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## Can mixed plastics be recycled and upcycled without separation?

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With more than 400 million tonnes of plastic waste produced each year, developing effective treatment options for post-use plastics is an urgent imperative to reduce plastic pollution whilst preserving their material and chemical value within a circular economy. Whilst recycling and upcycling are emerging as important strategies to improve plastic circularity, a key bottleneck for practical adoption of these technologies is the complexity of plastic feedstock, especially from municipal streams heavily contaminated with different plastic types and non-plastic waste. This often necessitates plastic sorting and cleaning, whose complexity and labour-intensiveness add to the cost of recycling and upcycling and hinder more widespread adoption. In this review, we critically examine and spotlight the possibility of recycling or upcycling mixed plastics as an alternative to utilising only clean, single-component plastic feedstock streams. We discuss strategies to chemically recycle and upcycle mixed plastics into industrially-relevant molecules and functional materials, as well as the possibility of repurposing mixed plastics for polymer blend materials. Through this critical discussion, we hope to highlight the pressing need for designing emerging technologies for addressing the inherent heterogeneity of real-life plastic waste streams, contributing to an economically-viable and sustainable post-use plastics economy.

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### Green foundation

- Approaches towards recycling and upcycling of post-use plastics are dominated by technologies that use single plastic types as feedstock. However, as real-life plastic waste is inherently heterogeneous and heavily contaminated, mixed/contaminated feedstock should be considered instead. This review critically assesses the feasibility of this approach.
- New approaches towards recycling/upcycling mixed plastics significantly reduces logistical challenges and costs. These approaches are interdisciplinary and will impact multiple fields in both academia and industry.
- The future of practical plastic waste recycling and upcycling will be to embrace mixed plastics as feedstock instead of clean, single plastic sources. By highlighting the feasibility of recycling and upcycling approaches to mixed plastic feedstock, this review challenges the current *status quo* in plastics end-of-life R&D and shapes the development of new methodologies that account for feedstock heterogeneity.

## 1. Introduction

Plastics play a wide range of important roles in modern society, favoured for their diverse properties, low production cost, durability, and ease of processing. However, end-of-use plastic products represent a major waste disposal problem as the majority possess linear and unsustainable take-make-throw life cycles. The bulk of post-consumer plastics are cur-

rently disposed of in landfills and incineration or irresponsibly disposed of in the environment, resulting in complete loss of their intrinsic chemical and material value, while releasing harmful by-products such as greenhouse gases, particulate matter, heavy metals and micro-plastics that are detrimental to human health and the environment. Recent efforts in improving plastic circularity have led to the rising demand for global plastic recycling and upcycling, with a projected industrial growth surpassing USD 100 billion by 2033.<sup>1</sup> Although both practices aim to improve plastic circularity, they differ conceptually. Plastic recycling primarily focuses on recovering the same polymers from post-use plastics for different products *via* either physical processing and/or sequential chemical depolymerisation and repolymerisation. Namely, rPET reprocessed from post-use PET plastic bottles (mechanically or

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chemically) could be used for similar applications (e.g. back to plastic bottles for beverages). In contrast, plastic upcycling focuses on transforming plastic feedstock into new products of higher economic value for different applications, without recovery of the original polymer. Advantageously, both recycling and upcycling processes allow recovery of the embedded material and chemical value present in waste plastic polymers that would otherwise be lost upon disposal, whilst providing an alternative feedstock from petroleum.<sup>2</sup>

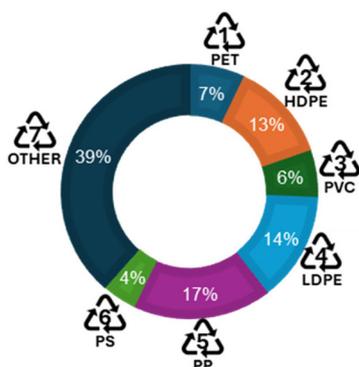
The composition of municipal plastic streams can vary greatly depending on the source and geographical location: global plastic waste produced in 2019 comprised of PET (7%), HDPE (13%), PVC (6%), LDPE (14%), PP (17%), PS (4%) and other plastics (39%), amounting to 353.3 million tonnes cumulatively (Fig. 1a). Plastic waste can come from both industrial and municipal sources. Post-industrial plastic waste (from industrial manufacturing processes) provides cleaner feedstock with well-known compositions albeit containing manufacturing additives<sup>3</sup> (Fig. 1b), while municipal plastic waste contains a mixture of additive-containing plastics that are often found contaminated with food, water and other materials (e.g. paper, metal or glass).<sup>4,5</sup> Furthermore, it is not uncommon for a single plastic product to incorporate a blend or composite of multiple plastic types with differing molecular composition and reactivity. Coupled with the unavoidable contamination during usage such as food remnants, they build up a complex plastic source which complicates the waste management process and inevitably hinders their recycling efficacy and the quality of recycled plastic products.

While there has been much attention on developing new strategies and processes to improve plastic waste recycling and upcycling, most still require relatively clean single polymer feedstock, necessitating prior sorting and cleaning of real-life plastic waste. At present, mechanical recycling is the most widely employed plastic recycling method, typically involving sequential sorting, sanitation, shredding, melting and reforming. Despite significant efforts and legislative support, only 9% of global plastic waste is recycled at present.<sup>7</sup> Recycling faces

numerous challenges, which include significant cost competition from inexpensive virgin plastics, the loss of material properties from inevitable polymer chain degradation over repeated cycles of heating and high shear conditions, and the presence of impurities which can be detrimental to the quality of the recycled products. Recycling rates are also highly dependent on the plastic type, its distinct chemical or physical properties and overall usage.<sup>8</sup> For instance, PET and HDPE have the highest recycling rates, while plastics like PS are rarely recycled.<sup>9</sup> While chemical recycling allows reformation of virgin-grade plastics through depolymerisation and repolymerisation, it requires high capital expenditure for infrastructure and can be disadvantaged by the high energy costs necessary. Chemical recycling is also more suitable for plastics containing site-cleavable bonds such as polyesters and polycarbonates, whilst polyolefin chemical recycling is plagued by low monomer recovery (typically <30%) despite the very high temperatures needed (typically >500 °C).<sup>10</sup>

Contaminants in plastic waste streams can be removed *via* different pretreatment strategies prior to recycling.<sup>11</sup> Typically, surface contaminants can be removed by washing waste plastics with water or with a combination of mechanical friction, elevated temperatures, sodium hydroxide or detergents for increased washing efficacy, while other methods such as hot air or steam stripping, dissolution–precipitation and solid–liquid extraction may be more effective at removing volatile organic compounds and colourants to address deodorisation or de-inking requirements.<sup>12,13</sup> For example, solid–liquid extraction involves removing solvent-soluble contaminants from the polymer without dissolving it, with more advanced methods assisted by surfactants, ultrasonic frequencies, microwaves, supercritical fluids and pressurised fluids.<sup>11,14</sup> While these pretreatment strategies are effective at removing undesirable contaminants, challenges in recycling or upcycling often lie with the incompatibility between multiple plastic types during processing. Thus, sorting is often a critical step in the recycling or upcycling of municipal plastic waste.

