UV light at the recycling facility [71]. DW works very similarly to TBS technologies. For DW, imperceptible postage stamp-sized codes are integrated into the packaging design. Specialized cameras can detect and act on these codes on high-speed sorting lines. An added benefit to using DW is that these codes can also be designed to provide consumers with relevant information such as nutritional values, provenance details, or recycling instructions [53].

Unlike precision waste-sorting robots, TBS and DW technologies require a systematic collaborative change to work as intended. Therefore, stakeholders' participation across the entire value chain is needed. We believe that aligning their interest and securing their commitment to change is a substantial hurdle. To overcome this hurdle, the Association des Industries de Marque, a European Brands Association, and the Alliance to End Plastic Waste, an NGO, introduced the "Digital Watermarks Initiative HolyGrail 2.0", the objective of which is to prove the viability of DW technologies for accurate sorting and the business case at large scale. The initiative has more than 160 participating companies and has made significant progress [72]. Overall, while TBS and DW technologies are nascent, they can be game changers if they can overcome their economic challenges [73] and be successfully implemented at scale, as they can increase recycling rates up to 80% [74] while reducing sorting and recycling steps [71].

4. Chemical Recycling

Chemical or feedstock recycling refers to any reprocessing technology directly affecting the formulation of polymeric waste or the polymer itself. It converts them into chemical substances and products, whether for the original or other purposes, excluding energy recovery [75]. Here, chemical substances and products include their constituents (e.g., monomers, oligomers), petrochemicals, and hydrocarbon mix [65,76,77]. All these can produce new plastic products of comparable quality to virgin plastics, thus eliminating downcycling [78–80]. Proponents believe it can complement mechanical recycling and valorize plastic waste it could not treat, thus increasing recycling rates and accelerating plastic circularity [48,80–82]. Conversely, critics still need to be convinced of its claimed environmental benefits, including the assumptions behind its life cycle assessments, and question its economic viability [45,65,81,83].

Presently, chemical recycling routes include solvent-based purification, depolymerization, pyrolysis, catalytic cracking, and gasification (Figure 2). By 2030, it is forecasted that pyrolysis and depolymerization will be the dominant chemical recycling routes, jointly capable of recycling at least 5% of today's plastic waste volume, a considerable step up from the state-of-the-art [48,84]. Conversely, recent scenario modeling by European Commission's Joint Research Centre concludes that gasification will not likely achieve economic viability within the next two decades [82]. Hence, as the scope of this article aims to identify gaps in plastic's circularity pathways and discuss ways to address them, we exclude gasification and focus on potentially feasible and viable chemical recycling routes of the near future.

4.1. Solvent-Based Purification

Even if plastic waste is perfectly sorted by type, the material composition of each plastic piece may still be contaminated with foreign elements. Depending on their end-use application, Roosen, et al. [85] estimated that the degree of contamination within each plastic piece could range from 10 wt % to 45 wt %. These innate contaminants can be removed using established solvent-based purification (SBP) techniques such as (a) extraction and (b) dissolution with precipitation. For extraction, the target polymer is washed with an appropriate solvent that dissolves and extracts the contaminants. While the extraction process is relatively simple and straightforward, it is only effective for cases involving low molecular weight contaminants [53]. Dissolution with precipitation technologies is preferred for complex cases involving multiple contaminants of varying solvency properties, molecular weight, or insoluble materials. Instead of washing, the target polymer is dissolved at the highest concentration of a suitable solvent, leaving the contaminants left behind to be filtered away. Antisolvents are then added to reprecipitate the polymers in solvents to retrieve virgin-like polymers [86–88].

SBP technologies for recycling various plastic types have been investigated for several decades. The fundamentals of recycling plastics with SBP techniques have remained unchanged since they were first established. Despite their heritage, we noted inconsistency in their classification. Some works consider them a chemical recycling process [89,90], while others do not [76,91]. The former argues that the processes constitute a chemical reaction and that fundamental chemistry knowledge is needed to comprehend and advance research in solvent–polymer interactions, their design, and recovery. Conversely, the latter argues that the chemical structure of the recovered polymers has not been significantly altered nor degraded during any SBP processes, which is a definitive feature of chemical recycling pathways [92]. Furthermore, plastic processing equipment can directly process the recovered polymers without further modifications [88].

