

## CRITICAL REVIEW

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Cite this: DOI: 10.1039/d5su00506j

# Recent advances in chemical recycling and upcycling of plastic waste into valuable materials, chemicals, and energy: a comprehensive review

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The global plastic waste crisis has increased in severity in recent years: annual plastic production is projected to reach 500 million metric tons by 2025, and plastic waste accumulation is expected to surpass 12 billion metric tons. Despite these growing volumes, only ~9% of plastic waste is currently recycled; the majority is either landfilled, incinerated, or mismanaged, contributing to escalating greenhouse gas emissions—from 1.7 Gt carbon dioxide equivalent (CO<sub>2</sub>-eq.) in 2015 to an estimated 6.5 Gt CO<sub>2</sub>-eq. by 2050—and physical environmental pollution. This review provides a comprehensive overview of advanced plastic upcycling strategies to address this issue and recover value from diverse plastic waste streams. Recent developments in solvent-based dissolution, chemical depolymerization, and thermochemical conversions are examined for major plastic types, including polyolefins, polycondensation polymers, and PVC. Underlying reaction pathways, catalyst designs, and processing parameters that govern product selectivity, efficiency, and conversion yields are discussed in depth. Emerging techniques such as microwave-assisted depolymerization, tandem catalysis, and co-processing approaches are highlighted for their potential to enhance efficiency under milder conditions. Emphasis is also placed on the production of high-value products such as monomers, naphtha-range hydrocarbons, and syngas, and discussion is provided on catalyst stability, contaminant removal, scalability, life cycle effects on the environment, and technoeconomic viability. Finally, the review outlines future research directions focused on catalyst innovation, integrated process design, supportive policy frameworks, and interdisciplinary collaboration. All recommendations are aimed at accelerating large-scale implementation of plastic upcycling technologies and advancing the global circular plastics economy.

Received 27th June 2025  
Accepted 15th December 2025

DOI: 10.1039/d5su00506j

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## Sustainability spotlight

This review highlights recent advances in chemical upcycling of plastic waste into high-value materials, fuels, and chemicals, offering sustainable alternatives to landfilling and incineration. By examining solvent-based dissolution, catalytic depolymerization, pyrolysis, hydrothermal liquefaction, and gasification across key plastic types such as polyolefins and condensation polymers, scalable pathways for circular plastics management are highlighted. Emphasis is placed on technologies that reduce energy input, enable contaminant removal, and valorize complex waste streams. Integrating reaction engineering, polymer chemistry, design economics, and lifecycle thinking, this work aligns with the United Nations Sustainable Development Goals (SDGs) for sustainable development, more specifically SDG 9 (Industry, Innovation, and Infrastructure), SDG 12 (Responsible Consumption and Production), and SDG 13 (Climate Action).

## 1. Introduction

Plastics are ubiquitous in contemporary society. From medical devices to everyday consumer products and electronics, they have found their way into the daily lives of all on this planet. Therefore, the demand for plastic products has increased significantly over the years, with an estimated production of ~500 million metric tons for the year 2025.<sup>1</sup> This increase has also resulted in ever-growing amounts of plastic waste being generated globally. Unfortunately, existing plastic waste

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recycling infrastructure is severely lacking; current recycling rates sit at only  $\sim 9\%$ .<sup>2,3</sup> This lack of scalable infrastructure has resulted in the following three outcomes becoming the most common for polymer waste: landfilling, mismanagement, and incineration for waste-to-energy conversion. Each of those outcomes is associated with negative impacts on the environment. Leaching of toxic polymer additives from landfilled plastic waste poses a health hazard to those who live in surrounding areas. For example, in the case of nonylphenol, a commonly used polymer stabilizer and antioxidant, concentrations of up to  $96 \mu\text{g L}^{-1}$  have been reported in landfill leachate, which is significantly higher than the  $2 \mu\text{g L}^{-1}$  limit set by the European Union for the substance in surface water.<sup>4</sup> Though incineration of polymers provides usable energy, it generates large quantities of greenhouse gas emissions. The amount of  $\text{CO}_2$ -equivalent generated from incineration alone is estimated to reach 6.6 metric gigatons by 2050.<sup>5</sup> Much of the mismanaged plastic waste generated finds its way into the world's oceans, where it represents a hazard to marine life. In fact, it is estimated that by 2050, the weight of plastic in oceans may surpass that of all marine animals.<sup>6</sup> Mismanaged plastic waste also impacts the global economy, with over 100 million

USD spent annually on handling/cleaning such plastic packaging material.<sup>7,8</sup>

Mechanical recycling has been widely implemented to enhance material circularity.<sup>9,10</sup> With respect to the minor fraction of waste polymer that is currently being recycled, mechanical recycling has increased by approximately 57%.<sup>11</sup> Such processes are effective at handling specific clean polymer products such as polyethylene terephthalate (PET) water bottles or rigid PE containers. However, they struggle with mixed and contaminated waste polymer streams, often leading to down-cycling. For instance, frequent discoloration of PET during reprocessing has been reported and is primarily attributed to the reduction of antimony ions and the presence of polyamide contaminants in the waste stream, as noted by Berg *et al.*<sup>12</sup> Moreover, metal-based additives tend to accumulate with successive recycling cycles in the absence of effective metal removal strategies, making the materials unsuitable for human-related applications. Chemical recycling and upcycling technologies represent an alternative solution toward enhancing polymer sustainability. Although such processes currently account for only about 0.1% of all recycled plastics, they are quite robust and can generate valuable products from virtually

**Table 1** Representative structures of common plastics and their major depolymerization products<sup>13,14,16,17</sup>

Polymer	Representative structure	Major depolymerization products
ABS		Acrylonitrile, butadiene, styrene monomer, ethylbenzene, toluene, light oils
PC		Bisphenol-A, phenol, isopropenyl phenol, carbon dioxide
PS		Styrene monomer, ethylbenzene, toluene, benzoic acid
PP		Propylene, branched alkanes, alkenes
PE		Short-chain alkanes/alkenes, waxes
PU		Polyols, amines, carbamates, carbon dioxide, alcohols
PVC		Hydrochloric acid, chlorinated hydrocarbons, alkenes, and aromatics
PET		Bis(2-hydroxyethyl) terephthalate, mono-(2-hydroxyethyl) terephthalate, terephthalic acid



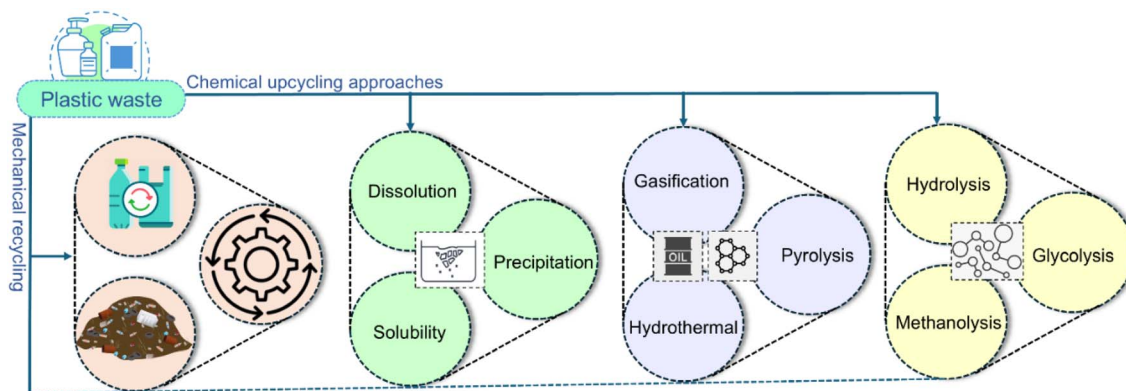


Fig. 1 Overview of recycling and upcycling strategies to convert plastic waste into valuable materials, chemicals, and energy.

all polymer waste types.<sup>11</sup> This has been proven by many lab-scale scientific studies and several other review articles published on the subject over the years.<sup>13–15</sup> More recently, novel approaches of chemical upcycling have been introduced.

This review presents an up-to-date, comprehensive overview of the state-of-the-art recycling and upcycling strategies to convert common plastic (summarized in Table 1 with representative structures and their corresponding depolymerized products) waste into value-added materials, chemicals, and energy (Fig. 1). Herein, energy denotes energy carriers such as liquid fuels, syngas, and hydrogen generated *via* thermochemical conversion; we do not imply a universal economic ranking relative to virgin polymers, as value is context- and market-dependent. This review covers strategies that enable the selective dissolution and recovery of pristine polymers, catalytic pathways for the depolymerization of PET and polyurethane (PU) into reusable monomers and polyols, and thermochemical techniques, including hydrogenolysis, hydrocracking, hydrothermal liquefaction (HTL), and gasification, for converting

plastic waste into naphtha-range hydrocarbons or syngas. In addition, the review explores microwave-assisted methods, tandem catalysis, and co-processing approaches that enhance product selectivity and energy efficiency. Key parameters influencing these processes such as catalyst composition, solvent choice, reaction conditions, and feedstock variability are examined. Finally, life cycle analysis (LCA) and techno-economic analysis (TEA) findings on such chemical recycling and upcycling technologies are presented to provide insight into both environmental impact and economic feasibility.

## 2. Upcycling of plastic waste *via* solvent-based processing into purified polymer

Upcycling of waste plastics can be defined as the depolymerization or conversion of the polymer into a product or products of higher value than the original waste itself. It can be achieved

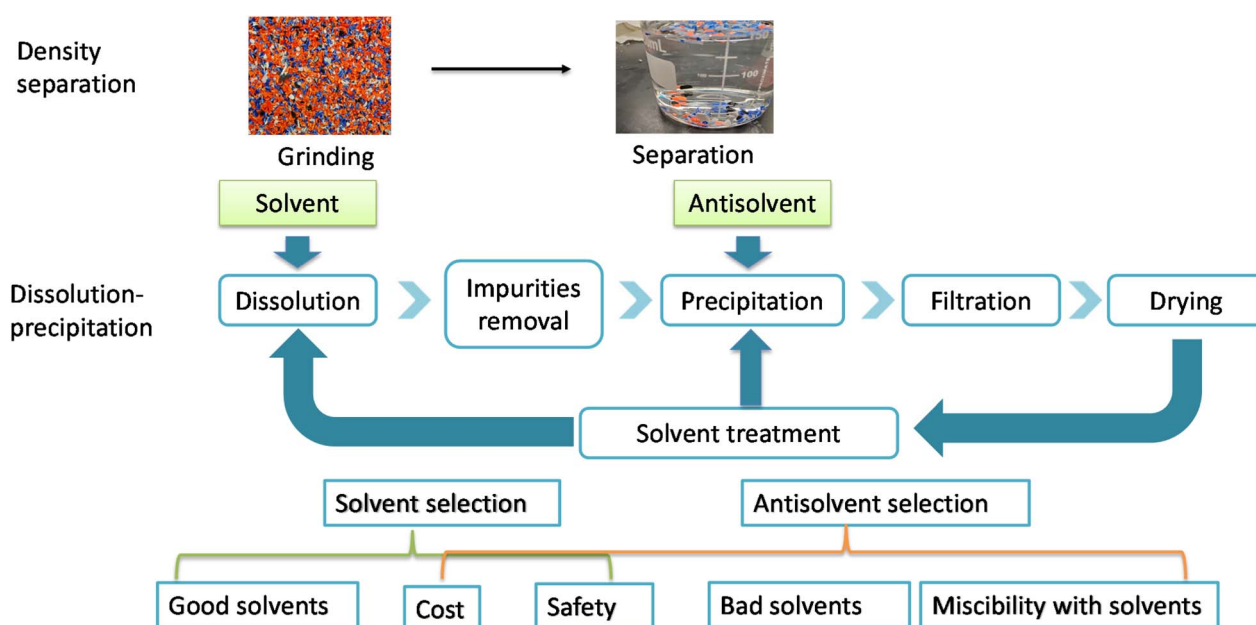


Fig. 2 Overview of polymer density separation and dissolution–precipitation.



via several different methods to generate various types of valuable products. One such upcycling pathway is the recovery of purified polymer through dissolution–precipitation or solvent-based extraction of waste plastics. Such methodologies, which employ solvents for polymer separation and purification of plastic waste, have emerged as promising complementary strategies to mechanical recycling. The general principle of solvent-based processing is selective dissolution. In the case of dissolution–precipitation, this refers to the dissolution of a single target polymer or all polymers except a target polymer; for solvent extraction, it relates to the targeted dissolution of unwanted contaminants exclusively. In either case, the key to achieving successful dissolution is to find an effectively selective solvent. An overview of both dissolution–precipitation and solvent extraction processes is shown below (Fig. 2).

## 2.1. Polymer solubility theory

In general, polarity is an important factor in chemical compatibility experiments. As described by the rule “like dissolves like,” a polymer with high polarity is likely to dissolve in a highly polar solvent, whereas a nonpolar polymer or a polymer with low polarity is soluble in nonpolar solvents or low-polarity solvents, respectively. It is also possible to build intermolecular interactions between the solute and the solvent. In this case, the polymer is easily dissolved as the interaction force exceeds the cohesive energy of the polymer.<sup>18,19</sup> Systems focusing on different aspects of dissolution relationships have been developed to describe and predict solubility behavior, including those of Hansen,<sup>18</sup> Kamlet–Taft,<sup>20</sup> Swain,<sup>21</sup>  $E_T(30)$  systems,<sup>22</sup> and Reichardt polarity.<sup>23</sup> In these solubility models, the polymer tends to dissolve in the solvents that have solubility parameters similar to its own.<sup>24</sup> The feasibility

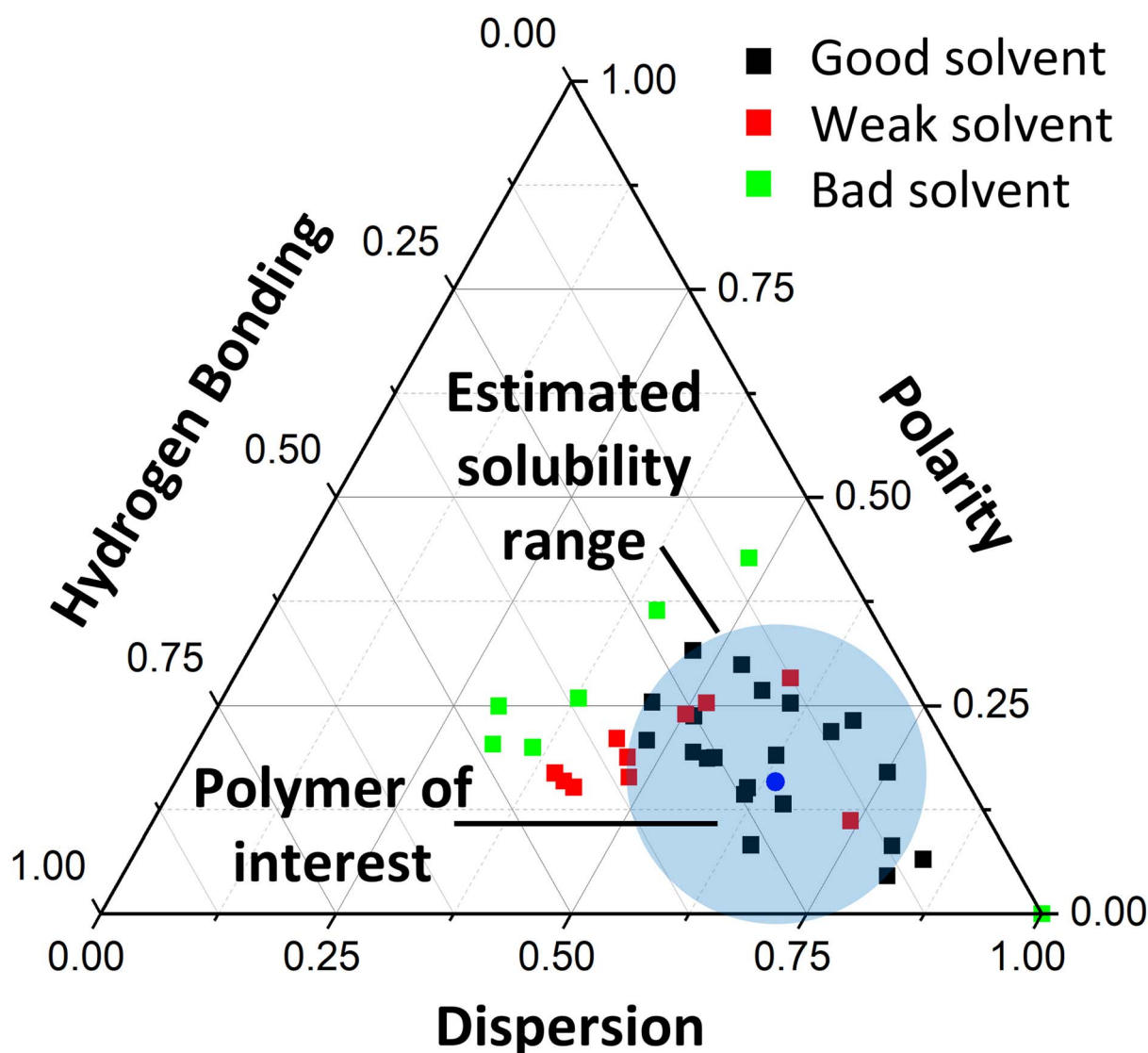


Fig. 3 Ternary Hansen solubility parameter plot showing the relative contributions of dispersion, polarity, and hydrogen bonding for solvents screened for the polymer of interest (open blue circle). Black, red, and green squares denote good, weak, and bad solvents, respectively; the shaded blue region indicates the acceptable solubility range (Hansen space) around the polymer, within which solvents are expected to dissolve the polymer effectively (adapted from literature).<sup>25</sup>



of solvent selection in dissolution-based upcycling largely depends on the accuracy of these predictions for polymer-solvent compatibility. Hansen solubility parameters (HSPs) are a comprehensive framework to evaluate solubility that incorporates three key intermolecular interaction components: dispersion forces, polar interactions, and hydrogen bonding.<sup>18</sup> A solvent is considered suitable for a given polymer if its coordinates in three-dimensional HSP space fall within the polymer's sphere of solubility (see Fig. 3 as an example).<sup>25</sup> This approach has been successfully applied in various polymer recovery studies. For example, HSP theory was used to identify appropriate solvents for selectively dissolving components in computer housing waste containing polystyrene (PS), polycarbonate (PC), and styrene-acrylonitrile copolymer.<sup>26</sup> It is important to note that consistent standards for the use of solubility parameters for solvent screening have yet to be established for such research work.