(a) Global waste generation by polymer type



(b) Additive composition in plastic products

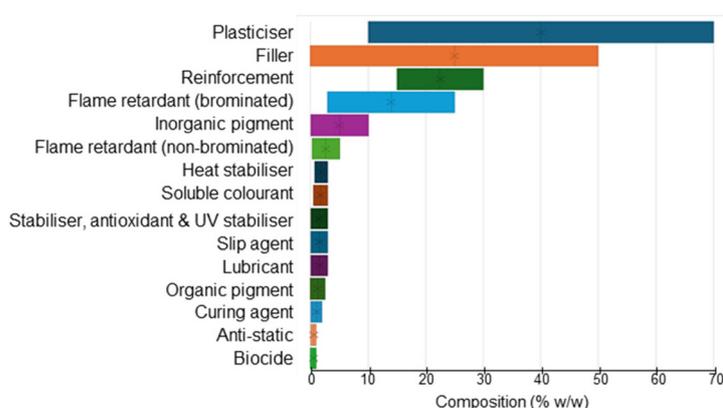


Fig. 1 (a) Global waste generation by polymer type, reproduced from data reported by OECD.<sup>6</sup> (b) Common additives and their composition in plastic products, adapted from ref. 3.

The simplest form of plastic sorting is manual sorting, which is both inefficient and labour intensive. In 1988, the Resin Identification Code was introduced by the US Society of the Plastics Industry (SPI code)<sup>15</sup> as a concise plastic identification system to facilitate the plastic sorting process during post-use recycling. The SPI codes 1–6 correspond to the six most common commercial plastics, also known as the “Big Six”, namely PET, HDPE, PVC, LDPE, PP and PS, respectively, which account for more than 75% of all plastics produced.<sup>16</sup> Although the SPI coding system is still widely employed worldwide today, identification of plastics in waste streams remains challenging due to different extents of compliance, non-uniformity of the resin identification codes, the vast volume of waste plastics that need to be processed, and inevitable human errors. Additionally, the presence of plastic blends and multi-packaging materials that utilise several types of plastics or other materials (e.g. paper) further complicates identification and sorting. With technological advancements in sorting processes, modern approaches incorporate more automation, such as density separation, electrostatic separation and sensor-based separation.<sup>17</sup> These processes rely on the innate properties of individual plastic types to enable improved accuracy and consistency during the sorting (or partial sorting) process. For instance, the deconstruction of multilayer plastic films into their constituents can be achieved through a series of solvent-targeted recovery and precipitation based on preferential dissolution of each constituent polymer in selected solvents at specific temperatures.<sup>18</sup> In the case of a PE–EVOH–PET multilayer film, selective dissolution of PE and EVOH in hot toluene and DMSO, respectively, followed by precipitation of the filtrate in suitable antisolvents, gave two components, while PET was isolated as a solid. Dissolution temperature selection was also crucial, with 95 °C chosen for the preferential recovery of EVOH in DMSO due to the known higher solubility of PE in DMSO near 100 °C. However, such advanced sorting processes require significant monetary investment, where the cost varies based on the operation scale and specific technology, with a projected cost amounting to ~USD 32 billion globally between 2021 and 2040.<sup>19</sup>

Thus, the inability to utilise contaminated municipal plastic waste directly and the need for costly and tedious sorting hinder the practicality of plastic recycling. Furthermore, despite the various sorting technologies available, either working individually or in collaboration, no approach is flawless, resulting in inconsistency in waste plastic management across different geographical regions. Considering the immense challenge to recover embedded value from millions of tonnes of mixed plastic waste generated annually and the technological and logistical requisites of plastic sorting, we reflect upon the question: can we bypass costly and tedious sorting processes and recycle or upcycle mixed plastics directly? Herein, this review considers the advancements in chemical recycling and/or upcycling strategies for mixed plastic feedstock containing at least two different plastic polymers and at least one “Big Six” plastic. Uncovering new methods to valorise municipal plastic waste

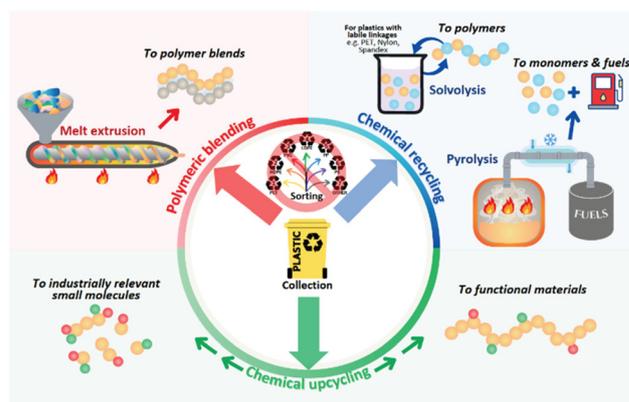


Fig. 2 Recycling and upcycling strategies for post-consumer mixed plastic waste without sorting.

without the need for prior sorting will simplify their post-use life cycle, likely resulting in cost reduction, and could potentially translate to improved economic viability and rates of recycling and/or upcycling. We examine three scenarios that are generally relevant for most commercial plastics (Fig. 2): (1) chemical recycling of mixed plastics where chemical depolymerisation into respective monomers occurs *via* bond cleavage, followed by repolymerisation of said monomers to reform the original plastic polymer; (2) chemical upcycling of mixed plastics into industrially relevant small molecules or functional polymers with an expanded range of applications; and (3) physical incorporation of mixed plastic feedstock into polymer blends.

## 2. Chemical recycling

Chemical recycling processes involve the recovery of monomers or oligomers from post-use plastics through chemical depolymerisation processes, followed by repolymerisation to reform the original polymer type. This process is an attractive strategy to promote a sustainable circular economy as it enables the continuous high-volume production and usage of existing plastics without the environmental impacts of landfilling or incineration.<sup>20,21</sup> As the products of chemical depolymerisation are small molecules, their purity can be ensured *via* common purification techniques such as recrystallisation, that are achievable on industrial scales and enable high yields in monomer recovery for closed-loop recycling. Advantageously, chemical recycling can recover plastics from multi-plastic mixtures and even multilayer packaging. While an infinite closed-loop plastic circularity can be achieved in theory, chemical recycling is often disadvantaged by the generally high energy demands, operating costs, and greater suitability for certain classes of plastics (e.g. PET, polycarbonates) compared to others (e.g. polyolefins, which give poor monomer recovery). Chemical recycling processes can be broadly classified into two strategies: site-specific chemical depolymerisation, whereby plastic polymers containing cleava-

ble bonds are selectively converted into their monomers, and thermal cracking which utilises heat to break down (or crack) polymers into a monomer-containing mixture of small molecules/oligomers. Here, we will discuss how these two chemical recycling strategies are applicable to mixed plastic feedstocks, as well as their advantages and disadvantages.

## 2.1 Recycling of plastics through site-specific depolymerisation

Site-specific chemical depolymerisation selectively converts plastic polymers into monomers, typically through solvolysis processes such as glycolysis, methanolysis or hydrolysis. This process is highly applicable to real-life plastic streams as it is tolerant of most additives and polymer mixtures or blends due to its specificity to certain chemical functionalities. Similarly, the main drawback of site-specific chemical depolymerisation is its restriction to only selected polymers with cleavable bonds (such as esters, amides, carbonates and urethanes). Because of this, PET is the only plastic currently amenable for site-specific chemical depolymerisation out of the “Big Six”. However, this restriction does not limit its practicality, with site-specific chemical depolymerisation of PET performed commercially by companies such as Garbo,<sup>22</sup> IBM<sup>23</sup> and Dupont-Tenjin<sup>24</sup> on large scales, yielding monomers such as bis(hydroxyethyl) terephthalate (BHET) or terephthalic acid (TPA) at a scale of 10 000 to 20 000 tons annually. The monomers are then reused as raw materials for the production of rPET with comparable properties to virgin PET.<sup>25</sup> Among all the chemical recycling processes for PET, glycolysis emerges as the most effective approach, especially on products with high PET content.<sup>26</sup> The most prominent example is the widely seen PET plastic bottles, which often contain a percentage of polyolefins in the form of bottle caps or print labels.<sup>26,27</sup> In spite of these contaminants, glycolysis experiments carried out by Aguado *et al.*<sup>26</sup> on actual PET waste showed that the unreacted PE contaminants could be easily separated from the reaction product after glycolysis *via* filtration due to its poor solubility in the reaction medium. Furthermore, depolymerisation of highly coloured PET (coloured PET bottles) and complex multilayered PET (beer or juice bottles) was also capable of affording high quality BHET and TPA, suggesting the resilience of hydrolysis and glycolysis processes towards different contaminants.