Regardless of the scholarly debate, the merits of SBP techniques are that the recovered polymers are high quality and do not need further reform. As a result, SBP technologies score the highest in carbon dioxide avoidance (up to 6 tons of carbon dioxide avoided for each ton of plastic waste) compared to all other treatment methods [93]. However, our review indicates that cost is the most significant drawback impeding their widespread adoption. Firstly, SBP technologies use strong and hazardous organic solvents, which require certain technical and safety conditions to be satisfied before use [86,88,93]. Next, the unit cost of solvent is expensive and adds up when it is used on a commercial scale. Lastly, recovering solvent and antisolvent for subsequent reuse is energy-intensive, predominantly when the employed process operates at a high solvent/polymer ratio [53]. The removal of solvents must also be complete, as any residual solvent affects the target polymer's properties and, correspondingly, its resale value. These factors have led to the closure of some of the most established SBP facilities [94] and incidentally paved SBP's research direction.

Today, researchers are actively investigating alternatives to conventional solvents and antisolvents. Ideally, these emerging alternatives should be safe. They should be synergistic with their target polymer to optimize for (a) greater process efficiency during application and (b) recoverability and recyclability for subsequent use while not compromising on output quality [93]. Optimizing these conditions would lead to cost savings across the unit cost and associated recovery systems, paving the way to greater industry adoption and future developments. Green alternatives, such as supercritical fluids (e.g., dimethyl ether or carbon dioxide) and natural solvents (e.g., terpene oil), may enable these desired outcomes, leading to heightened research interest in their technical feasibility and unit economics for commercial implementations. Utilizing these green alternatives to purify enhanced recycled plastics (described in Section 3.1) for recycling has also emerged as an area of interest [87]. Beyond that, scholars are also actively investigating the optimal integration of SBP technologies with other treatment methods to separate mixed plastic waste more effectively and efficiently [86].

4.2. Depolymerization

Depolymerization refers to reversing a polymer to its monomer(s) or a polymer of lower relative molecular mass [95]. Methanolysis, glycolysis, and hydrolysis are several established depolymerization methods that are commercially available [96]. These methods correspond to the chemical agent that splits the plastics' functional ester groups at high temperatures to retrieve the target monomers. Methanolysis, glycolysis, and hydrolysis yield at least 65%, 50%, and 70% of the target monomers, respectively [97]. Known chemical agents include solvents, reagents, and catalysts such as glycol, methanol, water, manganese acetate [98], cobalt acetate [99], lithium hydroxide [100], n-butyl titanium oxide [101], acetic acid, sodium sulfate, and potassium sulfate.

Depolymerization is the only chemical recycling process capable of recovering an intermediate product (monomers), making it one of the most economically valuable approaches. It also uses less energy than the other chemical recycling processes [102] and can recycle cross-linked polymers [103]. After filtering out colorants and additives, the recovered monomers can be repolymerized into virgin-grade quality plastics, making them suitable for industrial applications where the plastics' purity is required [93,96]. However, depolymerization is only effective for homogeneous plastic waste and certain polymer types [76,91]. As a result, it is challenging for recyclers to acquire acceptable recyclates in high volumes to make the depolymerization process cost-effective and scalable [76].

Recent developments in depolymerization research aim to address these issues by widening its applicability. Researchers have successfully demonstrated selective depolymerization of targeted polymeric waste materials in nonhomogeneous plastic waste using hydrosilanes as reductants with organic [104], iridium(III) [105], aldehyde [106], ketones [106], and other catalysts. In their investigations, researchers observed that the effectiveness of the selective depolymerization process correlates with the reaction conditions, suggesting the need to optimize the recyclate–chemical agent pair and its associated reaction conditions to maximize the yield and output quality of the target monomers [107,108]. However, optimization increases the cost of the chemical agents, further diminishing the cost-effectiveness of the depolymerization process.