## 2.2. Dissolution-precipitation

As described by the solubility parameters, a good solvent can be defined as a solvent with sufficient solvation energy to solvate the target polymer, while a bad solvent lacks the energy to dissolve the polymer. As such, a solvent system can be established to recover dissolved polymer in solution by adding a bad solvent (antisolvent) to the good solvent such that precipitation of the dissolved polymer occurs. Such a system is known as dissolution-precipitation (Fig. 4). In general, the process requires selective dissolution of polymers as the first step, with precipitation subsequently achieved. Polymer composition identification is required prior to the process as additives and impurities may interact with solvents and affect dissolution performance.<sup>18</sup> Some examples of dissolution-precipitation processes using common solvents are provided in Table 2. Additional discussion, including the development of designable and feasible multi-component recycling systems through

tailored dissolution-precipitation strategies, is provided following the table.

Several studies have demonstrated effective recovery of targeted polymers from mixed plastic waste, including systems containing polymers that are challenging to dissolve. For instance, selective dissolution using a dichloromethane (DCM)/acetone solvent blend enabled the recovery of PC from computer housing waste with high yield (>95%) and purity (>95%).<sup>26</sup> In a laboratory-scale electronic waste recycling study, a multi-step solvent extraction process employing mixed solvents was devised to recover individual polymers from a complex mixture containing PC, PS, acrylonitrile butadiene styrene (ABS), and styrene acrylonitrile (SAN).<sup>30</sup> In another study, a solvent-antisolvent system based on *N*-methyl-2-pyrrolidone (NMP) and methanol was employed to separate polycarbonate from a polycarbonate/polyamide mixture. Notably, the solvents were reused, and a polycarbonate recovery rate of 90% was still achieved in the second cycle.<sup>33</sup> Solvent systems informed by thermodynamic solubility predictions have also been employed to guide targeted polymer recovery. For example, Walker *et al.* demonstrated the solvent-targeted recovery and precipitation (STRAP) process, which effectively deconstructs multilayer films into virgin-like resins with near-complete material recovery.<sup>34</sup> Specifically, toluene was used to selectively dissolve polyethylene (PE), while dimethyl sulfoxide (DMSO) was employed for ethylene vinyl alcohol (EVOH) extraction. Weeden *et al.*<sup>26</sup> found discrepancies between the predicted dissolution capabilities for isopropyl alcohol and THF and the experimental results obtained. Predictions of polymer solubility are affected by the diffusivity of solvents and the dissolution temperatures.<sup>34</sup> The HSP model may also overlook the true effect of electrostatic forces.<sup>34</sup> To improve the solvent screening accuracy and optimize dissolution-precipitation processing, several calculation techniques have been explored

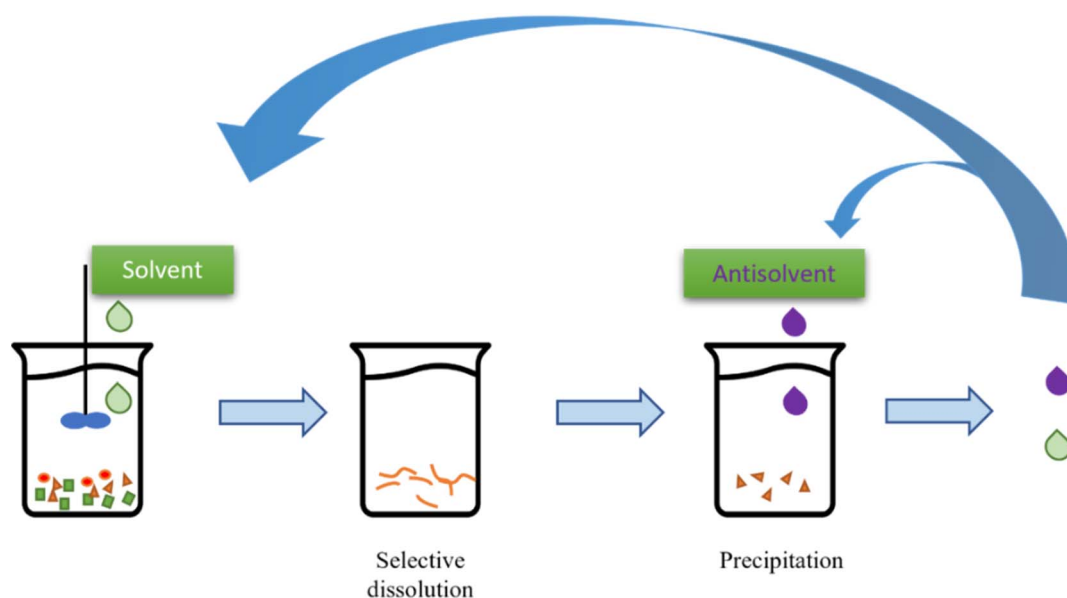


Fig. 4 Schematic drawing of the dissolution-precipitation technique.





Table 2 Examples of dissolution–precipitation processes using common solvents

Polymer	Solvents	Antisolvents	Description	Ref.
PE	Xylene	<i>n</i> -Hexane Methanol	Recovery of over 97% was obtained with solvent/antisolvent ratio of 1/3, and dissolution temperature of 100 °C	27
PE	Xylene	Propanol	Recovery of over 99% was obtained with solvent/antisolvent ratio of 1/3, and dissolution temperature of 85 °C	28
PP	Xylene	<i>n</i> -Hexane	Recovery of over 98% was obtained with solvent/antisolvent ratio of 1/3, and dissolution temperature of 140 °C	27
PS	DCM Toluene	Methanol	Recovery of over 92% was obtained with solvent/antisolvent ratio of 2/3, and dissolution temperatures from 25 °C to 100 °C	29
PS	Toluene Xylene	<i>n</i> -Hexane Methanol	Recovery of over 87% was obtained with solvent/antisolvent ratio of 1/3, and dissolution temperatures from 25 °C to 100 °C	30
PC	DCM Chloroform	Methanol	Recovery of over 95% was obtained with solvent/antisolvent ratio of 2/3, and dissolution temperatures from 25 °C to 100 °C	30
PC	Acetone/DCM (0–15 vol% ACE)	—	Acetone/DCM mixtures selectively extracted PC from ABS. Recovery of over 95% was obtained by evaporation–precipitation	29
PET	Benzyl alcohol	Methanol	Recovery of 99% was obtained with solvent/antisolvent ratio of 1/3, and dissolution temperature of 180 °C	31
ABS	DCM Acetone	Methanol	Recovery of over 90% was obtained with solvent/antisolvent ratio of 2/3, and dissolution temperatures from 25 °C to 100 °C	32
ABS	Acetone	—	ABS was recovered by dissolution at room temperature and evaporation–precipitation	32

for multiple-stage extraction, including molecular dynamics simulations and conductor-like screening models for realistic solvents (COSMO-RS).<sup>35,36</sup> For example, a combination of HSP, molecular dynamics, and COSMO-RS calculations was used to screen solvents, with predicted solubility over a range of temperatures also computed by COSMO-RS.<sup>37</sup>

### 2.3. Removal of polymer additives and impurities *via* solvent extraction

A major challenge in solvent-based closed-loop recycling is polymer contamination, either from other polymers or the

additives within the material itself.<sup>38,39</sup> It is possible to manipulate solvent systems to achieve polymer/polymer separation due to large differences in solubility. However, the removal of additives remains a problem. Polymer additives typically have a solubility range that is similar to or larger than that of the polymer itself and are often used in large quantities, such as 20–50 wt% for plasticizers and 10–20 wt% for flame retardants.<sup>40–42</sup> These organic chemicals can participate in the dissolution and remain as impurities in the recovered plastics, complicating the efficiency of the recycling process and affecting the reusability of reclaimed material. Hence, solvent-based additive extraction



has been attempted to purify polymers to enhance the quality of recycled plastics. Solid–liquid extraction is a widely used method in which targeted components are extracted from the solid matrix.<sup>43</sup> In principle, selective permeability is the main factor that dictates the extraction of desired substances. More specifically, solubility and diffusivity both directly affect the ability of a solvent to permeate through a solid matrix and remove unwanted chemicals. Because the solubility of additives is generally quite high in common solvents, multiple methods have been developed to improve the diffusion of such solvents into the permeable solid matrix. Accordingly, optimization of temperature and pressure as well as the addition of mechanical agitation forces has been explored.<sup>44,45</sup> Continuous extraction of additives can be achieved *via* both shake-flask extraction and Soxhlet extraction,<sup>46</sup> with the latter involving repeated boiling–condensation cycles. Ultrasonic extraction offers superior and better cavitation in the solid matrix, increasing permeation of solvent through the material.<sup>47,48</sup> Microwave-assisted extraction also promotes diffusivity by converting microwave energy to thermal energy in both the solid sample and the solvent.<sup>49,50</sup> Furthermore, supercritical fluids, following the principle of solid–liquid extraction, have been used in practice due to their improved solubility and diffusivity.<sup>51</sup>

#### 2.4. Reprocessing of polymers recovered from solvent-based processes

Compared to mechanical recycling, which usually leads to downcycling plastic mixtures, solvent extraction has the advantage of recycling or upcycling selective plastic material. Because of the superior ability to remove additives and contaminants, the recovered materials should have enhanced value as compared to other recycled plastics. However, information about the thermal and mechanical properties is lacking in depth; few studies have either re-processed or assessed the processability of polymers recovered from solvent-based processing. Evaluating how the polymers change during solvent-based polymer processing is important for not only remanufacturing but also for ensuring adequate performance of the recovered plastic that meets the requirements of the applications in which it is used. Arostegui *et al.* evaluated the degradation mechanism of ABS during dissolution-based recycling.<sup>52</sup> It was found that the polymer was more susceptible to heat and shear during subsequent injection processing due to the loss of desirable additives during the dissolution process. Walker *et al.* reported no significant effect of dissolution–precipitation processing on the  $T_g$  of the recovered PE.<sup>34</sup> Interestingly, this contrasts with findings by Yu *et al.* that identified changes in  $T_g$  for recovered PC after the dissolution–precipitation process.<sup>39</sup> They also identified differences in molecular weight in recovered PC samples processed with two different solvent/antisolvent systems, with probable degradation of the polymer occurring during NMP/water dissolution/precipitation treatment. Moreover, a study by Cecon *et al.*, focusing on the characterization of STRAP-recycled films, reported minimal changes in molecular weight and the absence of detectable heavy metals, highlighting potential safe use in food packaging

applications.<sup>53</sup> In conclusion, dissolution-based processes are capable of recovering purified polymer without sacrificing intrinsic properties. However, this is highly dependent on the solvent system utilized. A compounding process to add back lost desirable additives is likely required for the reclaimed polymer material prior to additional polymer processing. Additionally, polymer contamination and residual solvent retention are critical factors that influence the final properties and usability of the recovered materials.

Despite high yields and near-virgin quality of recovered polymer achieved through dissolution-based processing, several critical challenges arise from a techno-economic and environmental point of view. The environmental impact caused by the solvent-based processing itself represents a critical area of concern. Ideally, polymer dissolution should occur under mild conditions, paired with solvent recovery processes that are both accessible and energy-efficient.<sup>9</sup> Nevertheless, effective solvent recycling must also be integrated into process design to support comprehensive economic and environmental requirements as defined by industrial practice and legal regulations. The organic solvents themselves also often incur a substantial environmental burden, which has prompted growing interest in more sustainable alternatives such as bio-based solvents, switchable solvents, and supercritical fluids.<sup>54</sup> Moreover, to enable food-grade applications irrespective of the form or origin of the feedstock polymer, additional efforts are required to produce solvent-free, metal-mitigated polymers. This may be achieved through advancements in modular purification systems and improved drying technologies.

### 3. Upcycling plastic waste into valuable materials and chemicals

#### 3.1. PP and PE

Representing about half of all global plastics produced by volume, polyolefins are the most widely utilized class of polymer material. From oil containers to yogurt cups, agricultural films and much more, they have become an integral part of modern society due to high affordability, versatility, and lightweight properties.<sup>55</sup> In fact, PE alone represents ~34% of annual global plastic production.<sup>56</sup> Unfortunately, current recycling rates do not match this large production volume: a significant amount of polyolefin waste ends up in landfills, is incinerated, or is mismanaged as ocean plastic waste.<sup>17</sup> Given this issue, much recent attention in the scientific community has been placed on developing strategies to improve polyolefin end-of-life outcomes. More specifically, interest has increased in upcycling technologies rather than existing conventional chemical recycling methods. The past five years have seen a significant jump in the number of publications on the topic, including entire review articles specifically highlighting polyolefin upcycling research *via* methodologies such as functionalization and selective breakdown into valuable chemicals (Fig. 5).<sup>57,58</sup>

One of the more promising approaches investigated by researchers in the past few years has been oxidative upcycling of polyolefins. For instance, Chang *et al.* employed controlled



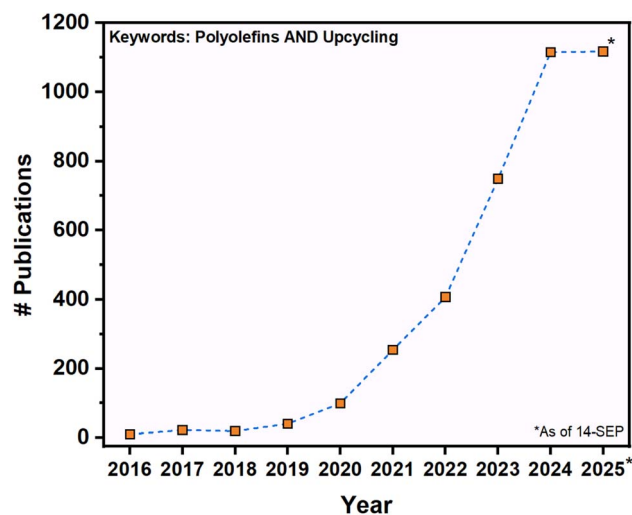


Fig. 5 Number of publications related to polyolefin upcycling since 2016. Data acquired from dimensions AI scientific database using the "AND" boolean operator. Specific unrelated scientific fields were excluded using the "NOT" boolean operator.<sup>59</sup>

oxidative cracking to treat polyolefins, obtaining functionalized oligomers from PE waste. The oligomers were subsequently demonstrated as a suitable feedstock in the synthesis of a dynamically crosslinked elastomer.<sup>60</sup> The introduction of nitrogen during the oxidative degradation process to increase the value of products obtained has also seen lab-scale success. Employing controlled oxidative degradation with nitric acid, Wei *et al.* acquired nitro-containing carboxylic acids from both PP and PS waste.<sup>61</sup> Similarly, Zhao *et al.* utilized  $\text{MnO}_2$  in the conversion of waste PP and PS into high value nitriles, amides, and acetone.<sup>62</sup> Nitriles and amides are common compounds in both the pharmaceutical and petrochemical industries.