Besides disposable PET bottles, another major source of PET waste comes from the textile industry. Glycolysis of PET from mixed textile waste is notably more challenging as PET-containing textile products are often spun tightly together with cotton, nylon, spandex and other types of polyesters while also mixed with other contaminants such as additives or colourants, resulting in high heterogeneity in the waste plastic feedstock.<sup>28</sup> Furthermore, textile mixtures may also contain other polyester or nylon polymers that are susceptible to depolymerisation into their respective monomers during glycolysis, complicating the process as these products may react with BHET to produce other products. In light of this, Chen *et al.*<sup>29</sup> developed a seamless three-step chemical recycling of PET into BHET from the mixed textile waste (Fig. 3a). First, the textile waste was success-

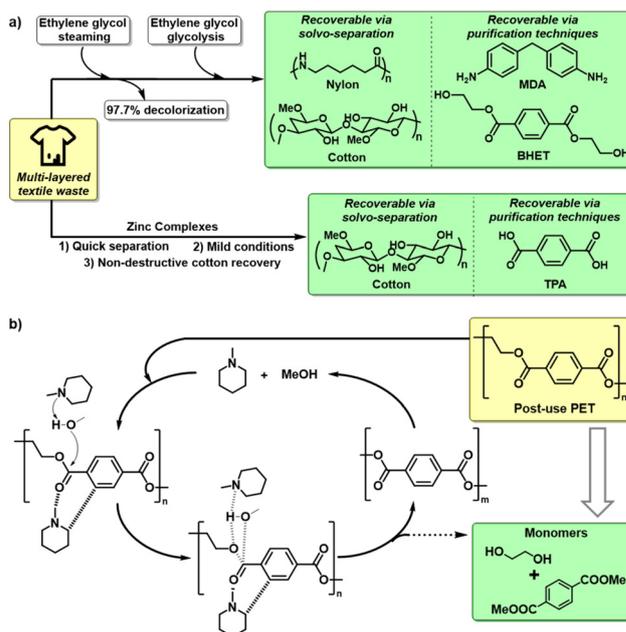


Fig. 3 (a) Chemical recycling of PET into monomers from the mixed textile waste.<sup>26,29,31</sup> (b) PET methanolysis catalysed by NMP, adapted with permission under the CC-BY-NC license.<sup>32</sup> Copyright 2024 Elsevier.

fully decolourised by steaming it with ethylene glycol vapor, achieving up to 97.7% decolourisation efficacy. Then, the decolourised textile mixture was glycolysed with ethylene glycol to form BHET. Finally, BHET was isolated with a high purity of >97% *via* sequential recrystallisation and sublimation with efficient removal of the other polymers or glycolysed products. Using this procedure, high quality BHET could be recovered and subjected to polycondensation with ethylene glycol to form rPET with comparable properties to the parent PET. Recently, Andini *et al.*<sup>30</sup> demonstrated a similar strategy to handle this textile mixture under microwave-assisted glycolysis in 15 min with ZnO as the catalyst. Besides chemical recycling of PET, this procedure accounts for the end-fate of other polymers within the mixed textile waste. While PET and spandex were completely degraded to BHET and diphenylmethane-containing molecules, respectively, it was noteworthy that other polymers such as nylon or cotton did not participate in the glycolysis chemistry. Subsequent recrystallisation could recover BHET, while 4,4'-methylenedianiline (MDA), the most valuable molecule from spandex degradation, could be isolated with common laboratory purification techniques such as solvent extraction, precipitation, or column chromatography. Unreacted cotton and nylon mixtures were also separable by solvent dissolution in 90% formic acid at room temperature, which solubilises nylon while keeping cotton intact.

In another study, Wu *et al.*<sup>31</sup> demonstrated a similar non-destructive cotton recovery process from a PET/cotton textile blend while hydrolysing the PET content into TPA under mild conditions (90 °C for 7 h with 0.1 M NaOH) using a binuclear zinc complex catalyst. This dual-purpose catalyst served to

both accelerate PET hydrolysis and enhance the cotton separation efficiency by 7-fold, with a 97.5% cotton recovery rate. Xie *et al.*<sup>32</sup> also demonstrated successful depolymerisation of different types of waste PET, including mixed packaging materials, *via* a methanolysis process catalysed by different tertiary amine organocatalysts. Out of all the amine organocatalysts evaluated, *N*-methylpiperidine (NMP) stood out as the most reactive catalyst, affording complete deconstruction of PET into dimethyl terephthalate (DMT) and ethylene glycol at 160 °C in 1 h (Fig. 3b). Further in-depth analysis revealed a strong correlation between basicity and methanolysis efficacy, proposing a complex formation between NMP and methanol that amplified its nucleophilicity. At the same time, NMP also activated the ester linkages in PET through two types of hydrogen bonding interactions: (1) interaction between the nitrogen of NMP and the hydrogen of benzene and (2) interaction between the carbonyl oxygen of PET and the methyl hydrogen of NMP. These concurrent interactions enable nucleophilic addition of methanol to the PET ester bonds, leading to an ester bond cleavage and a tetrahedral intermediate held together by hydrogen bonding. A new alcohol was then formed with the regeneration of NMP, and the deconstruction process repeats itself, resulting in the eventual formation of DMT and ethylene glycol monomers (Fig. 3b). This process was also performed on PET-containing textile or multi-material packaging waste to further exemplify the practicality of the process. Similar to previous reports, commonly present inert plastics such as PS, PP or PE did not affect the methanolysis process. Notably, the presence of other polyesters or polyamides such as PLA, PC, poly(butylene terephthalate) or nylons also did not affect the process significantly, affording >90% DMT and ethylene glycol recovery in all cases. On the other hand, the presence of organic dyes could sometimes compete with NMP during the methanolysis process if they possessed reactive functionalities, resulting in the formation of byproducts. However, these dyes are often soluble in methanol and could be removed easily *via* a methanol extraction pretreatment.

Alternatively, site-specific depolymerisation could also be facilitated by engineered enzymatic activity. For example, recombinant *Pichia pastoris* yeast could efficiently depolymerise PET-PE multilayer trays with more than 80% TPA recovery.<sup>33</sup> After which, the isolated TPA was repolymerised to rPET and the recovered PE layer was reprocessed into rPE films with properties comparable to virgin LDPE.

More recently, there has been growing interest in the use of ionic liquids (ILs) for chemical recycling, allowing good plastic dissolution, and they can also act as catalysts for site-specific depolymerisation of PET, PC, PLA and nylons *via* solvolysis. Such reactions can occur at lower temperatures compared to conventional solvents, reducing energy requirements and undesirable side reactions. Although recovery of ILs after depolymerisation has been demonstrated,<sup>35</sup> their environmental impact remains controversial due to their persistence and excellent water solubility, with similarities drawn to PFAS pollution.<sup>36</sup> Nonetheless, they can be potentially applied for depolymerisation of mixed plastic feedstock. For instance, Dou

*et al.*<sup>34</sup> reported the use of cholinium lysinate ([Ch][Lys]) IL for the hydrolysis of a PET/PLA mixed feedstock, achieving over 95% depolymerisation. High depolymerisation efficacy was attributed to the stronger contact probability of [Ch][Lys] with the polyesters as compared to other IL systems, enhancing solvation of the plastic mixture. The authors then utilised the monomer mixture as the sole carbon source for bio-upcycling with engineered *Pseudomonas putida* to produce biodegradable polyhydroxyalkanoates (PHAs). Table 1 summarises current chemical recycling methods employed for site-specific depolymerisation of mixed plastic waste and their recovery strategies.