The cost of depolymerizing plastic waste is its most significant impediment, encouraging future researchers to pay attention to its commercial viability as they optimize the recyclate–chemical agent pair and its associated reaction conditions. Cost aside, the depolymerization process is also confronted with several technical issues that must be addressed before widespread adoption. They include the (a) small contact area between the chemical agent and the target polymer and (b) the challenges in recovering chemical agents in high purity after application [93].

4.3. Pyrolysis

Pyrolysis (or thermal cracking) refers to the irreversible chemical decomposition caused solely by a rise in temperature [95]. It is not incineration, as the plastic waste is thermochemically decomposed without oxygen [109]. Depending on the applied treatment method, pyrolysis occurs between 200 °C and 1300 °C. Pyrolyzing plastic waste at temperatures of 500 °C and lower produces liquid oil, while exceeding 500 °C produces more gaseous or char products such as gas, wax, and coke [110,111]. Pyrolyzing plastic waste in ideal conditions derives pyrolysis oil of similar performance to conventional diesel oil, including its viscosity (up to 2.96 mm^2/s), density (0.8 kg/m^3), flash point (30.5 °C), cloud point (~18 °C), and energy density (41.58 MJ/kg), allowing it to be used as a fuel or a feedstock to manufacture other compounds [112]. With remarkable versatility, pyrolysis has emerged as the chemical recycling choice for major chemical companies [113], enabling it to reach commercial maturity [91]. Today, it is the most extensively studied chemical recycling technology, with the most research output and data available [65]. Figure 4 illustrates a typical plastic waste pyrolysis process.

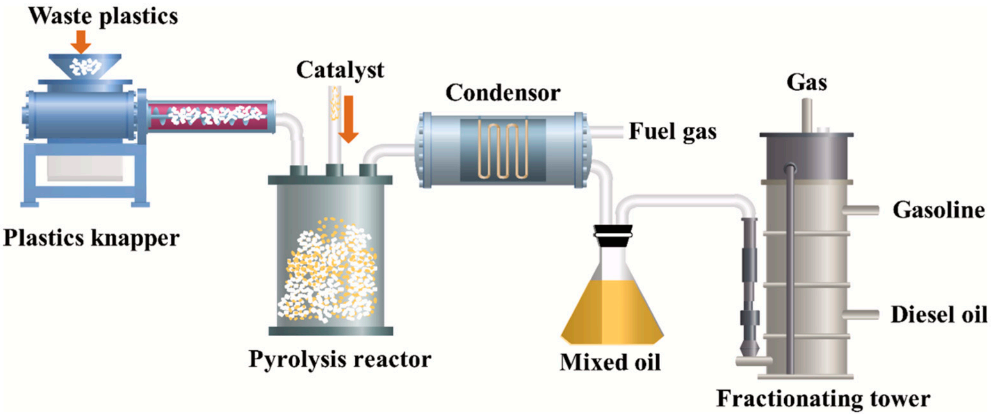

Figure 4. Flowchart of plastic waste pyrolysis process. Reproduced with permission from Maqsood, et al. [112]: "Pyrolysis of plastic species: A review of resources and products", published by Elsevier B.V, 2021.

Pyrolysis, like all other processes, is not perfect. While it can directly treat mixed plastic waste, it is not highly encouraged. The recyclates should be first sorted and purified for pyrolysis to produce virgin feedstock of comparable quality [83]. If pretreatment steps are not undertaken, the quality of pyrolysis oil produced may be inferior and unfit for its intended purposes. For example, subjecting polyvinyl chloride (PVC) to pyrolysis causes it to release corrosive hydrochloric acid. Therefore, if PVC is not removed before pyrolyzing an incoming batch of mixed plastic waste, its presence, even in small amounts, can contaminate and destroy the entire batch of recyclates and corrode the reactor. Researchers tried pretreating the recyclates at 300 °C for 60 min to mitigate this problem, reducing the embodied chlorine content by ~75 wt% [114]. They also tried to remove hydrochloric acid from PVC by grinding it with a planetary ball mill before adding calcium oxide to react with the hydrochloric acid to produce calcium salts that can be washed away [115]. Recently, plasma pyrolysis (pyrolyzing plastic waste between 1730 °C and 9730 °C in less than a second) and hydrocracking (adding pressurized hydrogen to the pyrolysis) have also emerged as feasible alternatives due to their ability to remove or prevent the formation of hydrochloric acid [96]. All in all, researchers today are actively investigating practical pretreatment strategies for pyrolysis to treat mixed plastic waste effectively. They are experimenting with better methods and agents, including inhibitors that prevent hydrochloric acid and other plastics from diminishing the output [116].