Recently, catalytic hydrogenolysis technology has emerged to selectively cleave the C–C bond within the polymer, allowing for

upcycling of PP and PE. Many heterogeneous (*e.g.*, noble metal catalysts such as Ru and Pt) and homogeneous catalyst systems have been explored.<sup>63</sup> Zhao *et al.* utilized  $\text{Ni/SiO}_2$  to hydrogenate PE and obtain gasoline and diesel with a hydrocarbon content of 81%.<sup>64</sup> Sun *et al.* used a Pt-based catalyst to hydrogenate LDPE into gasoline-range alkanes with a 63 wt% yield.<sup>65</sup> Rorrer *et al.* used a Ru-based catalyst for the hydrogenolysis of a PP and PE mixture (1:1) to obtain branched alkanes at conditions under 225 °C for 24 h and 40 bar  $\text{H}_2$ .<sup>66</sup> Ma *et al.* investigated the effect of temperature on hydrogenolyzing PP and PE into liquid alkanes by utilizing Ru/C as a catalyst.<sup>63</sup> Chen *et al.* utilized Ru as a catalyst at 200–225 °C and 30 bar and found that the high  $\text{H}_2$  pressure promoted hydrogenolysis of internal C–C bonds, reducing methane production and favoring linear alkane over branched alkane formation.<sup>67</sup> Zare *et al.* studied the effect of reaction media on hydrogenolysis of PE with Ru/ $\text{TiO}_2$  catalyst. The short-chain alkanes (octane, hexadecane and dotriacontane) influenced the polymer adsorption on Ru/ $\text{TiO}_2$  surfaces, altering the C–C bond cleavage kinetics.<sup>68</sup> Gdowski *et al.* addressed the impact of temperature under hydrogenolysis reactions catalyzed by Pt–Re/ $\text{SiO}_2$  at 140–200 °C under 500 psi  $\text{H}_2$ ; no hydrogenolysis was observed at 140 °C with rapid hydrogenolysis at 200 °C.<sup>69</sup>

Hydrocracking of polymers also appears to selectively recover useful chemicals from polyolefins. Jumah *et al.* used a bifunctional catalyst (Pt-loaded zeolite, Beta and USY) to hydrocrack LDPE, HDPE, PP and PS under mild reaction conditions (330 °C, 20 bar  $\text{H}_2$ , 30 min), resulting in low molar weight hydrocarbons with higher selectivity towards gases of  $\text{C}_3$ – $\text{C}_4$ .<sup>70</sup> In another study by Bin Jumah *et al.*, the kinetic model for LDPE hydrocracking with bifunctional zeolite (1% Pt-beta) at 250–300 °C and 20 bar  $\text{H}_2$  for 0–40 min was examined. The results showed that Pt- $\beta$  holds great potential to hydrocrack LDPE and yield a rich naphtha stream (25–30 wt%).<sup>71</sup> In addition to using hydrogen alone, Chen *et al.* investigated the effectiveness of different hydrogen donors (*i.e.*,  $\text{H}_2$ , tetralin and

Table 3 Hydrogenolysis and hydrocracking of PE and PP

Feedstock	Reaction conditions	Major products	Ref.
<b>Hydrogenolysis</b>			
PE/PP	200–225 °C, 30 bar $\text{H}_2$ , 3 h, Ru/C	$\text{C}_6$ – $\text{C}_{38}$ alkanes (57%)	67
PE/PP	250 °C, 30 bar $\text{H}_2$ , 18 h, 0.5 wt% Ru/ $\text{CeO}_2$	$\text{C}_6$ – $\text{C}_{40}$	77
LDPE/HDPE/PP	200 °C, 60 bar $\text{H}_2$ , Ru/ $\text{CeO}_2$	$\text{C}_5$ – $\text{C}_{21}$ , $\text{C}_{22}$ – $\text{C}_{45}$	78
PP	200 °C, 20 bar $\text{H}_2$ , 6 h Ru/ $\text{TiO}_2$	$\text{C}_5$ – $\text{C}_{21}$ (38%), $\text{C}_1$ – $\text{C}_4$ (11%)	79
LDPE/PP	270 °C, 30 bar $\text{H}_2$ , 8 h, Co/ $\text{SiO}_2$	$\text{C}_5$ – $\text{C}_{30}$	80
LDPE	300 °C, 12 bar $\text{H}_2$ , Pt/ $\text{SrTiO}_3$	$\text{C}_1$ – $\text{C}_8$	81
HDPE	250 °C, 8 bar $\text{H}_2$ , Pt/ $\text{SiO}_2$	$\text{C}_9$ – $\text{C}_{18}$	82
LDPE	250 °C, 40 bar $\text{H}_2$ , 40 h, Co/ZSM-5	$\text{C}_3$	83
PP	300 °C, 50 bar $\text{H}_2$ , 4 h, Ru/FAU	$\text{C}_4$	84
LDPE	250 °C, 50 bar $\text{H}_2$ , 16 h, Ru/C	$\text{C}_4$	85
<b>Hydrocracking</b>			
PE/PP	250 °C, $\text{H}_2$ , Pt- $\text{WO}_3$ / $\text{ZrO}_2$	Gasoline-range hydrocarbons (85 wt%)	73
HDPE	270 °C, 30 bar $\text{H}_2$ , 2 h, Ce/PtSn	$\text{C}_5$ – $\text{C}_{12}$ (77 wt%)	74
PE	180 °C, Pt-zeolite	$\text{C}_5$ – $\text{C}_{12}$ (80 wt%)	75
PE	250 °C, 30–60 bar $\text{H}_2$ , Ni/BEA	Naphtha (85 wt%)	76
LDPE/HDPE/PP/PS	330 °C, 20 bar $\text{H}_2$ , 30 min	$\text{C}_3$ – $\text{C}_4$	70
PP	280 °C, 10 bar $\text{H}_2$ , zeolite HMF1	$\text{C}_1$ – $\text{C}_5$	86





Table 4 Different chemolysis reactions to recover valuable chemicals from PU<sup>115,116</sup>

Process	Reagents	Products or yield
Hydrolysis	Water vapor and alkali metal hydroxide catalysts	Amines and polyols with undesired by-products
Alcoholysis	Alcohol and metal hydroxide	Polyols and urethane products
Acidolysis	Inorganic and organic acids	Amides, polyols, amine salts, and oligo-ureas
Aminolysis	Hydroxyl and amino-derived compounds	Polyols and polyamine compounds
Glycolysis	Diol compounds	Recovery and reuse of polyols

H<sub>2</sub> + tetralin) during hydrocracking of PE.<sup>72</sup> The results showed that the most effective H-donor for fuel production remains as H<sub>2</sub>, with 65.8% oil composition within the C<sub>6</sub>–C<sub>12</sub> range when PE was treated at 350 °C for 60 min.<sup>72</sup> Liu *et al.* evaluated tandem catalysis for hydrocracking of LDPE over Pt/WO<sub>3</sub>/ZrO<sub>2</sub> and HY zeolite in hydrogen at temperatures as low as 225 °C. The selective conversion of LDPE into gasoline-range hydrocarbons was achieved, and the resulting hydrocarbons had yields up to 85%.<sup>73</sup> Wang *et al.* investigated the incorporation of cerium into the PtSn catalyst for hydrocracking of HDPE at 270 °C and 30 bar H<sub>2</sub> for 2 h. The results demonstrated that the optimal performance was achieved at a catalyst loading of 0.5 wt%, yielding 77 wt% gasoline-range products (C<sub>5</sub>–C<sub>12</sub>).<sup>74</sup> Tian *et al.* presented a novel catalyst design strategy to achieve precise control of Pt cluster positioning in the channels of H-beta zeolite. The strategic approach led to a 98% yield of short-chain alkanes at 180 °C, with 80% C<sub>5</sub>–C<sub>12</sub> selectivity.<sup>75</sup> Vance *et al.* hydrocracked PE into naphtha (achieving a maximum 80% yield with 85% selectivity) at 250 °C and 30–60 bar H<sub>2</sub> by using Ni/BEA (nickel based Beta zeolite) as a catalyst.<sup>76</sup> The state-of-the-art hydrogenolysis and hydrocracking processes for both PE and PP are summarized in Table 3.

### 3.2. PET

In recent years, the chemical recycling of PET has shown significant progress, as various chemolytic methods have been successfully implemented for industrial-scale applications. The most widely studied approaches have been methanolysis, alcoholysis, hydrolysis, aminolysis, ammonolysis, and glycolysis.<sup>8,13,17</sup> Depending on the solvent utilized, PET can be depolymerized through the same basic degradation mechanism to yield a range of value-added products with the addition of a catalyst. Extensive studies on the topic of PET chemolysis have been reported in the literature.<sup>8</sup> Thus, this section focuses primarily on the fundamental reaction mechanisms, theoretical insights, and existing challenges associated with the chemistries involved during chemolytic PET depolymerization.

**3.2.1. Catalytic PET chemolysis.** The fundamental degradation mechanism is generally applicable to all transesterification catalysts used for PET depolymerization regardless of solvent type: the catalyst facilitates nucleophilic attack on the ester bond to accelerate depolymerization. Various metal catalysts have been used for PET degradation, primarily through the formation of coordination bonds between the metal center and the carbonyl oxygen of PET.<sup>87</sup> For example, metal catalysts such as zinc acetate (Zn(OAc)<sub>2</sub>) promote glycolysis *via* a first-order, Lewis-acid mechanism;<sup>88–90</sup> metal cations

form coordination complexes with the carbonyl oxygen of PET, enhancing susceptibility of the ester bond to nucleophilic attack and facilitating bond cleavage. The nature and strength of these metal coordination sites can significantly impact the catalyst's Lewis acidity, which affects both product selectivity and the depolymerization pathway.<sup>15</sup> The literature suggests that if Lewis acid–base interactions are too strong, there will be insufficient free acid and base to enable depolymerization. Conversely, if these interactions are too weak, then catalyst components may not form stable complexes under depolymerization conditions. Different metal halides have also been used for PET depolymerization, such as in the case of Delle Chiaie *et al.*; in their work, PET depolymerization was carried out using different metals (*e.g.*, Zn, Mg, Bi, Y, Fe, Sn, Ti) combined with different halogens (*e.g.*, Cl, Br, I). The results showed that an increase in the size of halides (from Cl to I) influenced catalytic activity. It was observed that hard metals, such as Mg, tend to bind more strongly to oxygen-based anions (*e.g.*, acetates) than soft metals, which can lead to a reduction in catalytic activity.<sup>91</sup>

In glycolysis, the central metal atom of the catalyst plays a vital role in influencing PET depolymerization and reaction mechanism through its coordination ability, facilitating bond formation between cations and the ester group of PET.<sup>87</sup> The formation of these metal–ligand complexes reduces the electron density of the carbonyl group, thereby promoting a nucleophilic attack by the hydroxyl oxygen. Additionally, the ligands in metal catalysis are important because they can significantly enhance the Lewis acidity of the metal center, provide additional coordination sites, and influence reaction selectivity, catalyst deactivation, and solubility.<sup>92,93</sup> Chiaie *et al.* explored combining organic bases with Lewis acidic metal complexes for PET conversion into BHET. It was discovered that magnesium (hard metal) binds strongly to oxygen-based anions (*e.g.*, acetate), which can reduce catalytic activity. In contrast, soft metals such as zinc exhibited higher catalytic activity when coordinated with oxygen-based anions as compared to halide ligands.<sup>91</sup>

Wei *et al.* explored DBN (1,5-diazabicyclo[4.3.0]non-5-ene)-based catalysts to investigate their catalytic activity and mechanism for PET depolymerization.<sup>61</sup> The combination of DBN/urea and EG was found to increase the electronegativity of –OH on EG, while the interaction between DBN/urea and PET enhanced the electrophilicity of carbonyl carbon on the PET ester group. This dual activation accelerated PET depolymerization rates. *In situ* FTIR analysis further confirmed that DBN/urea increased the nucleophilicity of the hydroxyl groups, facilitating the cleavage of ester bonds. The synergistic effect of



the hydrogen bond donor (*i.e.*, DBN) and acceptor (*i.e.*, urea) promoted the fracture of PET ester linkages, thereby enhancing the depolymerization kinetics.

Besides the different types of catalysts and reaction conditions, the co-solvent approach has also been adopted to boost PET depolymerization kinetics under mild reaction conditions. For example, Schlüter *et al.* investigated the co-solvent approach of  $\gamma$ -valerolactone (VL) with ethylene glycol (EG) in the presence of  $\text{Zn}(\text{OAc})_2$  catalyst. The study showed that the incorporation of VL significantly enhanced the reaction kinetics; the improvement was found to be due to the interaction between  $\text{Zn}(\text{OAc})_2$  and VL, which facilitated acid–base synergistic catalysis.<sup>94</sup>

One of the major challenges in the chemical upcycling of PET, particularly for textile waste, is the accumulation of dyes, colorants, and other various inorganic contaminants (*e.g.*,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Ca}^{2+}$  and  $\text{Zn}^{2+}$ ). Traditional solvents are often ineffective in removing these colorants during depolymerization. To address this issue, Wang *et al.* used hydrophobic deep eutectic solvents (DES) (*e.g.*, menthol:oleic acid (Men:OleA)), which are considered a green reaction medium.<sup>95</sup> The decolorization process followed a pseudo-second-order kinetic model and was primarily driven by physical interactions such as hydrogen bonding and van der Waals forces, thereby supporting the closed-loop recycling of PET textile waste.