## 2.2 Chemical recycling of plastics with saturated C–C backbones

Close to 60% of all plastic waste comprises polymers with saturated C–C polymer backbones.<sup>37</sup> Due to the lack of specifically cleavable sites, chemical recycling of these polymers typically involves thermal cracking (such as pyrolysis or gasification) at elevated temperatures (typically ranging from 400 °C to 900 °C) to break down polymers into small molecules and oligomers in an oxygen-free environment. In contrast to site-specific chemical depolymerisation, polymer thermolysis is largely non-polymer specific and applicable to a wide range of plastics,<sup>38</sup> making it a practical approach for mixed plastic depolymerisation with established industrial-scale dedicated facilities.<sup>39</sup> Unfortunately, thermolysis is highly energetically-demanding with poor product selectivity, yielding diverse products ranging from desired monomers to other chemical products that can be used as fuels or fed back into the chemical industry as hydrocarbon feedstock. While generally unselective, the product distribution can be tuned by varying thermolysis conditions such as temperature and catalysts,<sup>40</sup> with the latter playing a critical role in reducing energy demands. For example, with the use of a mixed WO<sub>3</sub>/SiO<sub>2</sub> and Na/γ-Al<sub>2</sub>O<sub>3</sub> catalytic system at 320 °C, Conk *et al.* demonstrated that PE and PP could be converted to propylene and isobutylene with high yields of >90% without the need for prior dehydrogenation.<sup>41</sup> These monomers could be repolymerised to form polypropylene or polyisobutylene. More energy-efficient heating sources such as electrified spatiotemporal heating that can achieve periodically high temperatures (~600 °C) with transient heating durations could also be employed to increase the degree of depolymerisation while minimising unwanted side reactions compared to conventional pyrolysis. As such, relatively high monomer yields of 36% and 43% were obtained for PP and PET depolymerisation separately using similar STH settings but different peak temperatures.<sup>42</sup> This shows the versatility of this technology toward both polyolefins and condensation polymers and its potential applicability for mixed plastic mixtures by tuning the peak temperature setting. Table 2 summarises some strategies to address current challenges in thermal cracking of mixed plastic feedstock.

While thermal cracking has seen much success for different plastic varieties, the presence of other polymers in mixed plastic feedstock can alter product distribution and affect the outcome of the cracking processes.<sup>49</sup> For instance,

**Table 1** Chemical recycling of mixed plastics *via* site-specific depolymerisation

Method	Feedstock	Products	Recovery strategy	Ref.
Glycolysis and alkali hydrolysis	Highly coloured or complex multilayered PET bottles with PE bottle caps or print labels	BHET, TPA, rPET	<ul style="list-style-type: none"> <li>Glycolysis and hydrolysis resilient to colourants and multilayer PET</li> <li>Unreacted PE separated <i>via</i> filtration</li> </ul>	26
Glycolysis with the K <sub>2</sub> CO <sub>3</sub> catalyst	Mixed PET-containing textile waste	BHET, rPET	<ul style="list-style-type: none"> <li>Steam with EG vapor to decolourise up to 97.7% and then glycolysis to BHET</li> <li>Recrystallisation—sublimation achieves &gt;97% purity BHET</li> <li>Recrystallisation and other purification recover BHET and 4,4' - methylenedianiline</li> <li>Unreacted nylon and cotton, nylon separated from cotton by dissolution in 90% formic acid at room temperature</li> </ul>	29
Microwave-assisted glycolysis with the ZnO catalyst	PET/nylon/cotton/spandex	BHET, diphenylmethane-containing molecules ( <i>e.g.</i> 4,4'-methylenedianiline), unreacted nylon and cotton	<ul style="list-style-type: none"> <li>Non-destructive recovery of cotton with 97.5% recovery rate</li> <li>TPA precipitated with sulfuric acid and collected by filtration</li> <li>90% DMT and ethylene glycol recovery in the presence of PS, PP, PE, PLA, PC, PBT and nylon</li> <li>Organic dyes that could compete with NMP are soluble in methanol and removed easily by extraction</li> <li>80% TPA recovery from PET-PE multilayer trays</li> <li>Unreacted PE layers treated with alkali to remove residual PET</li> <li>Recovered PE reprocessed into rPE films with properties comparable to virgin LDPE</li> </ul>	30
Hydrolysis with the binuclear zinc complex catalyst at a low alkali concentration	PET/cotton	TPA, unreacted cotton	<ul style="list-style-type: none"> <li>90% DMT and ethylene glycol recovery in the presence of PS, PP, PE, PLA, PC, PBT and nylon</li> <li>Organic dyes that could compete with NMP are soluble in methanol and removed easily by extraction</li> <li>80% TPA recovery from PET-PE multilayer trays</li> <li>Unreacted PE layers treated with alkali to remove residual PET</li> <li>Recovered PE reprocessed into rPE films with properties comparable to virgin LDPE</li> </ul>	31
Methanolysis with the NMP catalyst	Mixed PET packaging with PS, PP, PE, PLA, PC, PBT, and nylon	DMT, ethylene glycol, unreacted PS, PP, PE, PLA, PC, PBT, and nylon	<ul style="list-style-type: none"> <li>90% DMT and ethylene glycol recovery in the presence of PS, PP, PE, PLA, PC, PBT and nylon</li> <li>Organic dyes that could compete with NMP are soluble in methanol and removed easily by extraction</li> <li>80% TPA recovery from PET-PE multilayer trays</li> <li>Unreacted PE layers treated with alkali to remove residual PET</li> <li>Recovered PE reprocessed into rPE films with properties comparable to virgin LDPE</li> </ul>	32
Enzymatic depolymerisation by recombinant <i>Pichia pastoris</i> yeast	PET-PE multilayer trays	TPA, rPET, rPE	<ul style="list-style-type: none"> <li>80% TPA recovery from PET-PE multilayer trays</li> <li>Unreacted PE layers treated with alkali to remove residual PET</li> <li>Recovered PE reprocessed into rPE films with properties comparable to virgin LDPE</li> </ul>	33
Hybrid chemical-biocatalytic deconstruction using ILs	PET/PLA mixed feedstock	PHA	<ul style="list-style-type: none"> <li>&gt;95% depolymerisation efficacy</li> <li>Tandem upcycling of the monomer mixture into PHA</li> <li>Elimination of the need for PET/PLA sorting and purification</li> </ul>	34

Williams *et al.*<sup>43</sup> demonstrated the influence of mixing PS with five other plastics (HDPE, LDPE, PP, PVC, and PET) in a 1:1 mixture. All the mixtures resulted in a higher gas yield than predicted from the single plastic pyrolysis data, suggesting interactions between PS and the other plastics when mixed. It was particularly interesting to note that the LDPE/PS mixture gave higher ethylene production compared to single LDPE pyrolysis (7.48 wt% and 4 wt% respectively). Similarly, Donaj *et al.*<sup>10</sup> showed that improved monomer recovery up to 23.6 wt% could be achieved from mixed polyolefins (specifically LDPE, HDPE and PP) by fluidised bed pyrolysis with a Ziegler-Natta catalyst at 650 °C, compared to pyrolysis of their pure plastic feedstocks.