Another significant drawback is that pyrolysis is energy-intensive, typically carried out at very high temperatures of 500 °C [113]. Even though it is commercially matured, techno-economic analyses suggest it is not an economically viable solution yet [82]. Hence, in addition to devising practical pretreatment strategies, researchers are investigating cost-effective and environmentally friendlier approaches for pyrolysis. Presently, catalytic cracking is the most promising solution. It involves adding complementary transition metal catalyzers such as zeolites and silica–alumina to pyrolysis. These complementary catalysts enable pyrolysis to be more efficient at lower temperatures (300–350 °C) and use less energy [82]. It also facilitates the production of higher yield and better-quality pyrolysis oil. Both factors significantly improve its economic viability. However, unlike conventional pyrolysis, catalytic cracking is very sensitive to contaminants. Hence, the process cannot treat mixed plastic waste and is only suitable for pure polymers such as polyolefins and polystyrene. Plastic waste must also be pretreated before pyrolysis to achieve desired outcomes [117]. Adding catalysts and pretreatment processes increases the cost of pyrolysis. However, this is inevitable, as pyrolyzing polyolefins or polystyrene without catalysts produces pyrolysis oil of lower yield and quality to the extent that it requires further refining before use, resulting in an even higher processing cost [107]. Alternative solutions researchers explore today include microwave-assisted pyrolysis to lower energy consumption. In microwave-assisted pyrolysis, dielectric material or absorbents such as activated carbon, silicon dioxide, or graphene are introduced to absorb microwave energy. These absorbed microwave energies produce thermal energy that elevates the temperatures in the reactor, making it suitable for pyrolysis, thus reducing the energy supply needed to heat the reactor. However, microwave-assisted pyrolysis has several technical challenges, limiting it to laboratory and pilot scales only. These challenges include its inability to (a) uniformly heat the reactor and (b) precisely and consistently control the reactor's temperature with the absorbed microwave energies [96].

Other developments in improving its economic viability explore how to pyrolyze plastic waste to extract its high carbon content to produce other higher-valued materials. For example, Mohamed, et al. [118] co-pyrolyzed PET waste with zinc powder at 700 °C to create new carbon-based nanomaterials suitable for photocatalysis applications. On the other hand, Ko, et al. [119] valorized PET waste into synthetic graphite by first pyrolyzing the PET waste at 900 °C, followed by a boron-assisted catalytic graphitization at 2400 °C. Today, a kilogram of battery-grade synthetic graphite produces about 17 kgCO_2e [120] compared to a kilogram of virgin plastic at an average of 2.9 kgCO_2e [117]. Therefore, valorizing plastic waste into other higher-valued materials, such as synthetic graphite, helps offset the process cost and contributes more to lowering global emissions. However, this strategy does not accelerate plastic circularity. As such, we encourage researchers to stay focused on returning plastic waste to plastics.

5. Discussion

We have extensively reviewed the state-of-the-art of identified circularity pathways for plastics and determined where the gaps are. We conclude that every studied route and technology faces considerable challenges that diminish its adoption and impede society's progress toward achieving plastic circularity. The most significant challenge identified is the economic viability of the reviewed technologies and processes. The state-of-the-art is not cost-effective. Table 1 summarizes the key advantages and disadvantages of the technologies discussed.

Table 1. Key advantages and disadvantages of plastics recycling technologies.