**3.2.2. Energy-assisted PET chemolysis.** In recent years, microwave-assisted glycolysis of PET has gained significant attention as an effective approach to accelerating depolymerization. Several mechanisms have been proposed to explain the behavior of PET under microwave irradiation. One such mechanism is the thermal effect, where the reduction in PET depolymerization efficiency *via* glycolysis is primarily associated with the uncontrolled temperature growth of the reaction system.<sup>96</sup> Another specific microwave effect occurs when the applied electric field induces change in the orientation of molecules, resulting in an alteration of activation energy such that rapid depolymerization is favored.<sup>97</sup> Kržan *et al.* utilized the microwave-assisted (500 W) approach to investigate PET glycolysis using  $\text{Zn}(\text{OAc})_2$  and achieved complete depolymerization in 10 min.<sup>98</sup> However, the catalytic activity of  $\text{Zn}(\text{OAc})_2$  was reported to be independent of the heating method. Similarly, Chaudhary *et al.* studied PET glycolysis under microwave irradiation (2.454 GHz, 0.0016 eV) using  $\text{Zn}(\text{OAc})_2$  and confirmed that the Lewis acid mechanism of  $\text{Zn}(\text{OAc})_2$  operates similarly under both microwave and conventional heating conditions.<sup>99</sup> Chen *et al.* further studied the kinetics of  $\text{Zn}(\text{OAc})_2$  for PET glycolysis under microwave irradiation using the shrinkage core model, demonstrating that the film diffusion was the rate-limiting step. The microwave irradiation decreased the reaction time significantly by lowering the activation energy ( $\sim 36.5 \text{ kJ mol}^{-1}$ ), as compared to the same process observed under conventional heating.<sup>100</sup> Additionally, other catalysts including sodium carbonate, sodium bicarbonate, and barium hydroxide for PET glycolysis to obtain BHET under microwave irradiation were also studied by Pingale and Shukla. They reported a significant time reduction as compared to conventional heating: from 8 h to just 35 min.<sup>101</sup>

Microwave-irradiation depolymerization has also been explored using various nucleophiles including alkali, ammonia, alkali hydroxide, propylene glycol, ethylene glycol, and ethanolamine to obtain different distributions of products. Mohammadi *et al.* investigated the effect of product distribution obtained from microwave irradiation and conventional methods. Their findings revealed that both methods result in the same product distribution. However, microwave irradiation effectively reduced the temperature or reaction time, offering a more energy-efficient pathway for PET recycling.<sup>102</sup> Beyond microwave irradiation, Jamdar *et al.* investigated the effect of gamma irradiation on the properties of PET and subsequent treatment *via* glycolysis. The pre-processing of PET waste with gamma radiation resulted in a significantly higher yield of BHET as compared to non-irradiated PET processed through conventional methods.<sup>103</sup>

**3.2.3. Challenges for PET chemolysis.** PET has huge potential for chemical upcycling and recycling to be implemented at an industrial scale. However, several technical, economic and environmental challenges exist which need to be addressed to enable its operation. Techno-economic analysis (TEA) and life cycle assessment (LCA) are valuable tools that can help guide the design and operation of the systems at an industrial level and thus potentially address economic and environmental challenges. McNeeley *et al.* conducted a detailed assessment of different PET depolymerization routes for a circular economy, evaluating process design options and modeling for methanolysis, glycolysis, and hydrolysis.<sup>104</sup> This study concluded glycolysis to be the least energy-intensive process due to its simpler purification process. By contrast, methanolysis showed promising potential but posed purification challenges, while hydrolysis was identified as the most complex and energy-intensive route. Similarly, Uekert *et al.* performed TEA and LCA analyses on mixed feedstock of PET (including PET, polylactic acid, and polybutylene adipate terephthalate) across different depolymerization routes. The findings suggested that methanolysis can outperform glycolysis and hydrolysis both economically and environmentally. This was primarily due to its more efficient downstream separation. Methanolysis generated products with a 31% decrease in selling price and a 21–46% reduction in various environmental impacts including acidification, carcinogenic toxicity, fossil-fuel depletion, global warming potential, particulate formation, and smog formation compared to conventional polyester manufacturing.<sup>105</sup> The successful industrialization of PET chemical recycling requires not only optimized recycling technologies but also integrated waste management infrastructure and the development of economically viable processes.<sup>106</sup>

The design of catalysts for PET glycolysis is also a significant challenge in that efficiency (*i.e.*, the activity of the catalyst) and sustainability (*i.e.*, biodegradability) often involve a trade-off. The highly active catalyst might improve reaction speed and yields but could complicate the operations from a sustainability perspective. Concerning this, Pedrini *et al.* utilized the different ionic liquid catalysts for PET glycolysis to investigate the structure–activity relationships (SAR) for combining high activity with biodegradability. The SAR study revealed that



incorporating lipophilicity into IL-44 lowered the activation energy and enhanced depolymerization.<sup>107</sup> Despite this finding, much more work is needed in this area.

### 3.3. PU

Polyurethanes (PUs) are an important class of synthetic polymer obtained from a polycondensation reaction between diisocyanates and diols or polyols.<sup>108</sup> Their versatile properties allow their applications in coatings, adhesives, sealants, construction, packaging, automobiles and energy harvesting.<sup>109</sup> It is estimated that by 2030, the global PU market will be worth approximately 112.45 billion USD,<sup>110</sup> with production reaching 25.8 million metric tons or 6% of global polymer production.<sup>111</sup> Within the circular economy concept and considering the current need to recycle PUs, several approaches have been designed, assessed, and exploited for recovery and conversion into valuable products. In this section, the strategies for the upcycling and recycling of PU are highlighted, as well as perspectives on the associated challenges and possible future developments.

The mechanical recycling of PU, such as the rebinding approach, involves shredding PU waste and subsequent reprocessing with binders through the continuous molding process to regenerate new PU products.<sup>112</sup> The highly cross-linked structure of PU, however, limits the effectiveness of mechanical recycling techniques for producing high-value materials. Conversely, chemical upcycling offers a more promising route through the cleavage of urethane linkages, allowing the recovery of polyols and other valuable intermediates.<sup>113,114</sup> The chemical methods used for PU depolymerization into desired products are summarized in Table 4.

Liu *et al.* reported the fast and solvent-free acidolysis of toluene diisocyanate (TDI)-based flexible polyurethane foam (PUF) within 10 min at 200 °C, using maleic acid, with a 95% recovery of the recycled polyols.<sup>117</sup> Beyond conventional methods that primarily focus on breaking down PU into polyols to regenerate new PU, the integrated chemolysis approach has also been a focus in recent years. For instance, Sun *et al.* combined methanolysis and hydrogenation with a CO<sub>2</sub>/H<sub>2</sub> reaction medium, using the heterogeneous catalytic system (*i.e.*, ZnO/ZrO<sub>2</sub>/Cu at 200 °C), which resulted in the conversion of PU into intermediates including aromatic diamines and lactones. These valuable intermediates were further utilized to synthesize polyimide (for advanced engineering applications) and polylactone (as a biodegradable alternative to traditional plastics).<sup>115</sup> Additionally, another strategy by Bech *et al.* integrated chemolysis by utilizing combined acidolysis and hydrolysis for PU foam. This enabled the simultaneous recovery of polyol and aromatic diamines with efficient separation.<sup>118</sup> In that work, the succinic acid was identified as an optimal reagent that formed readily isolable succinimide intermediates, which were valorized to toluene diamine *via* simple alkaline hydrolysis. This method achieved up to 83 wt% recovery and showed applicability to both flexible and rigid PU foams.

Typically, in the chemolysis of PU, the isocyanate component, a highly valuable fraction of PU, is often lost during

depolymerization. In order to recover this part, Zahedifar *et al.* proposed a multistage chemolysis approach in which glycolysis was conducted to depolymerize PU, generating carbamate and carbamate oligomers as precipitated intermediates.<sup>119</sup> The intermediates were subsequently hydrolyzed at 200 °C, in order to regain the original isocyanate fraction into corresponding amine compounds, with an amine production yield of about 30% of the model system.

Alongside different chemolysis conversion and recovery strategies for PU, improvements to energy efficiency and reaction times have been the focus of researchers as well. In this regard, microwave-assisted glycolysis has emerged as an alternative approach for PU depolymerization. Unlike PET, only a few studies have been reported on the microwave irradiation of PU *via* glycolysis. Kiss *et al.* carried out a study to compare atmospheric glycolysis and microwave irradiation-assisted glycolysis, where the latter proved to be a faster process leading to recycled polyol with similar characteristics.<sup>112</sup> Nikje *et al.* further investigated microwave-assisted glycolysis on rigid PU foam using DEG (diethylene glycol) as a solvent with different catalysts including sodium hydroxide, potassium hydroxide, sodium acetate, and zinc acetate under various microwave powers at atmospheric pressure.<sup>120</sup> In another study, Nikje *et al.* investigated the effect of water content in microwave-assisted glycolysis reaction on PU depolymerization efficiency.<sup>121</sup> This study revealed that higher water content reduced the viscosity of the product, thereby enhancing the degree of depolymerization. Furthermore, Nikje *et al.* also optimized the effect of operative temperature under microwave-assisted glycolysis to reduce the reaction time by up to 30 min to recover polyols to regenerate PU.<sup>122</sup> Donadini *et al.* also examined the microwave-assisted glycolysis using potassium acetate and stannous octoate as catalysts to recycle rigid PU, resulting in a reduction of reaction time and energy consumption of 94% and 45%, respectively, as compared to conventional heating.<sup>123</sup>

Among the different chemolytic pathways, glycolysis has been extensively used and developed for adaptation at a pilot scale (*e.g.*, Lyntal International Inc.).<sup>124</sup> Notably, major players in the global plastics industry, including Evonik, REMONDIS, Covestro, and Dow, have introduced integrated PU production and recycling initiatives aimed at enhancing sustainability, such as PReSmart (Covestro) and RENUVA (Dow); both initiatives are implemented in the EU market to facilitate the PU-based circular economy.<sup>117</sup>

**3.3.1. Challenges to PU chemolysis.** The chemical upcycling and recycling of PU encounter substantial challenges that must be overcome for the successful future implementation of the technology. Besides the key challenge—the stability of the urethane linkages and the presence of the urethane groups within the polymer backbone hinder depolymerization performance—the presence of various additives (*i.e.*, flame retardants, plasticizers, fillers, *etc.*) complicates the advancement of suitable catalysts. The presence of these additives also complicates purification and separation processes. Different chemolytic processes inherently have unique disadvantages that must be considered. For example, hydrolysis requires high temperatures and long reaction times, leading to increased energy



consumption. Aminolysis, though effective at depolymerization into valuable products, raises concerns related to the safe handling of amine reagents. The curing histories of PU coating play a critical role in their recyclability as well. This was demonstrated by Sánchez-Cadena *et al.* using the single solvolysis system: KOH-catalyzed glycolysis with diethylene glycol (DEG) to investigate UV and electron beam (EB)-cured polyacrylate urethane coatings.<sup>125</sup> The UV-cured coatings, with lower crosslink density, degraded fully at 200 °C in 2 h, while EB-cured coatings required 230 °C for 5 h. This highlights the need to consider both chemical structure and curing methods when designing efficient recycling strategies for PU coatings. Additionally, removal of the PU-based tie layers, such as within complex multi-packaging films, further complicates the recovery and recycling process. This requires further exploration.

### 3.4. PS

For PS upcycling, two principal strategies are typically reported: chemical recycling using catalytic depolymerization and feedstock conversion *via* thermochemical decomposition (*e.g.*, pyrolysis). There exist other upcycling and recycling methods, such as photodegradation, enzymatic degradation, microwave-assisted degradation, and mechanical degradation.<sup>126</sup> However, these have been less studied. Regardless of the strategy employed, upcycling into valuable products is associated with several universal challenges specific to PS. One of the major challenges is its inherent low density-to-volume ratio, which makes the collection, storage and transport of PS waste economically challenging. This results in a lack of investment in infrastructure, compaction equipment, and logistical systems, which hinders the development of PS upcycling at an industrial scale.<sup>127</sup> In addition, the presence of stable phenyl groups and single C–C bonds in the PS backbone makes its depolymerization an energy-intensive process.

The most common approach used for the depolymerization of PS is direct thermal pyrolysis. Advancements have been made with the technology regarding the thermal degradation of PS including improved reactor designs. Still, some challenges with the pyrolysis of PS persist, such as the production of aromatic products with low thermal-oxidative stability, which can result in carbon formation issues.<sup>128</sup> One of the potential solutions to this issue is the use of a catalyst (*e.g.*, zeolites, alumina, silica) to tune product selectivity.<sup>129,130</sup> Kim *et al.* utilized the partial oxidation treatment of PS at 382 °C under supercritical water. This approach provided high selectivity for styrene recovery while minimizing coke formation.<sup>131</sup> In addition, Pinsuwan *et al.* focused on the efficiency of the nitric acid oxidation reaction of PS; the reaction at 170 °C for 4 h resulted in 42% carbon recovery.<sup>132</sup> Zayoud *et al.* explored the pyrolysis of PS in CSTR (continuous stirred tank reactor) at 450–660 °C and 0.02–1 bar pressure, resulting in 56% styrene yield.<sup>133</sup> Inayat *et al.* used pyrolysis using MgO catalyst at 400 °C for 60 min, resulting in a higher amount of oil with higher selectivity for styrene monomer.<sup>134</sup> Dement'ev *et al.* explored zeolite under pyrolysis for PS at 450–550 °C; this resulted in complete PS conversion in

only 1–2 s of residence time.<sup>135</sup> Wang *et al.* developed a mild and selective method for the degradation of PS using visible light and inexpensive iron salts as catalysts under air or oxygen. The process enabled oxidative cleavage of C–C and C–H bonds in PS, yielding benzoic acid as the main product. Remarkably, full PS conversion was achieved under ambient conditions with 63% isolated yield of benzoic acid.<sup>136</sup> Kumar *et al.* reported an efficient, low-cost method for PS depolymerization using table salt and oxidized copper as additives. The process achieved up to 84% styrene yield at 420 °C, significantly higher than the 66% yield from direct pyrolysis. The synergistic effect of NaCl and CuO enhanced radical suppression and heat transfer, improving product selectivity. The recovered styrene was also successfully repolymerized without purification, producing polystyrene with comparable properties to commercial PS. This strategy demonstrates a scalable and environmentally friendly pathway for PS recycling.<sup>137</sup>

The tandem strategy has also been practiced for PS upcycling. This approach comprises two or more sequential reactions to amplify upcycling: degradation of PS to generate intermediates, followed by transformation of the intermediates into high-value products *via* subsequent upcycling reactions.<sup>138</sup> Xu *et al.* utilized tandem upcycling to convert PS into high-value chemicals by using UV light and AlCl<sub>3</sub> (facilitated Friedel–Crafts-type alkylation), depolymerizing PS into aromatic intermediates, followed by upcycling to diphenylmethane with 87% selectivity.<sup>139</sup> For further advancement, more research is needed to clarify the role of different solvents (*i.e.*, alcohols, supercritical *n*-hexane, *etc.*) when converting plastic waste into fuels under HTL.

Microwave-assisted heating for PS has a key advantage over conventional heating methods: internal, homogeneous, and volumetric heating.<sup>140</sup> Kachhadiya *et al.* used a multimode research-grade microwave oven to conduct the pyrolysis of PS using biochar as a susceptor. This yielded up to 60 wt% oil rich in styrene with a heating value of 39–42 MJ kg<sup>−1</sup>.<sup>141</sup> The results also revealed that higher microwave power and lower susceptor improved styrene selectivity by reducing the secondary reactions. Luong *et al.* also used the microwave catalytic approach for PS, using H-ZSM-5 at 400 °C and achieving ~98% conversion yield with 65 wt% of BTX.<sup>142</sup> This study revealed another advantage of microwave-assisted heating: the hotspots at the catalytic sites were induced, which resulted in lowering of the activation energy with increased selectivity for BTX. In another study, Ahmad *et al.* investigated the microwave-assisted copyrolysis of PS with PP using SiC particles as a microwave absorbent.<sup>143</sup> The optimal oil recovery of 93.84 wt% was achieved with a PS/PP weight ratio of 2 : 1 and microwave power of 600 W at 500 °C.

## 4. Upcycling plastic waste to naphtha

Plastic waste, particularly polyolefins, represent a source of carbon and hydrogen that can be converted into fuels and valuable chemicals such as naphtha.<sup>16</sup> Naphtha is a mixture of paraffins, naphthenes, and aromatics ranging from C<sub>5</sub> to C<sub>12</sub> that can be used as a feedstock for manufacturing plastics (*e.g.*,





ethylene).<sup>144</sup> Based on the carbon number, naphtha can be classified into different groups for different applications (Fig. 6). Typically, light naphtha can be used as a solvent for paints and cleaners, or as a component of gasoline. In contrast, medium and heavy naphtha can be used in manufacturing olefins and aromatics.<sup>145</sup> In terms of chemical composition, naphtha is typically comprised of a specific amount of paraffin, naphthene, aromatics, and olefins (Fig. 7).

Plastics-to-naphtha technology holds the potential to replace petroleum-derived naphtha through various thermochemical methods such as pyrolysis, catalytic pyrolysis, and HTL. All can be utilized to convert plastic waste into naphtha, and their state-of-the-art technology is discussed in the following sections.