Post-consumer waste plastics are often contaminated with heteroelements due to the presence of often complex, heterogeneous mixtures containing organics, papers or cardboards.<sup>50</sup> These contaminants can compromise catalytic activity for depolymerisation. Two common catalyst deactivation mechanisms are acid-pair complexation by nitrogen-containing additives and strong metal-aromatic ring interactions. To circumvent these deactivation processes, Ngu *et al.* reported that a combination of strong acidity, hierarchical porosity, elevated

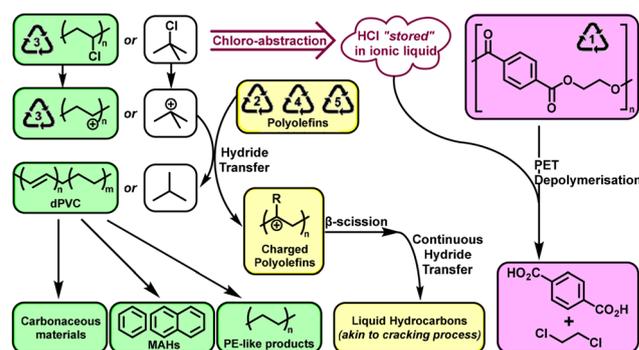
temperatures and rapid heating could achieve efficient recycling of additive-containing plastics.<sup>46</sup> Specifically, they proposed microwave-assisted catalytic cracking over metal-free hierarchical zeolites at moderately high temperatures with short reaction times as an effective method to avoid severe deactivation due to fast and uniform heating, quick removal of products, minimised coking and further cracking for secondary reactions. A range of organic additives were considered, including primary antioxidants (butylated hydroxytoluene and Irganox® 3114), secondary antioxidants (Doverphos® 213 and Irgafos® 168), hindered amine light stabilisers (2,2,6,6-tetramethylpiperidine and Tinuvin® 770), UV light absorbers (Drometrizole and Tinuvin® 571), lubricants (octadecanoic acid and 1-octadecanol), and a slip agent (oleamide). Employing a HY solid-acid catalyst that operates above the desorption temperature of most additives mitigates the impact of weakly adsorbing additives but might not be sufficient for aromatic-containing additives that bind strongly to catalyst metal sites with desorption temperatures >500 °C. In addition, the presence of both mesopores and micropores in the HY catalyst was deemed essential for polymer diffusion to active sites and providing high acidity for rapid polyolefin depolymerisation.

**Table 2** Strategies to address challenges in the thermal cracking of mixed plastic feedstock

Strategy	Feedstock	Products	Ref.
Tuning pyrolysis product distribution with a mixed $\text{WO}_3/\text{SiO}_2$ and $\text{Na}/\gamma\text{-Al}_2\text{O}_3$ catalytic system at 320 °C	PE/PP	Propylene and isobutylene (>90%)	41
Electrified spatiotemporal heating for periodic and transient heating minimises side reactions and maximises the degree of depolymerisation in pyrolysis	PP or PET	Propylene (36%) or 1,4-benzenedicarboxylic acid (43%)	42
Mixed plastic pyrolysis could produce higher monomer yields compared to single plastic pyrolysis	LDPE/PS	Polyethylene (7.48 wt%), higher than pure LDPE pyrolysis (4 wt%)	43
Coupling polyolefin cracking with alkylation using the Lewis acidic chloroaluminate ionic liquid catalyst yields gasoline-range liquid alkanes with high selectivity	LDPE/HDPE/PP PE with $\text{C}_4\text{-C}_6$ paraffins	Improved monomer recovery up to 23.6 wt% 100% PE conversion with $\text{C}_6$ to $\text{C}_{10}$ gasoline-range liquid alkanes (126 wt%), gaseous isobutane (48 wt%), and $\text{C}_{11}$ to $\text{C}_{36}$ large alkanes (27 wt%)	10 44
Pt/ $\text{WO}_3/\text{ZrO}_2$ and HY zeolite systems for hydrocracking of mixed polyolefins to fuels with high selectivity	PE/PP/PS	Gasoline- and diesel-range hydrocarbons with a high conversion yield of 60 to 85 wt%	45
Microwave-assisted catalytic cracking over metal-free hierarchical zeolites at moderately high temperatures with short reaction times	LDPE containing 11 different additives	$\text{C}_3\text{-C}_8$ paraffins with >90% conversion for most additives	46
Polyolefin depolymerisation using the chlorine tolerant chloroaluminate ionic liquid for chlorine/PVC-containing mixtures <i>via</i> tandem cracking-alkylation	PP/PVC	Liquid hydrocarbons $\text{C}_4\text{-C}_7$ (35.5 wt%), $\text{C}_8\text{-C}_{12}$ (36.4 wt%) and $\text{C}_{13+}$ (28.1 wt%)	47
Metal-acid bifunctional catalytic system ( <i>e.g.</i> $\text{MoS}_x\text{-HBeta}$ ) for hydrocracking of heteroatom-containing mixtures	LDPE/HDPE/PP/PS with and without added heteroatom polymers such as TPU, PC, PLA and PVC	Liquid hydrocarbons (71% or 81% without heteroatom polymers), mainly $\text{C}_4\text{-C}_{16}$ iso-alkanes from PE and PP, and a mixture of alkylbenzenes and alkanes from PS	48

As such, catalytic cracking of 11 different additive-containing LDPE mixtures using a HY solid-acid catalyst at 370 °C yields primarily  $\text{C}_3\text{-C}_8$  paraffins with a high conversion of >90% for all additives except Doverphos® 213 and Drometrisole with a conversion of >80%. The effect of small molecule impurities on the pyrolysis product of mixed polyolefins was also studied by Genuino *et al.*<sup>51</sup>, who applied washing pre-treatment on a complex post-consumer plastic mixture (DKR-350) comprising PE, PP, PS, PET, clogged materials, multilayer flexibles, and a significant amount of organic and inorganic residues. Washing pre-treatment reduced the amount of solid and gaseous products attributable to the removal of organic impurities (such as sugars, proteins and fatty acids) and metal impurities that acted as the source and catalyst for gas formation but had minimal effect on the yield of pyrolysis oil or wax despite effectively lowering both organic and inorganic contents.<sup>50</sup> Monomers such as ethylene, propylene and styrene were recovered from the pyrolysis of the polyolefin mixture DKR-350 and could potentially be repolymerised to PE, PP and PS.

Other than organic additives and non-plastic contaminants, PVC can be highly detrimental to the thermal cracking process of post-consumer polyolefin waste due to the liberation of corrosive HCl gas. Conventionally, PVC has to be isolated from mixed plastic waste streams and separately pre-treated by dechlorination followed by deconstruction *via* conventional processes.<sup>52</sup> Recently, a novel process introduced the utilisation of chlorinated contaminants to facilitate polyolefin depolymerisation. Polyolefins could be deconstructed *via* a tandem cracking-alkylation process by using a Lewis acidic chloroaluminate



**Fig. 4** Advancements in cracking involving PVC at low temperatures, adapted with permission.<sup>44</sup> Copyright 2023 AAAS.