Advantages	Disadvantages
Mechanical Recycling	
• Straightforward process and effective for most thermoplastics [38,44,45]. • Least negative environmental impacts [40,43].	• Degrades recyclates' properties [36,49–53]. • Limited recycling cycles [54]. • Low tolerance to contaminants [52,65].
Solvent-Based Purification	
• Able to treat contaminated plastic waste using dissolution with precipitation [86–88]. • Recovers high-quality polymers [86–88].	• Health and safety concerns arise from using strong and hazardous solvents [86,88,93]. • Unit cost of solvents and its associated recovery processes can be costly [53].
Depolymerization	
• Recovers monomers [95]. • Uses less energy than other chemical recycling processes [102].	• Low tolerance to contaminants [76,91]. • Expensive and only applicable to certain polymers [76,91].
Pyrolysis	
• Straightforward and highly versatile process, where different treatment parameters yield different outputs [110,111].	• Not cost effective [82]. • High energy requirements [113].

Fortunately, these technologies and their processes are under heavy development. Hence, scholars and practitioners believe that with the right conditions, costs can be lowered while increasing output yield and quality, leading to improved economic performance and adoption [82]. However, due to how nascent these respective technologies and their processes are, most studies lack real-world data and evidence to devise a path to achieving the right conditions. This is also a limitation of our study.

Nevertheless, scholars believe that achieving the right conditions may require collaborations with other industry players, who may be direct competitors. Therefore, scholars recommend that state actors intervene and facilitate early interactions among industry players by utilizing shared objectives, resources, and knowledge exchange to accelerate progress toward achieving the right conditions [17,19,121,122]. In addition to improving the technologies and their processes, scholars and industry practitioners believe that product design, collection, and sorting systems are equally critical factors and have provided recommendations for implementing them. We discuss their proposals and offer our perspectives and outlook in this section.

5.1. Product Design

One of the most frequently mentioned recommendations is to improve product design. Here, scholars refer to designing plastic products "for recycling" and "from recycling." Design "for recycling" refers to synergizing upstream design elements with downstream end-of-life treatment methods [123,124]. Successfully designing plastic-based products for recycling can reduce the inherent inefficiencies in the target circularity pathway, leading to potentially lower process costs, higher yield, and better quality. Presently, anecdotal evidence suggests low adoption of "design for recycling" principles, as industry players lack the incentive and knowledge to adopt them. Therefore, scholars recommend that policymakers lead the way with national-level goals, education, standardization, and certification programs [124,125].

For example, the South Korean government set and achieved a national-level goal of eliminating colored multi-material disposable plastic products by 2020, which they assessed as detrimental to their domestic plastic recycling efforts. They reached it using various policies and mechanisms, including education and standardization programs, that strongly encouraged companies to rethink their product designs and choices and only incorporate transparent mono-material plastics in their disposable products if necessary [126].

Singapore's Technical Reference (TR) 109 on Sustainable Packaging Guiding Framework and Practices is another excellent example of an industry standardization and education program. The TR specifies guidelines, criteria, and best practices in implementing the 3Rs (reduce, reuse, recycle) for business-to-business and business-to-consumer packaging, considering the life cycle of packaging, starting upstream from packaging design to end-of-life management, in Singapore's context. The TR targets Singapore's business leaders and believes its successful adoption can improve Singapore's recycling rates and capabilities.

Design "from recycling" refers to incorporating recycled base materials into the finished product. Designing the product to use these recycled base materials encourages material circulation in our ecosystem. In the case of plastics, it diverts them from the linear economic systems and reduces their footprint [127]. Plastic producers in places such as the European Union, Germany, and the United Kingdom, where targets for replacing virgin plastics with recycled plastics have been introduced, notably invest more in plastic recycling innovation and capacity. However, the methods and rules for calculating the recycled content of plastics today are still unclear, deterring industry players from committing more resources to scaling up their technologies and efforts [128]. Nevertheless, scholars believe that design from recycling will be a crucial strategy for society to accelerate our progress to plastic circularity if these critical issues can be resolved.

5.2. Collection and Sorting

In the previous section, we discussed the significance of collecting and sorting plastic waste before treating it. We also established that the waste stream's purity directly affects the output quality of the recycled plastics. Several scholars reported that most individuals have low awareness and understanding of this relationship and its implications, leading to unintentional actions undermining recycling rates [125]. They believe that consumer education would be sufficient to improve recycling rates to a desirable level.