#### 4.1. Naphtha from non-catalytic conversion of plastic waste

Many non-catalytic conversion methods using pyrolysis, HTL, and solvothermal liquefaction have been used to convert plastic waste into naphtha (Table 5). For example, Hassibi *et al.* used pyrolysis to convert PP waste into naphtha. They found that

lighter naphtha could be obtained by using reflux temperature.<sup>146</sup> However, most literature suggests that pyrolytic oil converted from plastic waste is not suitable for naphtha applications and requires further processing such as hydro-processing or hydro-isomerization. Keeping this in view, Neuner *et al.* coupled pyrolysis with hydro-processing, converting LDPE into oil products of higher-quality naphtha.<sup>147</sup>

In addition to LDPE, non-catalytic pyrolysis has also been used to convert mixed plastic waste into naphtha. For instance, Kusenbergh *et al.* pyrolyzed municipal plastics (including EVOH, PA, PUR, PVC and EVA) and highly contaminated marine litter into oil at 400 °C.<sup>151</sup> Afterwards, distillation and steam cracking were applied to the pyrolytic oil. It was found that integrating pyrolysis, distillation with subsequent steam cracking could effectively valorize highly mixed, contaminated, and difficult-to-recycle plastic waste for naphtha production.<sup>152</sup> To upcycle single-use plastic packaging waste with greater than 40% of polyolefin composition, Genuino *et al.* pyrolyzed DKR-350 (European plastics packaging waste with lower recycling value,

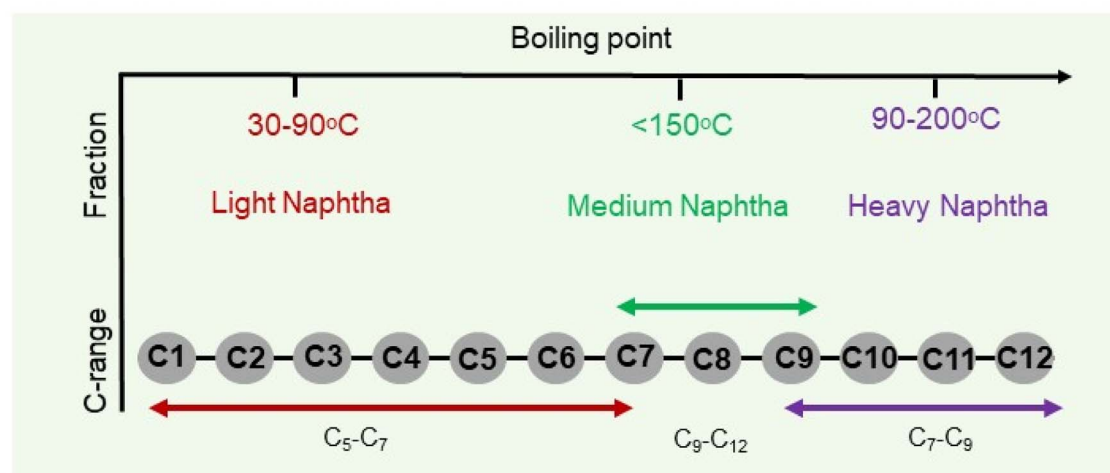


Fig. 6 Fraction of naphtha based on carbon number range and boiling point.

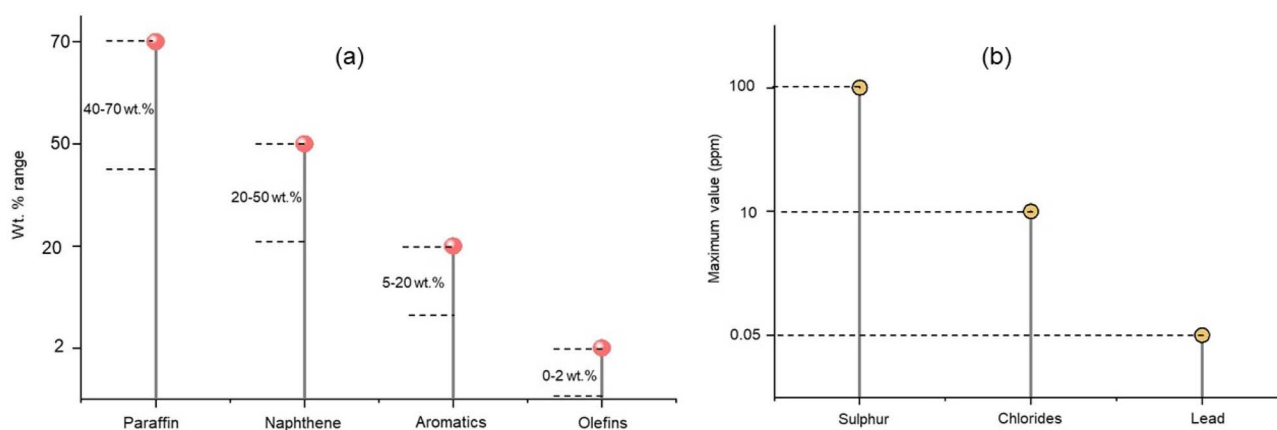


Fig. 7 Typical naphtha specifications for (a) paraffin, naphthene, aromatics and olefins (all in wt%), as well as (b) sulfur, chloride and lead (all in ppm).





mixed composition, high level of contamination, and variable properties) to obtain naphtha. The obtained naphtha was found to have a boiling point of <180 °C, and post-processing of the naphtha for future applications (27% of the sample) from DKR-350 was highly recommended.<sup>153</sup> Lu *et al.* carried out HTL at 450–475 °C with a 2–4 h reaction time to convert pretreated ocean-bound plastic waste into oil. Approximately 66–78% of the oil product recovered shared a similar boiling point distribution to that of naphtha.<sup>16</sup> When the reaction conditions were changed from 450 °C for 4 h to 475 °C for 1 h, the naphtha fraction decreased, likely due to some parts of the naphtha being converted into gaseous products. This finding indicates the importance of optimizing reaction conditions for higher naphtha production. The molar mass of plastic waste can also play an important role in the production of naphtha. Jan *et al.* conducted HTL on linear low-density polyethylene (LLDPE) with supercritical water with varying molar mass (ranging from  $7.20 \times 10^3$  to  $1.29 \times 10^5$  g mol<sup>-1</sup>). They discovered that lower molar mass samples achieved complete conversion at 425 °C for 2 h. Additional works obtaining naphtha through non-catalytic processes are presented below in Table 5.

#### 4.2. Naphtha from catalytic conversion of plastic waste

Catalytic thermochemical conversion methods to recycle plastic waste into naphtha have been investigated to improve naphtha quality, reduce by-products, and enhance process efficiency. This has involved the addition of catalysts to thermochemical

conversion processes including HTL, pyrolysis, and hydrocracking. Table 6 summarizes state-of-the-art catalytic thermochemical conversions of plastic waste.

Dai *et al.* coupled pyrolysis with tandem catalysis reactions to convert polyolefins (HDPE, LDPE and PP) into short chain paraffin rich naphtha. The catalysts used were Zn/SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> pillared montmorillonite clay (M-Clay), which were able to facilitate conversion of the polyolefins into naphtha with minimal aromatic products. The tandem catalysis approach proved to accelerate hydrogenation for producing naphtha with composition in the C<sub>5</sub>–C<sub>12</sub> range, which can be used to manufacture new plastics.<sup>161</sup> In a subsequent study, Dai *et al.* used the montmorillonite clay (M-Clay) for one-step catalytic pyrolysis of pure LDPE and HDPE/LDPE/PP blend. The results showed that the incorporation of Al with M-Clay favored conversion into naphtha, with 60.3% of the naphtha fraction consisting of C<sub>5</sub>–C<sub>12</sub> alkanes.<sup>160</sup> Another study by Dai *et al.* explored an SBA-15 (SiO<sub>2</sub>)-supported metal catalyst for upcycling the same polyolefins into naphtha, with zeolites (USY and ZSM-5) used as reference catalysts. Results indicated that Cs, Fe, Mg, Co, and Cu/SBA-15 did not prove to be active for C–C bond dissociation. However, based on proper acidity and pore structure, Zn/SBA-15 exhibited better activity for naphtha production as compared to pure SBA-15 with aromatics generated.<sup>161</sup> Another catalyst, SAPO-11 (AEL structure) zeolite with 0.3% platinum loading, was reported to increase oil yield with a higher value of naphtha.<sup>147</sup>

Table 5 Producing naphtha from different non-catalytic conversion technologies of plastic waste

Feedstock	Process	Reaction condition	Remarks	Ref.
PE	Hydrothermal liquefaction	425–475 °C 23 MPa	<ul style="list-style-type: none"> <li>From a full conversion reaction, 66% of the converted oil can be used as naphtha</li> <li>The amount of naphtha produced was 7–18% lower than that from PP at full conversion reaction, likely due to different reaction pathways under HTL.</li> </ul>	16
PP	Hydrothermal liquefaction	0.5–4 h 380–500 °C 23 MPa	<ul style="list-style-type: none"> <li>An increase in reaction time (2 to 6 h) results in a higher fraction of naphtha (73 to 94%) with a decrease in heavier fraction of fuels (<i>e.g.</i>, diesel)</li> <li>425 °C, 4 h or 450 °C, 1 h proved to be the optimal reaction condition for the highest naphtha yield (~80 wt%)</li> </ul>	148
PP, PS (packaging)	Thermal cracking	0.5–6 h 510–520 °C 15–30 min	<ul style="list-style-type: none"> <li>Increased PS concentration to the PP increased the aromatic content in the naphtha fraction</li> <li>The octane and motor number of naphtha fraction was increased due to the presence of higher olefins and aromatic content</li> </ul>	149
LDPE, HDPE, PP, PS, PVC, PET (individual polymer and mixture)	Pyrolysis	700 °C 25 °C min <sup>-1</sup>	<ul style="list-style-type: none"> <li>PET oil contained more alcohol and carboxylic acid</li> <li>The boiling point of converted oil was within range of naphtha application and can be utilized in a steam cracker for ethylene production</li> </ul>	150





Table 6 Producing naphtha from different catalytic conversion technologies of plastic waste

Feedstock	Process	Reaction condition	Remarks	Ref.
Tetra Pak (LDPE + Al foil + paper fiber)	Hydrothermal liquefaction	360 °C	<ul style="list-style-type: none"> <li>Naphtha content increased with the addition of catalyst (Ni-xCe/CNTs)</li> </ul>	154
		20 MPa	<ul style="list-style-type: none"> <li>Increased loading of Ce led to an increase in naphtha content whereas diesel oil content decreased</li> </ul>	
		0.5 h		
Tetra Pak (LDPE + Al foil + paper fiber)	Hydrothermal liquefaction	Ni-xCe/CNTs	<ul style="list-style-type: none"> <li>Order of catalytic activity for oil conversion: <math>\text{Ni}_{10}\text{Ce}_5/\gamma\text{-Al}_2\text{O}_3 &gt; \text{Ni}_{10}\text{Ce}_7/\gamma\text{-Al}_2\text{O}_3 &gt; \text{Ni}_{10}\text{Ce}_3/\gamma\text{-Al}_2\text{O}_3 &gt; \text{Ni}_{10}\text{Ce}_1/\gamma\text{-Al}_2\text{O}_3</math></li> </ul>	155
		360 °C	<ul style="list-style-type: none"> <li>Increased gasification reaction rate led to a decrease in naphtha fraction</li> </ul>	
		20 MPa	<ul style="list-style-type: none"> <li><math>\text{Al}_2\text{O}_3</math> and <math>\text{CeO}_2</math> favored the production of diesel over naphtha</li> </ul>	
PP + <i>Prosopis juliflora</i> biomass	Hydrothermal liquefaction	0.5 h		156
		$\text{Ni}_{10}\text{Ce}_5/\gamma\text{-Al}_2\text{O}_3$ (with different Ce loading)		
		320–440 °C		
HDPE	Microwave-assisted pyrolysis coupled with <i>ex situ</i> catalytic reforming	1 h	<ul style="list-style-type: none"> <li>With the addition of catalyst, a decrease in naphthalene oligomers was observed</li> </ul>	157
		Alumina-supported metal-oxides	<ul style="list-style-type: none"> <li>Nb/<math>\text{Al}_2\text{O}_3</math> catalyst can be reused up to 10 times</li> </ul>	
		500 °C	<ul style="list-style-type: none"> <li>High silica zeolite with low Brønsted acidity effectively inhibit the aromatic formation in producing naphtha</li> <li>ZSM-5 (800) did not change after 8 regeneration cycles</li> </ul>	
HDPE/vacuum gas oil blend	Hydrocracking	ZSM-5 (25)		158
		ZSM-5 (800)		
		ZSM-5 (280)		
HDPE	Microwave-assisted pyrolysis	ZSM-5 (1500) $\text{SiO}_2/\text{Al}_2\text{O}_3$	<ul style="list-style-type: none"> <li><math>\text{H}_2</math> pressure proved to be an important parameter to control the quality of naphtha</li> <li>Pressure has a direct relation with the selectivity of fuel index (naphtha)</li> </ul>	159
		440 °C		
		$\text{H}_2$		
PP	Pyrolysis	2–10 MPa	<ul style="list-style-type: none"> <li>Higher gas yield was obtained</li> </ul>	160
		PtPd/HY	<ul style="list-style-type: none"> <li>Addition of ZSM-5 was not suitable for producing naphtha</li> </ul>	
		500–740 °C	<ul style="list-style-type: none"> <li><math>\text{Al}_2\text{O}_3</math> led to a more selective naphtha composition, composed of 60.3% <math>\text{C}_5\text{--C}_{12}</math> alkanes</li> <li>ZSM-5 produced higher aromatics</li> </ul>	
LDPE	Pyrolysis	ZSM-5		160
		450–650 °C		
		Kaolin		
Mixture (41% LDPE, 24% HDPE, 35% PP)	Pyrolysis	M-Clay	<ul style="list-style-type: none"> <li>Advanced distillation was required to upgrade the liquid oil for naphtha applications</li> </ul>	160
		$\text{Al}_2\text{O}_3$ pillared M-Clay		
		Zeolite		



Table 6 (Contd.)

Feedstock	Process	Reaction condition	Remarks	Ref.
HDPE	Microwave-assisted pyrolysis	500 °C	• HDPE and LDPE showed similar product selectivity for lower aromatic naphtha application	161
LDPE		Metals/SBA-15	• ZnO incorporation with SBA-15 increased the catalyst acidity, which in turn improved the cracking activity of catalyst	
PP		USY		
Mixture (41% LDPE, 24% HDPE, 35 wt% PP)		ZSM-5		
PE	Pyrolysis	H $\beta$	• The addition of acidic support to H $\beta$ (bifunctional) promoted naphtha production	162
		HY	• The Pd impregnation changed the distribution of naphtha significantly	
LDPE		HZSM-5		
HDPE	Hydrocracking	375–450 °C 1 h	• PP generated more naphtha than PE	163
			• The use of catalyst did not affect the amount of naphtha but changed the distribution of carbon numbers	
PP		H <sub>2</sub> – 5 MPa Bifunctional DHC-8 HYDROBON (18% NiO on Al <sub>2</sub> O <sub>3</sub> ) DHC-8/HYD		
Fischer-Tropsch wax LDPE	Pyrolysis Hydro-processing	SAPO-11 CoMo	• Noble metal catalyst reduced the formation of aromatics	147
Mixed plastics feed (HDPE, PP, PS)	Catalytic hydrocracking	CoMo/Al <sub>2</sub> O <sub>3</sub> 370–430 °C 0.5 h 7 MPa Akzo KC 2600	• Better selectivity of naphtha achieved at 370 °C	164

Table 7 Factors affecting the distribution and production of naphtha converted from plastics

Factor	Influence on naphtha fraction
Use of catalyst	Catalysts with large surface area, good dispersion, more Brønsted acidic sites, and stable pore size with higher adsorption capacity can lead to higher formation of valuable naphtha
Temperature/time	Higher reaction temperature and time both favor the formation of naphtha, but optimized temperature and time are suggested for naphtha generation
Feedstock type	Mixed plastic waste is difficult for naphtha conversion
Operating pressure	High pressure can accelerate naphtha production but results in higher capital cost
Solvent	Organic solvents with lower critical temperature and pressure favor naphtha production but will increase the cost of processing and separation
Contamination	Contaminants ( <i>e.g.</i> , sulfur, nitrogen) should be removed; otherwise, they will poison catalysts

Wang *et al.* processed Tetra Pak waste (a pharmaceutical and food packaging composed of paper, LLDPE, and Al foil) *via* catalytic HTL for naphtha production. Using a bimetallic Ni-Ce<sub>x</sub>/γ-Al<sub>2</sub>O<sub>3</sub> catalyst, naphtha production was accelerated. In contrast, a slight decrease in naphtha production was observed due to gasification under HTL.<sup>155</sup> In another similar study processing Tetra Pak waste *via* HTL (360 °C, 22 MPa, 0.5 h) with Ni-Ce/CNT catalyst for naphtha production, an increased yield was observed with an increased loading of Ce. However, GC-MS analysis showed that the bimetallic catalyst produced a higher hydrocarbon fraction than the monometallic catalyst through a swift shift between Ce<sup>3+</sup> and Ce<sup>4+</sup>. It was also found that deoxygenation and decarbonylation played key roles in facilitating the production of naphtha.<sup>154</sup> More specifically, Ni-xCe/CNTs catalysts with high loading of Ce were able to maximize naphtha yield.<sup>154</sup>

Typically, water (supercritical or subcritical) is used as the reaction medium under HTL. However, recently organic solvents (*e.g.*, toluene, alcohol, acetone) have also been used (*i.e.*, solvothermal liquefaction). For example, Banivaheb *et al.* solvothermally converted a plastic mixture (LLDPE, PP, PS, PU) with toluene and Pd/C catalyst in hydrogen at 350 °C for 1–5 h. The Pd/C catalyst was found to effectively promote hydrodenitrogenation and hydrodeoxygenation.<sup>165</sup> A 5% Pd/C catalyst loading and increased hydrogen pressures were also found to favor higher naphtha production.