IL ( $[\text{C}_4\text{Py}]\text{Cl-AlCl}_3$ , dissolved in  $\text{CH}_2\text{Cl}_2$ ) (Fig. 4).<sup>44</sup> The dechlorination process of *tert*-butyl chloride forms a *tert*-butylcarbenium ion that is capable of abstracting a hydride from the polyolefin, deconstructing aliphatic hydrocarbon-based polymers to form small alkenes. Notably, unlike traditional thermal cracking processes, this process occurs at significantly lower temperatures ( $\sim 70$  °C). Likewise, PVC de-chlorination could be incorporated into polyolefin cracking processes with a similar ionic liquid medium, utilising the reactive intermediate generated from PVC de-chlorination as a hydride abstractor to react with aliphatic hydrocarbon-based polymers like PP and form carbenium for deconstruction into small alkenes at room temperature.<sup>47</sup> The PVC de-chlorination process could be further exploited for site-specific PET hydrolysis (Fig. 4)<sup>53</sup>

where the HCl by-product could be “stored” within the IL medium to assist in PET hydrolysis into TPA and dichloroethane. Although not demonstrated, the TPA could potentially be recovered to produce rPET while the leftover dPVC could be subjected to Ru catalysed hydrocracking<sup>54</sup> with up to 83.7% carbon recovery, through a reaction akin to that of PE hydrocracking.<sup>55</sup> Furthermore, the highly unsaturated dPVC residues could be subjected to intramolecular cyclisation or intermolecular crosslinking reactions to either form carbonaceous material with a capacitance value of  $399 \text{ F g}^{-1}$  (at  $1.0 \text{ A g}^{-1}$ ),<sup>56</sup> or porous materials for  $\text{CO}_2$  absorption with an uptake capacity of  $7.29 \text{ mmol g}^{-1}$  (32.1 wt%) and  $5.16 \text{ mmol g}^{-1}$  (22.7 wt%) at 0 and 25 °C, respectively.<sup>57</sup> Finally, dPVC cracking reactions at extremely high temperatures (>550 °C) also allow for the formation of monocyclic aromatic hydrocarbons.<sup>58</sup> While these studies demonstrate the possibility of recycling PVC-containing mixed plastic waste without any form of pre-treatment, difficulties in the handling of water sensitivity, recovery costs of ionic liquids and the use of toxic solvents need to be considered before actual implementation.

Besides pyrolysis, hydrocracking is another form of catalytic cracking that occurs in the presence of hydrogen gas and typically involves a bifunctional catalytic system mixed by metal sites and acid sites.<sup>59,60</sup> Due to its exothermic nature, hydrocracking could be performed at considerably lower temperatures than pyrolysis, especially for plastics with an inert hydrocarbon-only backbone (such as PE, PP, and PS).<sup>61</sup> Whilst not strictly considered a recycling process as the saturated hydrocarbon products cannot be repolymerised, these metal–acid bifunctional catalytic hydrocracking systems have been shown to be suitable for mixed plastics feedstock, having been demonstrated on a combination of different polymer resin types to mimic real-life LDPE/PP waste streams. For instance, a mixture of LDPE, HDPE, PP and PS with and without added heteroatom polymers such as TPU, PC, PLA and PVC was used to mimic municipal mixed plastic waste. Subjecting the mixtures to 250 °C and 20–30 bar  $\text{H}_2$  with a  $\text{MoS}_x\text{-HBeta}$  catalyst yielded liquid hydrocarbons with high yields of 71% and 81% respectively.<sup>48</sup> Hydrocracking products of PE, PP and PS were mainly iso-alkanes ( $\text{C}_4\text{-C}_{16}$ ) or a mixture of alkylbenzenes and alkanes respectively. The catalyst demonstrated its effectiveness in handling mixed plastic feedstocks by exhibiting tolerance towards heteroatom-containing contaminants and polymer mixtures, which are prevalent in mixed plastic waste streams. As such, a mixture of polyolefins and common heteroatom-containing polymers (PVC, PU, PC or PLA) was successfully converted to small molecules with this same process. Other catalytic systems such as  $\text{Pt/WO}_3/\text{ZrO}_2$  and HY zeolites were also found to be effective at catalytic hydrocracking of PP, PE and PS-containing waste plastic mixtures, providing gasoline- and diesel-range hydrocarbons at a high conversion yield (60 to 85 wt%).<sup>45</sup>

### 2.3 Assessment

Chemical recycling of plastics is technically viable for mixed plastic waste streams and has been implemented on industrial

scales. Site-specific chemical depolymerisation shows selectivity to certain chemical bonds while being tolerant to other polymers especially those containing saturated C–C backbones. This was clearly demonstrated by their ability to process complex, tightly woven mixed textile feedstock containing multiple hydrolysable polymers such as PET, nylons or spandex in the presence of different coloured dyes.<sup>29,30</sup> The monomers/oligomers from the depolymerisation processes could be isolated, purified and recirculated for repolymerisation. Comparatively, the advantage of thermal cracking processes (e.g. pyrolysis) lies in its generality to almost all forms of commodity plastics, including polyolefins. Although product selectivity is often poor, judicious catalyst design can aid in tuning product distributions and improving yields of desired products, including higher-value gasoline- and diesel-range hydrocarbons. While metal catalysts can be susceptible to deactivation by heteroatom-containing contaminants, these could be overcome either through process control and design (e.g. *via* microwave reactions) or pretreatments to afford desirable hydrocarbon product fractions. The challenging-to-recycle PVC could also be deconstructed with ILs under mild conditions whereby the liberated Cl heteroatoms could conveniently assist in the degradation of other plastics *via* a tandem cracking–alkylation process.<sup>47</sup>

Although these advancements in chemical recycling demonstrate some competency for handling mixed plastic feedstock, the complexity of municipal solid waste remains a tremendous hurdle.<sup>62</sup> With the need for pretreatments that add cost, chemical recycling of plastics from complex mixtures faces stiff cost competition from virgin plastics. It is thus important that techno-economic analyses (TEAs) be performed to assess the economic viability of these chemical recycling processes *via* a cost-benefit comparison, focusing mainly on economic viability through cost, revenue and risk analysis. This comprises factors such as raw materials, energy consumption, and operational and maintenance costs. For example, Aguado *et al.*<sup>26</sup> compared the economic viability of two chemical depolymerisation methods for PET packaging waste—hydrolysis and glycolysis—considering costs required for energy usage and waste management. In a pilot plant with a 200-ton annual capacity for waste PET, the economic costs were found to be 2.71 € per kg for hydrolysis (to form TPA) and 4.23 € per kg for glycolysis (to form BHET). The primary cost drivers for both processes were identified to be excessive solvent use in alkaline hydrolysis and waste management in glycolysis. After identifying the main costs, a scaled-up process, up to an 8000-ton per year plant with appropriate process optimisations, reduced the production cost to 1.02 € per kg for hydrolysis and 1.99 € per kg for glycolysis. Secondly, Andini *et al.*<sup>30</sup> highlighted the intricate relationship between product sales and the economic viability of microwave-assisted glycolysis for mixed textile waste *via* TEA. The analysis uses a profitability index as a key metric, with a profitability index of 0.95 and 1.29 for low and high product sales respectively. This study suggested the importance of the economies of scale of the expected product in reducing production costs and gener-

ating profits. Finally, in addition to the expected product from waste plastic valorisation, isolatable side products could also be valuable and may tip the cost–revenue scale of the technology. For instance, mixed textile waste contains valuable materials such as cotton, which could potentially be isolated after chemical deconstruction of PET. Wu *et al.*<sup>31</sup> demonstrated a remarkable profit gain because of the non-destructive cotton recovery after PET hydrolysis, driving the cost–revenue scale in favour of the otherwise less profitable chemical recycling of PET to TPA.

### 3. Chemical upcycling

Chemical upcycling focuses on generating products of higher economic value from waste plastic feedstock. The value of these products can arise from a combination of high industrial demand and/or product market value (*e.g.* specialty chemicals). Spurred by the diverse possibilities, recent years have witnessed the emergence of novel chemical upcycling processes to convert low-value commodity plastics into higher value chemicals or functional materials.<sup>63–71</sup> Although most upcycling technologies are currently lower on the technology readiness scale compared to recycling, upcycling is a viable complement to recycling for addressing plastics' end-of-life. Like recycling, many upcycling strategies developed thus far are specific to specific types of plastic feedstock. This section will focus on chemical upcycling strategies suitable for mixed plastics for the production of either industrially relevant chemicals (summarised in Table 3) or functional materials (summarised in Table 4).