However, we are skeptical of how much it can improve further. We can use Singapore's waste recycling efforts to illustrate our point. The National Environment Agency (NEA) in Singapore launched the "Recycle Right" campaign in 2019 to encourage Singaporeans to recycle correctly. As part of this campaign, recycling bin labels were redesigned to clarify what can or cannot be recycled. A comprehensive list and an easily accessible recycling search engine complement labels and aid residents in making the most informed decision [129]. Yet, after nearly three years of Singapore's best efforts to increase public awareness, advocacy, and education, 52% of waste in Singapore's recycling bins was contaminated or not recyclable [130]. Additionally, Singapore's plastic recycling rates only increased by 2% from 2019 to 2021 [131].

These results led us, and other scholars, to believe that complementing consumer education efforts with improvements to collection and sorting systems may yield better results [132–134]. One recommended improvement is establishing an environment that facilitates accurate sorting of plastic waste at the source. If carried out correctly, sorting at the source effectively reduces contamination, thus increasing the purity, recyclability, and value of recyclates. For example, the National University of Singapore reported that they effectively reduced the contamination rate of collected plastic bottles from about 60% to 27% on campus, and 79% to 29% in local shopping malls, with redesigned bins that facilitate and encourage the accurate sorting of plastic waste at the source [131].

Another recommended improvement is introducing extended producer responsibility (EPR) policies to extend a producer's responsibility to the post-consumer stage of their product's life cycle [135]. Effectively implementing these policies shifts the responsibilities away from municipalities and towards the producer, thus compelling them to consciously consider the environmental aspects of their products and their associated cost as they design them. Effective EPR policies can lead to increased collection and recycling rates, reduction in public spending on waste management, reduction in overall waste management costs, and more [136].

In the context of accelerating plastic circularity, this means mandating manufacturers and distributors to register the amount of plastic used in their products and take responsibility for collecting and recycling their products and product packaging [126,134,137]. These companies must contribute to a designated recycling facility for subsequent collection if they cannot fulfill their current obligations. Failure to do so incurs monetary penalties, typically higher than the contribution fees [134]. These mechanisms have been implemented in places such as Germany, Korea, and Belgium and contributed to increasing their respective recycling rates. Therefore, scholars believe that a localized EPR approach would significantly improve plastic recycling efforts and urge countries that have yet to enact any EPR policies to do so. Countries with an EPR policy should gradually work on expanding their policies to cover more types of plastic goods. For example, in 2022, Korea grew the range of plastic products covered in its EPR policies from 43 to 63 varieties, significantly increasing its domestic plastic recycling rates [134]. Finally, as all plastics within the system boundary are accounted for, data collected by implementing EPR contributes to developing national-level statistics for plastic material flow monitoring and analysis models.

6. Conclusions

Achieving plastic circularity is imperative. It helps to address the ongoing plastic crisis and mitigate the adverse effects of adopting plastics while allowing them to continue contributing to society's betterment. Today, recycling mechanically or chemically is the only accepted circular pathway for plastics. The remaining end-of-life treatment pathways for plastics do not allow the recyclates to be channeled back into producing new plastics. Modern mechanical recycling is the most established and widespread plastic recycling method, accounting for almost all recycled material. Nevertheless, chemical recycling technologies, except gasification, are expected to play a more prominent role by recycling at least 5% of today's plastic waste volume by 2030, a considerable step up from the state-of-the-art.

Regardless of the recycling technologies and methods, our review concludes that the most significant challenge impeding its scalability and widespread adoption is its economic viability. The state-of-the-art is not cost-effective, and the primary reasons can be attributed to the (a) cost of collecting, sorting, and preparing recyclates; (b) treatment process cost; and (c) any cost necessary to optimize the yield and quality of the output for commercial offtake, e.g., the catalyst required for catalytic cracking. While researchers and the industry are actively investigating ways to optimize these technologies' cost-effectiveness, other critical elements, such as product design and collection and sorting systems, must also be improved, as they significantly influence our progress in achieving plastic circularity.

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