The catalysts utilized for high naphtha formation should retain high stability and be capable of reuse after recovery. For these purposes, the microwave-assisted pyrolysis with *ex situ* catalytic reforming of HDPE was studied by Dai *et al.* at different ratios of SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> to ZSM-5 zeolite catalyst. The catalyst was reused eight times in this study. The high silica ZSM-5 zeolites (ZSM-5 (800) and ZSM-5 (1500)) showed a reduced aromatic selectivity in the naphtha fraction.<sup>166</sup>

#### 4.3. Factors influencing the conversion of plastic waste to naphtha

The quantity and quality of naphtha obtained from plastic waste conversion methods are greatly influenced by reaction

parameters. The composition of the feedstock as well as the contamination associated with it also have a large impact on the naphtha obtained. Table 7 below provides an overview of the various factors and how they specifically affect naphtha production.

A promising catalyst for naphtha formation must be developed considering the following characteristics: surface area, pore size, channel size, adsorption capacity, and thermal/electrical properties. Related to this, the quantity and quality of naphtha can be tuned by optimizing reaction conditions and the type of solvents/catalysts used. Particularly, catalysts play a vital role in controlling the formation and quality of naphtha. For example, metal-based catalysts in general are more prone to coke formation, reducing naphtha yields. In such cases, the use of a promoter and support modifier is recommended for catalyst stability and activity.<sup>167,168</sup> Despite this, heterogeneous catalysts are often preferred based on their higher rate of hydrodeoxygenation reaction, catalytic efficiency, ease of recyclability, and lower cost. Catalysts that exhibit higher dispersion and interaction tend to promote enhanced hydrogenation of aromatic compounds for higher-quality naphtha. It has also been found that the acidity of the catalyst is positively correlated with higher rates of hydrogenolysis of C–O bonds for producing high-value naphtha.<sup>169</sup> However, excessively high acidity also promoted the formation of cokes, deteriorating the catalyst activity.<sup>13,170,171</sup>

A lower cost clay-based material (Kaolin, montmorillonite clay, Al<sub>2</sub>O<sub>3</sub> pillared M-Clay) has been explored for converting polyolefins into lower carbon synthetic naphtha. The lower pore volume of Kaolin (0.105 cm<sup>3</sup> g<sup>−1</sup>) with the incorporation of Al to M-Clay enhanced catalyst acidity and lowered the reaction temperature (381 °C) at 50% conversion of LDPE. A different study identified that Ni-Ce<sub>x</sub>/γ-Al<sub>2</sub>O<sub>3</sub> catalyst promoted the glycosidic bond of plastic packaging waste while increased loading of Ce aided in generating more oxygen vacancies. These oxygen vacancies acted as precursors to promote deoxidization and hydrogenation for higher quality naphtha at larger quantities.<sup>155</sup> Furthermore, it has been demonstrated that the oxidative calcination approach can effectively regenerate



deactivated catalyst during microwave-assisted pyrolysis for naphtha conversion, with the re-activated catalyst retaining most of the pore structure and acidity of the original.<sup>166</sup> The promising results, including those outlined previously, provide valuable insights into future catalyst design works. Catalysts with larger pore size, more active sites, or more Brønsted acid sites seem to favor naphtha production from plastic waste and should be targeted.<sup>160</sup> The lifetime of the catalyst should also be extended by making acidic sites more accessible while coke formation should be inhibited as much as possible.

Temperature in thermochemical conversion of plastics to naphtha has a pronounced effect. For instance, a study showed that when the pyrolysis temperature was increased from 350 °C to 390 °C, the amount of naphtha increased significantly while the fraction of heavy residues decreased.<sup>147</sup> In another study, when the temperature was raised from 320 °C to 520 °C, the product distribution became narrower with a greater shift toward the naphtha range of hydrocarbons centering at C<sub>8</sub>. However, higher temperatures do not always lead to better results. For example, in a catalytic conversion study, the temperature was reduced from 500 °C to 460 °C because higher monocyclic aromatics were obtained at higher temperatures. These aromatic components are undesirable as they can be converted into coke, which covers the active sites of any catalyst during catalytic cracking of naphtha.<sup>166</sup>

Solvent selection for thermochemical recycling of plastics into naphtha also plays a crucial role. Besides water (the most commonly used solvent) in HTL, other oxygenated solvents (*i.e.*, methanol, ethanol, acetone) have also been explored in the solvothermal conversion of plastics.<sup>165</sup> By using organic solvents, solvothermal conversion has the advantage of lower reaction conditions as compared to HTL. For instance, toluene, which possesses a lower critical temperature and pressure than water, can lessen the reaction severity required for conversion.<sup>172,173</sup> However, it is suggested that solvents with higher oxygen content be avoided for conversion into naphtha. This is not only because the presence of oxygen increases the amount of impurities in oil recovered but also leads to higher expenses in processing and separation. Impurities found in untreated pyrolytic oil containing naphtha require removal. Otherwise, they can poison any catalysts, especially in the case of noble metal and zeolite catalysts. Effective pre-treatment of plastic waste, including mechanical sorting, washing, and shredding to remove physical impurities (labels, food residues, and adhesives), is therefore essential.<sup>174,175</sup> For oxygen removal, different processes such as decarboxylation and hydrodeoxygenation have been explored.<sup>167,176</sup> The effect of other process parameters in the solvothermal liquefaction area, including reaction time and pressure, has yet to be evaluated in detail and should be explored in the future.

## 5. Upcycling plastic waste to syngas

Gasification is an environmentally friendly thermochemical conversion technology for upcycling of plastic wastes into valuable CO/H<sub>2</sub> mixtures (*i.e.*, syngas).<sup>177</sup> Along with syngas, byproducts such as char and tar are also produced.<sup>178</sup> Syngas

can be directly incinerated to produce energy or can be used to produce gasoline and diesel fuels *via* Fischer–Tropsch synthesis.<sup>179</sup> The gasification of plastic wastes typically occurs at high temperatures (*e.g.*, ≥800 °C) and atmospheric pressure, frequently with the use of catalysts.<sup>180</sup>

The gasification process involves several complex reactions, mainly including drying, pyrolysis, cracking, and reforming.<sup>178</sup> Based on the shape and composition of plastic wastes, gasification reactors can be divided into fixed-bed and fluidized-bed reactors.<sup>177</sup> Gasification offers several advantages.<sup>181</sup> For example, it can be effectively applied to generate power from various waste-derived feedstocks and results in lower environmental and air pollution compared to incineration. The gasification system can provide a simple operation in terms of feedstock flexibility and operation cost.<sup>181</sup> It imposes no limitations on the type and size of the plastic wastes.<sup>182</sup> Additionally, it offers wide flexibility in gasifying agent (*e.g.*, oxygen, steam) and adaptability to tailor the composition of the syngas product.<sup>180</sup> Moreover, plasma gasification is able to destroy some toxic components (*e.g.*, furans, dioxins) in the waste plastic as compared to conventional incineration.<sup>180</sup> Gasification also produces a cleaner gas compared to combustion and pyrolysis.<sup>183</sup>

However, gasification has some limitations. It is sensitive to contaminants or impurities, which can deactivate the catalysts.<sup>180</sup> Prior to gasification, the feedstock is typically pre-treated through mechanical sorting, washing/leaching, drying, and shredding to eliminate physical debris and reduce soluble impurities such as alkali metals, chlorine, and sulfur.<sup>174,184,185</sup> Additional thermal pre-treatments such as torrefaction or low-temperature pyrolysis may be applied to further improve feed uniformity and lower contaminant concentrations.<sup>174</sup> In addition, tar can be produced and block reactor pipes during gasification.<sup>180</sup> Catalysts play a critical role in the production of syngas *via* gasification.<sup>186</sup> The active phases of catalysts can be divided into precious and non-precious metals. For example, Wu *et al.* developed a nickel core and ceria shell-based catalyst for waste plastic gasification. The core–shell structure effectively inhibits the metal sintering, which improves the catalytic stability.<sup>186</sup> Catalysts also reduce tar formation and can lower both the operating temperature and overall process cost compared to non-catalytic gasification.<sup>178</sup> The quality of syngas depends on operating conditions in the reactor, gasifying agent types, and catalyst activity.<sup>177</sup> Highly active catalysts are essential for efficient tar and methane reforming, should be easily regenerable, and should reduce energy input to meet industrial demands.<sup>177</sup>

### 5.1. Syngas from PE and PP

Pyrolysis and gasification are two major thermochemical routes for the conversion of PE and PP into syngas and other hydrocarbon fuels. Both catalytic and non-catalytic pyrolysis primarily yield hydrocarbons,<sup>187–189</sup> whereas the gasification process can be tuned to produce syngas along with hydrocarbons.<sup>181</sup> The H<sub>2</sub> and CO composition of syngas obtained from the gasification of PE and PP depends on a variety of parameters including, but not





limited to, reactor type, gasifying agent, temperature, equivalence ratio, residence time, and catalyst formulation.

The profound technical knowledge of fluidized-bed reactors gained from decades of biomass conversion studies prompted them to be widely used in the gasification of plastics.<sup>55,190–202</sup> For example, Arena *et al.* utilized bubbling fluidized bed reactors at 850 °C to gasify PE on quartz sand and olivine (magnesium iron silicate, (Mg, Fe)<sub>2</sub>SiO<sub>4</sub>) beds. They found that compared to the inert quartz sand, the catalytically active olivine bed led to higher H<sub>2</sub> and CO formation as well as reduced tar generation.<sup>190</sup> Sancho *et al.*<sup>199</sup> investigated both dolomite (calcium magnesium carbonate, CaMg(CO<sub>3</sub>)<sub>2</sub>) and olivine as catalytic bed materials in fluidized bed gasification of PP and concluded that while dolomite provides better quality syngas with lower tar than olivine, the latter is more attrition-resistant, leading to fewer particulate impurities in the product gas. Mechanical strength and resistance towards attrition are therefore important criteria for fluidized bed catalysts. A dual bed gasifier system with two stages at 600 °C and 800 °C was utilized by Yang *et al.* for air gasification of PE and PP to simultaneously produce syngas and carbon nanotubes. They used Ni/Al-SBA-15 catalysts produced by the wet impregnation method and polyol process in both gasifier stages; the polyol process synthesized catalyst with a Si/Al ratio of 10 was found to be the best catalyst for carbon nanotube and syngas formation. The increased surface acidity upon Al incorporation into these catalysts was posited to be the primary reason for their enhanced activity.<sup>55,202</sup>

Although fluidized-bed reactors have seen widespread application in plastic gasification, a major fraction of these systems are non-catalytic. On the other hand, fixed bed reactors are mostly used with a catalyst for PE and PP conversion.<sup>203–210</sup> Supported Ni-based catalysts are primarily used for this purpose due to their activity in gasification, tar removal, methane reforming, and water gas shift reaction that helps adjust the H<sub>2</sub>/CO ratio in product. He *et al.* studied catalytic steam gasification of PE over a NiO/γ-Al<sub>2</sub>O<sub>3</sub> catalyst in the temperature range of 700–900 °C that produced syngas at a H<sub>2</sub>/CO ratio of 0.83–1.35, a suitable feedstock for Fischer–Tropsch process.<sup>206</sup> Several Ni-based catalysts such as Ni/Al<sub>2</sub>O<sub>3</sub>, Ni/MgO, Ni/CeO<sub>2</sub>, Ni/ZSM-5, Ni–Al, Ni–Mg–Al, and Ni/CeO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> were investigated by Wu *et al.* in a two-stage pyrolysis-gasification of PP where the first stage was non-catalytic pyrolysis at 500 °C and the downstream second stage was catalytic gasification at 800 °C. Their study found Ni–Al (1 : 2), Ni–Mg–Al (1 : 1 : 2), and Ni/ZSM-5 to be great catalysts for syngas production with low coke formation, with Ni/MgO being the least active for this application. Other authors also investigated Ni-based catalysts such as Ni–Mg–Al–CaO, NiO/dolomite, and Ni–CaO–C for syngas production from PE and PP.<sup>204,205,208,210</sup> The formation of coke and carbon fibers on the active Ni-sites as well as the support has been reported as the main reason for catalyst deactivation in these studies.

Mixed gasification of PE and PP with various plastics, biomass and other municipal solid wastes has been shown to produce better quality syngas with higher gas formation and lower tar and char formation.<sup>191,192,196–198</sup> For example, Ruoppolo *et al.* reported enhanced H<sub>2</sub> formation from a mixture of PE and

wood pellets with NiO/γ-Al<sub>2</sub>O<sub>3</sub> catalyst in a fluidized-bed reactor.<sup>198</sup> Friengfung *et al.*<sup>205</sup> studied various combinations of PP, LDPE, HDPE, PS to investigate co-gasification behavior with Ni/dolomite catalyst in a drop-tube fixed-bed reactor.<sup>37</sup> The addition of HDPE or LDPE to PP decreased the H<sub>2</sub> and CO yields, whereas PS addition to PP at 0.5–0.75 wt% led to a significant increase in syngas formation. A synergistic interaction between the different plastics and their gaseous products is thought to be the driver behind such improvement in syngas production from co-gasification.

Operating parameters such as temperature, gasifying agent, and equivalence ratio have a significant impact on the product yield from plastic gasification. The endothermic gasification process makes it highly susceptible to temperature changes. Toledo *et al.* investigated the gasification of PP on an olivine catalyst in a fluidized bed reactor to show that an increase in temperature from 750 °C to 910 °C leads to higher H<sub>2</sub> and CO formation.<sup>200</sup> He *et al.* performed steam gasification of PP on NiO/γ-Al<sub>2</sub>O<sub>3</sub> catalyst in a fixed bed in the temperature range of 700–900 °C.<sup>206</sup> They reported that an increase in the operating temperature increased syngas formation, H<sub>2</sub>/CO ratio, and decreased char and coke formation thereby giving better long-term stability to the catalyst. The type of gasifying agent is another important variable. Researchers have used air, oxygen, steam, CO<sub>2</sub>, and their mixtures as gasifying agents; among them, steam and oxygen have been found to lead to better H<sub>2</sub> and syngas production.<sup>190,193,196,198</sup> The equivalence ratio and steam/fuel ratio can alter plastic conversion and product distribution. Toledo *et al.* showed that a decrease in the equivalence ratio in air gasification of PE improves gaseous product yield and their calorific value.<sup>200</sup> Simultaneous optimization of all these operating variables is required to maximize syngas production from PE and PP and minimize the formation of undesired by-products such as tar and coke.