#### 3.1 Chemical upcycling to industrially relevant chemicals

In upcycling by depolymerisation, plastics are converted into chemicals different from their monomers. For example, instead of recovering the styrene monomer from PS, the polymer can be converted through oxidative depolymerisation reactions into oxygenated compounds, such as benzoic acid,

that are industrially relevant in food preservation, cosmetics and pharmaceuticals.<sup>65,66,72,73</sup> Most upcycling methods currently developed are designed for specific types of plastics, with reactions selective to the targeted polymer's chemical reactivity in order to obtain specific degradation products. However, while not explicitly mentioned, such processes often demonstrate potential for a mixed plastic system, despite the need for further product separation procedures. This section will focus on oxidative degradation processes (either thermal- or photo-assisted) that introduce new heteroatoms and functionalities into the small molecule chemicals derived from the “*Big Six*”.

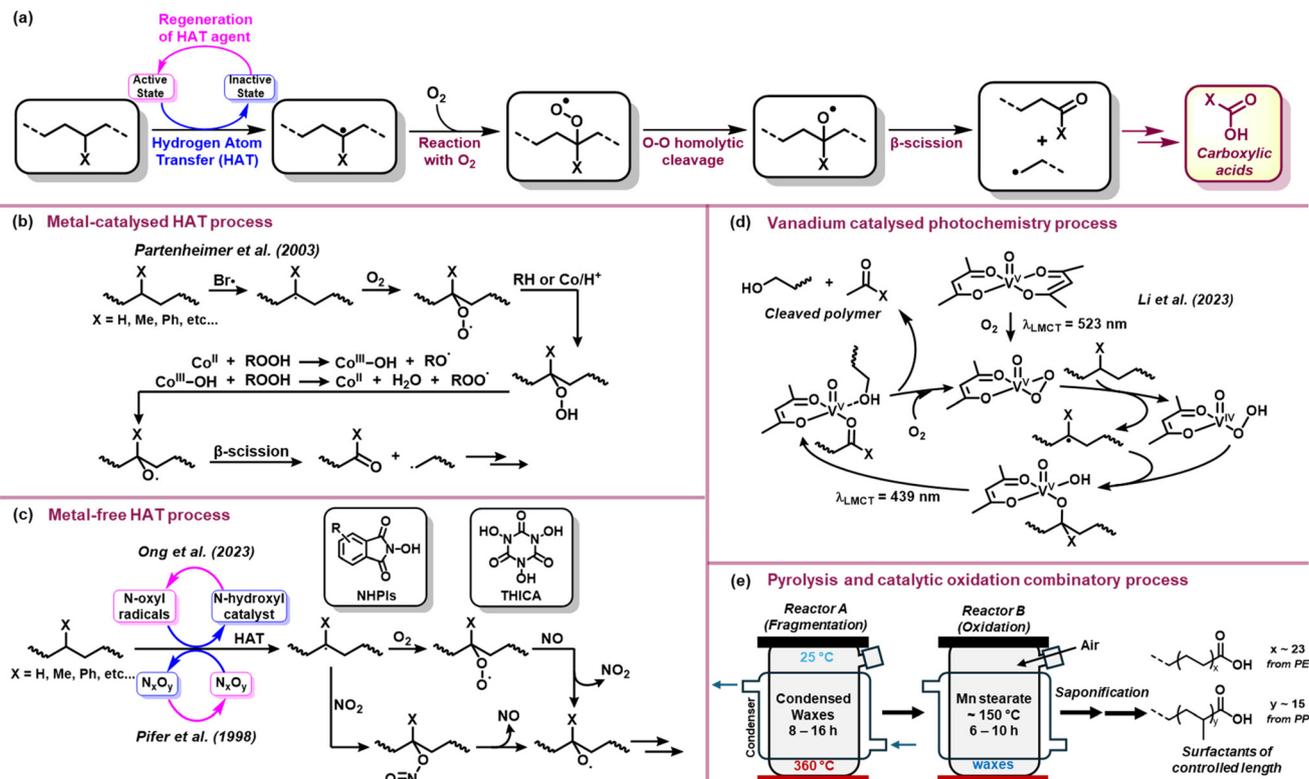
Oxidative degradation takes place in the presence of an oxidant, which can be O<sub>2</sub> from air – a desirable choice due to its abundance, cost-effectiveness (can be practically free), and the absence of stoichiometric organic waste generation typically encountered with the usage of organic oxidants. When applied to plastics with saturated C–C backbones, degradation occurs *via* radical-based reaction cascades involving reactive oxygen intermediates, typically initiated by a hydrogen atom transfer (HAT) agent which abstracts hydrogen atoms to form carbon-centred radicals on the polymer backbone. These radicals can be trapped by O<sub>2</sub> to form a peroxy radical species, allowing further reaction propagation through homolytic O–O cleavage to form oxo-radicals and/or  $\beta$ -scissions that result in C–C cleavage, ultimately forming the final product (often carboxylic acids)<sup>74</sup> (Fig. 5a). The application of oxidative degradation on plastics could be traced back to 1960,<sup>75</sup> when PE was oxidised into acids and esters using metal catalysts. In modern iterations, the catalytic system typically employs a bromide source such as NaBr as a precursor to the Br radical that acts as a potent HAT agent responsible for initiating the first radical formation (Fig. 5b), as well as transition metal salts such as Co(OAc)<sub>2</sub> or Mn(OAc)<sub>2</sub>, which enables constant regeneration of the Br radicals and facilitates homolytic O–O cleavage (Fig. 5b).<sup>76</sup> While the predecessor of this system was only applied on PEs, Partenheimer<sup>77</sup> demonstrated the viability of the Co/Mn/Br catalytic system on all the “*Big Six*”, along with

**Table 3** Chemical upcycling of mixed plastic feedstock to industrially relevant chemicals

Method	Catalyst system	Feedstock	Product	Ref.
Thermal oxidative degradation	NaBr, Co(OAc) <sub>2</sub> , Mn(OAc) <sub>2</sub> or Zr(OAc) <sub>4</sub> with V	PE, PP, PS, PVC, P4VP, P(2VP-co-S), PBT, PEN, PC	Oxidised forms of monomers	77
	N <sub>x</sub> O <sub>y</sub> , high temperature and pressure	HDPE, LDPE, PP, PS, PMMA, PAA or nylon-6,6	Oxidised products PS with nitrated aromatic acids	80
Photo-oxidative degradation	N <sub>x</sub> O <sub>y</sub> with the <i>N</i> -hydroxyl catalyst, 120 °C, air	PS, P4MS, PSSA, polyindanes	PS → Benzoic acid and 4-nitrobenzoic acid P4MS → TPA PSSA → 4-sulfobenzoic acid Polyindane → phthalic acid/phthalic anhydride $\beta$ -Keto adipate, PHA	65 and 82
	NHPI with the Co/Mn co-catalyst → <i>Pseudomonas putida</i>	HDPE, PS, PET or HDPE/PS/PET		83
Tandem pyrolysis–oxidation process	VO(acac) <sub>2</sub> under photoirradiation	LDPE, HDPE, PS, PP, PVC and other co-polymers	Carboxylic acids	78
CBP biological–chemical cascade upcycling process	Temperature-controlled reactor setup	PE or PP	Long-chain aliphatic carboxylic acids or esters and surfactants	79
	<i>Ideonella sakaiensis</i> 201-F6	PET	TPA, EG	84