Recently, plasma-assisted plastic gasification using high temperatures—by microwave, electric arc plasma, or radio-frequency—presents a promising solution.<sup>211</sup> For instance, Fathi *et al.* carried out the high-power microwave plasma gasification (reactor temperature ~1400 °C) of PP into syngas with a maximum hydrogen yield of 19.7% with 60% energy efficiency.<sup>212</sup> Additionally, the by-product, carbon black nanoparticles, were recovered and upcycled into titanium carbides to promote a circular approach. In another study, microwave plasma gasification was explored by Tamošiūnas *et al.* for medical waste (PP and PE) using CO<sub>2</sub> as a gasifying agent. The results showed a high syngas yield of 3.13 Nm<sup>3</sup> kg<sup>−1</sup> with a syngas heating value of 13.88 MJ Nm<sup>−3</sup>, where H<sub>2</sub> and CO were the main components (~80.46 vol%).<sup>213</sup> The co-gasification approach has been practiced as well to enhance syngas production. For example, Mukeru *et al.* found that co-gasification of PE with glycerol resulted in increased syngas yield, hydrogen production, and cold gas efficiency (~64%) through synergistic effects between the two feedstocks.<sup>214</sup>

Contaminants originating from additives, pigments, and halogenated polymers in both PE and PP waste streams pose challenges for stable and efficient gasification. For example, chlorine from residual PVC releases HCl during thermal



decomposition, which can corrode reactor internals and poison downstream catalysts.<sup>215</sup> Nitrogen-containing polymers (e.g., PUR, PS) and sulfur-bearing additives may form  $\text{NH}_3$  and  $\text{H}_2\text{S}$ , which deactivate metal catalysts and contribute to acid gas emissions.<sup>216</sup> Pre-treatment strategies such as mechanical sorting, solvent washing, and blending with low-contaminant feedstocks are commonly employed to reduce halogen and metal contents.<sup>217</sup> In addition, in-bed sorbents like dolomite and  $\text{CaCO}_3$  are used to capture  $\text{HCl}$  and  $\text{SO}_2$  *in situ*, thereby reducing corrosion and enabling cleaner syngas production.

Catalyst performance is also sensitive to contaminants. Ni-based catalysts (e.g.,  $\text{Ni}/\text{Al}_2\text{O}_3$ ,  $\text{Ni-Mg-Al}$ ,  $\text{Ni-ZSM-5}$ ) are widely used in PE and PP gasification due to their high activity in tar reforming and methane conversion. However, prolonged exposure to chlorine, sulfur, or heavy metals can lead to deactivation *via* sintering, carbon deposition, or active site poisoning.<sup>218</sup> Advanced catalyst designs, such as Ni-core/ $\text{CeO}_2$ -shell or Zr-doped supports, have been shown to enhance thermal stability and coke resistance.

Post-gasification syngas cleaning is essential to meet the purity requirements for Fischer-Tropsch or methanol synthesis. Scrubbers and guard beds are used to remove acid gases (e.g.,  $\text{HCl}$ ,  $\text{H}_2\text{S}$ ), while particulate filters and tar crackers ensure long-term catalyst protection in upgrading reactors.<sup>219</sup> The contaminant profile can vary significantly depending on the source and composition of the plastic feedstock; thus, integrated pre-treatment, reactor design, and gas cleanup strategies are essential for robust and scalable syngas production from PE and PP waste.

## 5.2. Syngas from PET

PET, making up roughly 7% of all plastics consumed globally, is one of the most often utilized polymers.<sup>220</sup> According to recent data, the global PET market is projected to reach approximately USD 52.94 billion in 2024 and grow to USD 109.63 billion by 2032, reflecting a compound annual growth rate (CAGR) of 9.5% during the forecast period.<sup>221</sup> This growth is driven by increasing demand for lightweight, durable, and recyclable packaging materials across various industries, including food and beverage, pharmaceuticals, and personal care. PET was initially used in textiles, films, video, photography, and X-ray films and cassettes. Recently, because of its glass-like clarity, sufficient gas barrier for carbonation retention, lightweight characteristics, and high capacity, PET is increasingly utilized for food and beverage containers.<sup>222</sup> Even though transparent PET bottles are highly recyclable into fibers, bottles, amorphous PET sheets, strapping tape, and other products, the recycling rate of post-consumer PET, including opaque PET bottles, PET trays and films still falls short of the pace of PET consumption due to the broad range of colors, additives, multilayer structure, labels, and other complexities.<sup>223</sup>

Identifying a new processing method of PET is essential but not easy. Bioremediation of PET remains challenging, as the ester bonds within its polymer chain are inherently resistant to degradation under ambient (room temperature) conditions. Additionally, PET is considered a challenging plastic for

thermochemical recyclers due to its high yield of benzoic acid during thermal deconstruction, which adversely affects the quality of pyrolysis oil. Researchers compared the pyrolysis products from PP, LDPE, and PET and found that PET, compared with PP and LDPE, tends to produce more tar, which has a mostly aromatic structure.<sup>224</sup> Therefore, high temperature pyrolysis/gasification of PET for high-quality syngas production has been considered an effective pathway to recover energy since the obtained syngas can be directly used for energy, fuels, or chemicals. It was demonstrated that the gas product from PET steam gasification at 750–800 °C was mainly composed of  $\text{H}_2$ ,  $\text{CO}$ , and  $\text{CO}_2$ . Among the gaseous products,  $\text{CO}_2$  was dominant due to the large number of carboxyl groups in the PET chain.<sup>225</sup> Increasing temperature can notably improve the yields of  $\text{H}_2$  and carbon conversion to gaseous products with  $\text{CO}_2$  as the dominant gas product. However,  $\text{CO}_2$ -assisted gasification at 800–950 °C effectively increased the  $\text{CO}$  yield to a higher level of 0.5–0.9  $\text{g g}^{-1}$ , which accounted for about 90 wt% of the total syngas yield.<sup>226</sup> It was estimated that 0.1–0.7  $\text{g CO}_2$  was utilized for each gram of PET gasified. This process offers a robust solution in converting PET to alternative syngas fuels along with  $\text{CO}_2$  utilization.

To improve the quality of syngas generated from PET, co-pyrolysis has been the most common method adopted. Combining PET with PP and/or biomass is conducive to syngas production.<sup>227</sup> For example, Ahiekpor *et al.* found that the optimal syngas yield (68.1 wt%) was achieved at 800 °C with a nitrogen flow rate of 6  $\text{NL min}^{-1}$ , using a feedstock composed of 50 wt% sawdust and 50 wt% plastic.<sup>228</sup> Nevertheless, reaction conditions need additional study for better syngas yield in co-pyrolysis of PET with biomass. Apart from co-pyrolysis, there are still some positive studies for producing syngas from PET with other methods. Wang *et al.* found that using  $\text{CO}_2$  as the gasifying agent for syngas production from PET can be a reliable way to turn waste PET into alternative syngas fuels and address the problem of greenhouse gas emissions at the same time. They studied the gasification of PET in  $\text{CO}_2$  by using TGA and a fixed bed reactor. The results showed that  $\text{CO}$  was the dominant component, comprising over 73.3 vol% of the total syngas, with each unit mass of PET consuming 0.1–0.7 mass unit of  $\text{CO}_2$  depending on the reaction temperature.<sup>226</sup> Xu *et al.* also applied photocatalysis using  $\text{Co-Ga}_2\text{O}_3$  as catalyst to realize the plastic-to-syngas conversion at ambient conditions. As a result, commercial plastic items including PE bags, PP boxes, and PET bottles could be effectively photodegraded into  $\text{CO}$ , whereas  $\text{Co-Ga}_2\text{O}_3$  nanosheets could photo reduce  $\text{H}_2\text{O}$  into  $\text{H}_2$ .<sup>229</sup> Additionally, Wang *et al.* co-pyrolyzed cellulose and PET (sourced from waste textiles) to enhance syngas production through synergistic effects. This approach yielded a maximum fuel gas output of 13.9  $\text{mmol g}^{-1}$  at 850 °C, which further increased to 20.0  $\text{mmol g}^{-1}$  with the use of an  $\text{MgO-CaO}$  catalyst derived from oyster shells.<sup>230</sup> The catalyst not only enhanced the syngas yield, but also reduced the tar formation by promoting the cracking of volatiles. This approach provides a sustainable strategy for mixed waste conversion, utilizing biomass-derived catalysts to valorize PET waste into syngas.



### 5.3. Syngas from PS

Syngas yields and quality from catalytic gasification of PS were enhanced compared to non-catalytic conditions. Liu *et al.* investigated the catalytic gasification of PS in CO<sub>2</sub> over Ni-Mg/Al<sub>2</sub>O<sub>3</sub> in a fixed-bed reactor at 900 °C.<sup>231</sup> A quasi *in situ* strategy was used, in which the catalyst was placed downstream of the feedstock in the same gasification reactor. Catalytic gasification of PS resulted in a 22% increase in syngas yield compared to non-catalytic conditions. The results also showed enhanced yields of 63% H<sub>2</sub>, 20% CO, 119% CH<sub>4</sub>, and 85% C<sub>2</sub>–C<sub>3</sub> yields. Additionally, cold gas efficiency improved from 44% to 57%, and syngas composition analysis showed enhanced quality, as indicated by a higher H<sub>2</sub> mole fraction and a reduced CO fraction. In a related study, Choi *et al.* investigated the catalytic pyrolysis of PS using CO<sub>2</sub> and Ni-based catalysts (2/5/10 wt% Ni/SiO<sub>2</sub>). They observed that H<sub>2</sub> yield from catalytic pyrolysis was over an order of magnitude higher than in the non-catalytic process, with H<sub>2</sub> production increasing proportionally with catalyst loading.<sup>232</sup>

Process temperature significantly influences the yields of syngas, hydrogen, and energy. Ahmed *et al.* studied the pyrolysis and gasification of PS in a semi-batch reactor at 700, 800, and 900 °C.<sup>233</sup> At 700 °C, gasification produced lower yields than pyrolysis; however, the trend reversed at 900 °C. For PS gasification, syngas, hydrogen, and energy yields increased exponentially with temperature, while in pyrolysis, the increases were linear. Compared to gasification, pyrolysis produced higher syngas quality at all temperatures. Yao *et al.* investigated the pyrolysis-catalytic steam reforming of PS over three Ni/Al<sub>2</sub>O<sub>3</sub> catalysts for syngas production using a two-stage tube reactor.<sup>234</sup> The reactor consisted of two stages (pyrolysis stage and catalytic stage) with operating temperatures of 500 °C and 900 °C, respectively. The catalysts were synthesized using co-precipitation, impregnation, and sol-gel methods. Among them, the Ni/Al<sub>2</sub>O<sub>3</sub> catalyst prepared *via* the sol-gel route achieved the highest syngas yield, producing 62.26 mmol H<sub>2</sub> per g plastic and 36.10 mmol CO per g plastic. This enhanced performance was attributed to the sol-gel catalyst's higher surface area and finely dispersed nickel particles with uniform distribution.

The gasification atmosphere also affects the syngas production from PS. Zhao *et al.* studied the gasification of PS in supercritical water with CO<sub>2</sub> at 400–700 °C. The carbon conversion efficiency of PS reached 47.6% at 700 °C; however, no conversion occurred at 400 °C.<sup>235</sup> The addition of CO<sub>2</sub> to supercritical water increases the production of H<sub>2</sub>, CH<sub>4</sub>, and CO<sub>2</sub>, which suggests that the presence of CO<sub>2</sub> promotes the gasification of PS. Furthermore, Choi *et al.* found that syngas (H<sub>2</sub>/CO) formation during the catalytic pyrolysis of PS with 10 wt% Ni/SiO<sub>2</sub> was five times higher under CO<sub>2</sub> compared to N<sub>2</sub>.<sup>232</sup>

Co-processing (Co-gasification and Co-pyrolysis) of the PS and other components, such as biomass and plastics, has also been reported with improved performance observed due to synergetic effects.<sup>236–238</sup> Basha *et al.* investigated the co-gasification of PS and palm kernel using an electrically heated

downdraft gasifier at 700–900 °C in air without a catalyst.<sup>236</sup> Their results showed that higher gasification temperatures increased the H<sub>2</sub> and CO volume fractions while decreasing CH<sub>4</sub> content in the syngas. Additionally, increasing the PS proportion in the feedstock blend enhanced overall syngas production at 900 °C. Janajreh *et al.* studied the gasification of the PS and PS/PE/PP mixture using the high-fidelity entrained-flow-gasification model, which was validated experimentally.<sup>237</sup> Gasification was also conducted on a drop-tube-reactor with an equivalence ratio of 1.8 at 1000 °C under atmospheric pressure. The results found that the cold gasification metrics increased from 73.13% (PS) to 89% for the co-gasification mixture, suggesting that there would be no need for the extra cost of sorting the plastic waste before the entrained flow gasification. Déparrois *et al.* studied syngas production through co-pyrolysis and CO<sub>2</sub> co-gasification of PS, using both thermo-gravimetric and a laboratory-scale tubes at ~900 °C.<sup>238</sup> Total gas yield increased with CO<sub>2</sub> co-gasification, accompanied by enhanced synergistic conversion. This synergistic effect reduced the char available for reaction with CO<sub>2</sub> during gasification, resulting in a lower CO yield.

To quantify the synergistic effects in co-pyrolysis of PS with biomass, Burra *et al.* investigated the co-pyrolysis of pinewood (PW) and PS blends at 900 °C in a fixed-bed reactor.<sup>239</sup> The results showed that co-pyrolysis synergistically enhanced the yields of syngas components; H<sub>2</sub> and CO increased by 80% and 67%, respectively, compared to the weighted yields from the individual pyrolysis of PS and pinewood. Different interactions, including volatile–volatile, volatile–feedstock, and volatile–char interactions in the co-pyrolysis, as well as individual pyrolysis of PS and PW, were deconvoluted through uniquely arranged sample loading configurations. Volatile–volatile interactions contributed to a 33% increase in H<sub>2</sub> yield, whereas PS volatile–PW intermediate interactions added 17%. In contrast, inhibitive interactions between PW volatiles and PS intermediates

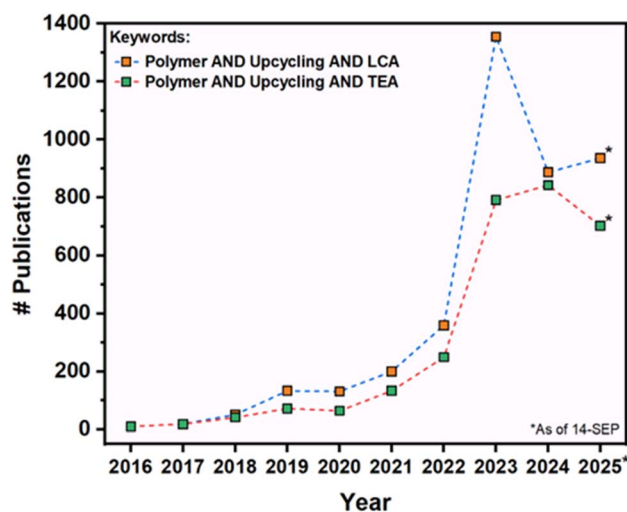


Fig. 8 Number of publications related to LCA and TEA on the topic of polymer upcycling since 2016. Data acquired from dimensions AI scientific database using the "AND" boolean operator. Specific unrelated scientific fields were excluded using the "NOT" boolean operator.





Table 8 Selection of LCA studies on polymer upcycling since 2020

Feedstock	Process	Products	Remarks	Ref.
Waste plastic pyrolysis oil from mixed polyolefin Post consumer waste	Oil to olefin conversion	Fuels	<ul style="list-style-type: none"><li>• 1.334 kg feedstock + 0.283 kWh electricity + 0.313 kWh natural gas per kg product</li><li>• 2.32 kg CO<sub>2</sub> equivalent per kg product</li><li>• GWP for same technology varies based on electricity source + global region</li><li>• “Cradle to gate” following ISO 14040 (OpenLCA 1.10.3 + Ecoinvent v3.7 + AspenPlus)</li><li>• Reduction in GWP and nonrenewable energy use by over 70% and 40% respectively as compared to virgin PET (assuming Europe or China as location)</li><li>• Scaled up aspen models evaluated with OpenLCA</li><li>• Solid/liquid loading has big impact on GHG emissions – 0.04 g ml<sup>-1</sup> minimum requirement for GHG emissions to be below that of virgin production</li><li>• Following ISO 14040 (Quantis Suite 2.0/IMPACT vQ2.2 + Ecoinvent 2.2)</li><li>• Results vary dramatically based on LCA boundaries/assumptions</li><li>• Downcycling is most favorable if model based on virgin material substitution rate</li><li>• Upcycling performs more favorably when target market for recycled material is high, and quality of material is considered</li></ul>	247
		Chemicals LDPE		
	Acetolysis	TPA		
Waste polyesters (PET, PETG, PEF, PEN, PBT)			EGDA (ethylene glycol diacetate)	248
Post consumer PET (transparent & colored)	Alkaline hydrolysis	TPA	EG	249
Printed plastic film waste	Comparison (upcycling, downcycling, waste to energy incineration)	Dependent on process		250
HDPE waste	Conventional pyrolysis vs. thermo oxo-degradation	Ethylene propylene naphtha diesel waxes		251
LDPE waste	Comparison (pyrolysis, gasification, hydrocracking, HTL, hydrogenolysis)	Dependent on process		245
Agricultural film waste	Catalytic hydrocracking	Naphtha		252





Table 8 (Contd.)

Feedstock	Process	Products	Remarks	Ref.
5 layer PE-PA plastic film waste	Comparison (landfill, incineration, solvent-based reuse, STRAP, pyrolysis, downcycling)	Dependent on process	<ul style="list-style-type: none"> <li>• "Cradle to grave" using TRACI + Ecoinvent V3.8</li> <li>• STRAP results in lowest environmental impact in most categories – incineration the highest impact in most categories</li> <li>• Credits from substituted products significantly impact overall GWP of processes</li> </ul>	253
Post-consumer PE waste	Hydrogenolysis	Lubricants	<ul style="list-style-type: none"> <li>• Conceptual 250 MT facility modeled with Aspen Plus v12</li> <li>• Significantly lower GHG emissions than petroleum-based processes assuming use of H<sub>2</sub> and electricity from renewable sources</li> </ul>	254

resulted in an 8% decrease. The work provides a unique strategy for studying the synergetic effect of co-processing. More studies are needed to improve understanding of the co-processing process.

#### 5.4. Challenges with contamination

Contaminants in plastic waste, such as chlorine from PVC, nitrogen-containing additives, metals, dyes, and food residues, pose serious challenges to gasification technologies. In gasification, these contaminants can deactivate catalysts, poison active sites, and promote tar formation, which clogs reactor systems and reduces syngas quality.<sup>180</sup> The degree of contamination and polymer compatibility vary significantly across plastic types, necessitating tailored strategies. For example, polyolefins tend to tolerate more heterogeneity, whereas PET and PVC require stricter control due to thermal and chemical sensitivities.<sup>240</sup> To address these issues, advanced catalysts with robust thermal and chemical stability are employed. Recently, Ni-based catalysts with core-shell structures have demonstrated resistance to sintering and deactivation.<sup>241</sup> Overall, a combination of catalyst engineering, process optimization, and feedstock pre-treatment is essential for effective contaminant management across different recycling technologies. Additionally, pre-treatment steps such as mechanical sorting, washing, or chemical extraction are often used to reduce feedstock impurities.<sup>54,242</sup>

## 6. Life cycle assessment (LCA) and techno-economic analysis (TEA)

LCA is a comprehensive methodology towards evaluating the total environmental impact of a product throughout its entire life cycle. Techno-economic analysis (TEA) is a similarly comprehensive methodology that assesses the economic performance of specific processes from beginning to end. Both have been applied towards the development of practical upcycling solutions for polymer waste. LCA helps inform the status of upcycling technologies from an environmental standpoint through factors such as energy consumption and global warming potential. TEA helps to ascertain the feasibility of upcycling technologies from an industrial perspective through capital expenditure (CapEx) and operational expenses (OpEx). Data from both LCA and TEA studies help towards achieving the 17 Sustainable Development Goals outlined by the United Nations. More specifically, LCA and TEA studies on polymer upcycling fall directly under goal 9 (Industry, Innovation, & Infrastructure), goal 12 (Responsible Production and Consumption), and goal 13 (Climate Action).<sup>243</sup> The number of publications related to both LCA and TEA on polymer upcycling has increased considerably over the past decade. Interestingly, however, there has been a decline in scientific works since a peak in 2023 for LCA studies and 2024 for TEA studies (Fig. 8).

The majority of LCA studies related to the topic of polymer upcycling conducted were carried out with the assistance of specialized LCA software and databases: SimaPro™, OpenLCA™, and Ecoinvent are some of the most commonly used research tools. These LCA software and databases are compliant with the





Table 9 Selection of TEA studies on polymer upcycling since 2020

Feedstock	Process	Product	Remarks	Ref.
PLA waste	Hydrolysis, alcoholysis	New PLA	<ul style="list-style-type: none"><li>• Aspen Plus with NRTL model</li><li>• Heating/cooling represents significant portion of total utility costs</li><li>• Annual equipment/installation cost higher for hydrolysis than for alcoholysis</li><li>• Annual operational cost lower for hydrolysis than for alcoholysis</li></ul>	257
Post-consumer PE waste	Hydrogenolysis	Lubricants	<ul style="list-style-type: none"><li>• Conceptual 250 MT facility modeled with Aspen Plus v12</li><li>• Leasing of catalyst results in lower total capital costs, higher annual operating costs</li></ul>	254
PS waste	Pyrolysis	Styrene monomer	<ul style="list-style-type: none"><li>• Aspen Plus v12 + Monte Carlo simulation methods</li><li>• Total product costs higher than total capital investment, difference between the two aforementioned values increases with increasing plant capacity</li><li>• Levelized production costs decrease with increasing plant capacity</li></ul>	258
HDPE waste	Pyrolysis	Ethylene	<ul style="list-style-type: none"><li>• BioSTEAM process model + discounted cash flow rate of return method</li></ul>	251
	Thermo	Propylene	<ul style="list-style-type: none"><li>• Thermo oxo-degradation has lower equipment cost, higher operating cost</li></ul>	
	Oxo-degradation	Naphtha	<ul style="list-style-type: none"><li>• Addition of hydrocracking significantly increases both capital cost and value of products produced</li></ul>	
5 layer PE-PA Plastic film waste	Comparison (landfill, incineration, solvent-based reuse, STRAP, pyrolysis, downcycling)	Diesel Waxes Dependent on process	<ul style="list-style-type: none"><li>• Incineration has by far highest net supply chain and net end of life costs</li><li>• Pelletization the only process to show negative net end of life cost (economically favorable)</li></ul>	259
LDPE waste	Comparison (pyrolysis, gasification, hydrocracking, HTL, hydrogenolysis)	Dependent on process	<ul style="list-style-type: none"><li>• Discounted cash flow method</li><li>• Gasification, HTL, and hydrocracking all have negative ROI</li><li>• Collection and sorting represent over 50% contribution of total process costs (except hydrogenolysis at 42.5% contribution)</li></ul>	245
Agricultural film waste	Catalytic hydrocracking	Naphtha	<ul style="list-style-type: none"><li>• Net profit only at production scales above 2000 tons per year</li><li>• OPEX directly proportional to amount of feedstock processed</li></ul>	252

environmental standards ISO 14040 and 14044 outlining the principles, framework, requirements and guidelines for all LCA.<sup>244</sup> The streamlining of the LCA process, required data inputs, and consolidation of existing data provided by the online resources have created an environment where comprehensive LCA studies can be efficiently carried out. It is this favorable environment which has largely enabled the large increase in LCA publications related to polymer upcycling in recent years. For example, net CO<sub>2</sub> emissions for gasification and HTL of LDPE were found by a 2023 study to be greater in value compared to not only hydrocracking and pyrolysis, but also conventional landfilling.<sup>245</sup> LCA on the aminolysis of rigid PET packaging into monomers with subsequent aramid polymer synthesis also highlighted the effect of amine selection on various environmental factors.<sup>246</sup> It was found by the 2025 study that the utilization of 1,4-phenylenediamine could result in an increase in greenhouse gas emissions of up to 25 times as compared to virgin Kevlar synthesis depending on the amine loading percentage. Use of 1,6-hexanediamine on the other hand was found to have up to a 50% reduction in greenhouse gas emissions and 92% reduction in ozone depletion as compared to virgin Kevlar. It is important to note however that the final monomer yield obtained with that amine was quite low at only 22%. Such findings highlight how LCA helps to expose the true complexity of identifying and developing polymer upcycling technologies which effectively output valuable products while simultaneously minimally impacting the environment. An overview of a selection of recent LCA studies on polymer upcycling technologies is provided below (Table 8).

In many cases, publications which present LCA data on polymer upcycling processes also present corresponding TEA findings for those same systems. For instance, a 2023 evaluation of potential full scale thermochemical upcycling pathways identified that the incorporation of product valorization infrastructure to pyrolysis-based processes to further refine pyrolysis oil into monomers can approximately double the annual revenue generated as compared to pyrolysis into oil without further treatment, all with a payback period of less than four years.<sup>255</sup> The study examined infrastructure related to conversion of post-consumer waste plastics in the Upper Midwestern states of Illinois, Iowa, Minnesota, and Wisconsin in the United States. Similarly, a 2021 German case study was carried out evaluating both life cycle analysis and techno-economic performance of waste polymer pyrolysis into monomer for new virgin polymer synthesis as compared to more conventional mechanical recycling strategies and combined pyrolysis mechanical recycling solutions.<sup>256</sup> The net production cost per kilogram of feedstock processed was found to be significantly less for pyrolysis and combined pyrolysis mechanical recycling as compared to pure mechanical recycling, indicating an increase in profits. Despite higher production costs as compared to mechanical recycling, the value of the monomer obtained from pyrolysis led to increased net profit, thus highlighting the importance of value-added products *via* upcycling toward the feasibility and implementation of polymer waste handling technologies. An overview of a selection of other recent TEA studies on polymer upcycling technologies is provided below (Table 9).

## 7. Conclusion and recommendations

The upcycling and recycling of plastic waste into valuable chemicals, materials and energy has emerged as a necessity to address the global plastics crisis. In this review, state-of-the-art developments from the most recently developed chemical upcycling strategies, reaction mechanisms, product selectivity, reaction chemistries, and efficiencies were systematically discussed. More specifically, upcycling advancements in solvent-based dissolution, chemical depolymerization, thermal conversion methods (*i.e.*, pyrolysis, hydrothermal liquefaction, hydrocracking, hydrogenolysis), and gasification for the conversion of plastics such as PP, PE, PET, PU, PS and PVC into value-added products (*i.e.*, monomers, syngas, naphtha, fuels and chemicals) were highlighted. Solvent dissolution holds the ability to preserve polymer integrity for reuse, while chemical depolymerization enables the selective recovery of monomers and oligomers from condensation polymers such as PET and PU for repolymerization of new polymers. Thermochemical routes including pyrolysis and HTL are highly effective at converting polyolefin-rich feedstocks into high-yield, high-carbon fuels and naphtha. Finally, gasification can produce valuable syngas.

Process efficiency and product quality are highly sensitive to variables such as temperature, heating rate, catalyst type, and feedstock composition. For example, increasing pyrolysis temperature during pyrolysis from 400 °C to 450 °C significantly enhances the oil yield from PS, while temperatures above 800 °C during gasification promote greater hydrogen and CO generation. Both gaseous products are suitable for clean energy applications. Co-processing strategies such as co-pyrolysis of plastic mixtures have also demonstrated synergistic effects, improving both product selectivity and carbon utilization efficiency. Future efforts should focus on developing cost-effective, durable, and selective catalysts, particularly for complex and mixed plastic waste streams. Advancements are needed in integrated process designs that combine upstream sorting, pretreatment, and downstream refining to maximize yield and product purity. Novel technologies, including microwave-assisted pyrolysis, tandem catalysis, and AI-driven process optimization, should be further explored to enhance efficiency and selectivity. Expanding pilot-scale upcycling followed by comprehensive TEA and LCA is crucial for evaluating real-world feasibility. At the same time, fostering close collaboration across disciplines, along with developing supportive policies and regulatory frameworks, will be crucial in turning plastic upcycling technologies into practical, widely adopted solutions.

## Author contributions

Conceptualization, K. J., E. Y., X. Z., and W.-T. C.; methodology, K. J., D. D., E. Y., T. L., K. L., H. J., K. N., E. W., S. O., R. Z., and L. D.; validation, K. J., E. Y., T. L.; formal analysis, K. J., D. D., E. Y., T. L.; investigation, K. J., D. D., E. Y., T. L., K. L. H. J., K. N., E. W., S. O. R. Z., L. D., X. Z., and W.-T. C.; data curation, K. J. and E. Y.; software validation, K. J. and E. Y.; writing – original draft preparation, K. J., D. D., E. Y., T. L., K. L. H. J., K. N., E. W., S. O. R. Z., and L. D.; writing – review and editing, K. J., D. D., E. Y.,



T. L., K. L. H. J., K. N., E. W., S. O. R. Z., L. D., H. L., R. R., X. Z., and W.-T. C.; visualization, K. J.; supervision, X. Z. and W.-T. C.; project administration, X. Z. and W.-T. C.; funding acquisition, W.-T. C.

## Conflicts of interest

The authors declare no competing financial interests.

## List of abbreviations

ABS	Acrylonitrile butadiene styrene
ACE	Acetone
BHET	Bis(2-hydroxyethyl)terephthalate
BTX	Benzene, toluene, xylene
CO <sub>2</sub>	Carbon dioxide
COSMO-RS	Conductor-like screening model for real solvents
COSMOtherm	Conductor-like screening model for thermodynamics
DCM	Dichloromethane
DES	Deep eutectic solvent
DMF	Dimethylformamide
DMSO	Dimethyl sulfoxide
DP	Degree of polymerization
EG	Ethylene glycol
EPA	Environmental protection agency
EPR	Extended producer responsibility
ESI	Electrospray ionization
EVOH	Ethylene vinyl alcohol
FTIR	Fourier transform infrared spectroscopy
HDPE	High-density polyethylene
HSP	Hansen solubility parameters
HTL	Hydrothermal liquefaction
IL	Ionic liquid
LDPE	Low-density polyethylene
LLDPE	Linear low-density polyethylene
M-Clay	Montmorillonite clay
MD	Molecular dynamics
MMT	Montmorillonite
NMP	N-Methyl-2-pyrrolidone
PA	Polyamide
PC	Polycarbonate
PE	Polyethylene
PET	Polyethylene terephthalate
PLA	Polylactic acid
PMMA	Polymethyl methacrylate
PP	Polypropylene
PS	Polystyrene
PU	Polyurethane
PUR	Polyurethane resin
PVC	Polyvinyl chloride
SAN	Styrene acrylonitrile
SAPO-11	Silicoaluminophosphate
SAR	Structure-activity relationship

STRAP	Solvent-targeted recovery and precipitation
TDI	Toluene diisocyanate
TEA	Techno-economic analysis
THF	Tetrahydrofuran
$T_g$	Glass transition temperature
VL	$\gamma$ -Valerolactone
WtE	Waste-to-energy
Zn (OAc) <sub>2</sub>	Zinc acetate
Zn/SBA-15	Zinc on mesoporous silica

## Data availability

No new data were generated or analyzed in this study. This article is a review and does not include primary research results, software, or code.

## Acknowledgements

This study was supported in part by the Harnessing Emerging Research Opportunities to Empower Soldiers (HEROES) Initiative (contract number: W911QY2020005), the U.S. Department of Energy's Office of Energy Efficiency and Renewable Energy (EERE) Advanced Manufacturing Office (AMO, award number DE-EE0009944), and the U.S. Department of Energy's Office of EERE Bioenergy Technologies Office (Award Number DE-EE0009666 awarded to Cascadia Consulting Group). This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, products, or processes disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof. The authors thank Dr Yang Xiao for his initial interest and discussion. The authors acknowledge the support from the U.S. Department of Energy's Bioenergy Technologies Office. This manuscript was authored in part by UT-Battelle LLC under contract DE-AC05-00OR22725 with DOE. The US government retains and the publisher, by accepting the article for publication, acknowledges that the US government retains a nonexclusive, paid-up, irrevocable, worldwide license to publish or reproduce the published form of this manuscript, or allow others to do so, for US government purposes. DOE will provide public access to these results of federally sponsored research in accordance with the DOE Public Access Plan (<http://energy.gov/downloads/doe-public-access-plan>).



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