**Table 4** Functional materials derived from mixed plastic feedstock and their properties

Functional materials	Feedstock	Material properties and possible applications	Ref.	
Vitrimers	HDPE/PP	<ul style="list-style-type: none"> <li>• Associative transesterification between ester and hydroxyl groups enables dynamic bonding</li> <li>• Improved tensile strength and stable crystallinity, with minimal deterioration after multiple processing cycles</li> <li>• More robust, with higher activation energy (124 kJ mol<sup>-1</sup>) than other vitrimer systems</li> </ul>	90	
	LDPE/HDPE/PP	<ul style="list-style-type: none"> <li>• Reversible siloxane bond exchange enables adaptable covalent bonding</li> <li>• Strong crosslinked network with enhanced tensile strength and full melt reprocessability</li> </ul>	91	
	LDPE/PP	<ul style="list-style-type: none"> <li>• Perfluorophenylazide-based nitrene crosslinkers introduce disulfide, imine, and acetal dynamic covalent bonds</li> <li>• Improved tensile strength with higher elongation at break compared to precursor plastics of the same blend</li> </ul>	92	
Carbon materials	Activated carbon	HDPE/LDPE/PP/PS/PMMA	<ul style="list-style-type: none"> <li>• Reversible vitrimer <i>via</i> carbene C–H insertion in thermoplastics</li> <li>• Mechanical properties (elongation and tensile toughness) enhanced up to 8 times through reversible crosslinking</li> </ul>	89
		LDPE/HDPE/PP/PS/PET	<ul style="list-style-type: none"> <li>• Fully microporous structure with a CO<sub>2</sub> uptake of 1.58–2.88 mmol g<sup>-1</sup> under equilibrium conditions</li> <li>• Stable adsorption and high regeneration efficiency (&gt;96%) over 10 cycles, with a total CO<sub>2</sub> uptake of &gt;14 mmol g<sup>-1</sup></li> </ul>	94
	Porous carbon nanosheets	LDPE/HDPE/PP/PVC/PET/PS	<ul style="list-style-type: none"> <li>• Highly microporous carbon with the maximum CO<sub>2</sub> uptake of 2.7 mmol g<sup>-1</sup> at 298 K and 1 bar</li> <li>• Fast adsorption rate, strong CO<sub>2</sub> selectivity over N<sub>2</sub> and good cycling performance</li> </ul>	95
		PE/PP/PS/PET/PVC	<ul style="list-style-type: none"> <li>• High specific surface area (1734 m<sup>2</sup> g<sup>-1</sup>) and large pore volume (2.441 cm<sup>3</sup> g<sup>-1</sup>)</li> <li>• Excellent gas adsorption: CO<sub>2</sub> uptake (up to 18.00 mmol g<sup>-1</sup>) and H<sub>2</sub> uptake (up to 5.2 mmol g<sup>-1</sup> at 45 bar), outperforming conventional carbon materials</li> <li>• Graphitic layers and hierarchical micro-/mesopores drive strong electrochemical performance</li> </ul>	96
		PE/PS	<ul style="list-style-type: none"> <li>• High-performance supercapacitors: energy density up to 30.4 Wh kg<sup>-1</sup> (at 0.1 A g<sup>-1</sup>), specific capacitance up to 207 F g<sup>-1</sup> (aqueous) and 120 F g<sup>-1</sup> (organic) at 0.2 A g<sup>-1</sup>, retains 72–79% capacitance at 10 A g<sup>-1</sup></li> <li>• Superior to commercial activated carbon across all tested electrolytes</li> <li>• Graphene oxide additive increases carbon yield by up to 250% and improves crystallite size</li> <li>• Produce graphitic carbons with lamellar morphology and high structural order</li> <li>• Suitable for energy storage applications, particularly as lithium-ion battery anode materials</li> </ul>	98
	Graphite	PE/PS	<ul style="list-style-type: none"> <li>• Cylindrical multi-walled CNTs formed from carbon deposition and crystallisation</li> <li>• High hydrogen yield (up to 44.1 mmol g<sup>-1</sup> plastic), 2–3× higher than traditional thermal methods</li> </ul>	99
	CNTs	LDPE/HDPE/PP	<ul style="list-style-type: none"> <li>• Pyrolysis–catalysis mainly produces multi-walled CNTs (12–25 nm diameter), with the Ni–Fe/MCM-41 catalyst affording high-quality CNTs due to the high surface area and mesoporosity</li> <li>• Hydrogen by-product (up to 38.10 mmol g<sup>-1</sup> plastic) usable as process fuel</li> </ul>	100
Carbon dots	PE/PP	<ul style="list-style-type: none"> <li>• Blue-fluorescent carbon dots (1–8 nm) with excitation-dependent photoluminescence</li> <li>• High solubility in water and thermal/photo stability</li> <li>• Compatible with flexible polymers for tunable photoluminescence applications like anti-counterfeiting and optoelectronics</li> </ul>	102	
Membranes	Oleophilic sorbent	LDPE/HDPE/PP	<ul style="list-style-type: none"> <li>• Highly porous structure with 600 cavities per cm<sup>2</sup> (0.5–200 μm in size); each cavity can expand up to 20× its thickness</li> <li>• Sponge-like swelling behaviour, with an oil uptake capacity of 70–140 g g<sup>-1</sup></li> <li>• Outperforms commercial sorbents in oil absorption and recovery</li> </ul>	104
	BP-GS	LDPE/HDPE/PP/PS	<ul style="list-style-type: none"> <li>• Blended plastic substrate with superabsorbent properties (high water absorption capacity ~1188%)</li> <li>• Porous, uniform structure supported by stable covalent bonding with a superabsorbent polymer and a polyolefin elastomer to create a hydrophilic, flexible material</li> <li>• Supports plant growth in drought or slope conditions, promotes micro-ecosystem formation ⇒ potential use in vertical greening, landscaping, and slope restoration</li> </ul>	106
	Zwitterionic hydrogel	LDPE/PP/PVC	<ul style="list-style-type: none"> <li>• Plastic-grafted hydrogel with zwitterionic properties for heavy metal removal</li> <li>• Simultaneously adsorbs cationic and anionic heavy metals in acidic and alkaline environments</li> <li>• Max. adsorption capacities: Pb<sup>2+</sup> (132.13 mg g<sup>-1</sup>), Cd<sup>2+</sup> (85.58 mg g<sup>-1</sup>), Ba<sup>2+</sup> (69.92 mg g<sup>-1</sup>), and Cr(vi) (85.15 mg g<sup>-1</sup>)</li> <li>• Strong mechanical strength, reusability, and structural stability</li> </ul>	107
MOFs	UiO-66, MIL-53(Al)	PE/PET/PS/PEF	<ul style="list-style-type: none"> <li>• Excellent porosity and crystallinity, comparable properties to MOFs from commercial feedstocks</li> <li>• Applicable in energy storage, separation, and catalysis</li> </ul>	108



**Fig. 5** Upcycling of mixed plastics into relevant chemicals *via* oxidative degradation. (a) General reaction progress for oxidative degradation of polymers. (b) Metal catalytic oxidative degradation of plastics, adapted with permission.<sup>77</sup> Copyright 2003 Elsevier. (c) Metal-free catalytic oxidative degradation of plastics, adapted with permission.<sup>65</sup> Copyright 2023 American Chemical Society. (d) Proposed mechanism for VO(acac)<sub>2</sub>-catalysed photo-oxidation of plastics, adapted with permission.<sup>78</sup> Copyright 2023 Elsevier. (e) Tandem pyrolysis–oxidation combinatory process to generate surfactants, adapted with permission.<sup>79</sup> Copyright 2023 Science.

other derivatives and co-polymers such as poly(4-vinyl-pyridine), poly(2-vinylpyridine-*co*-styrene), poly(butylene terephthalate), poly(ethylene naphthalate) and PC, yielding the oxidised forms of various monomers where applicable. This study was particularly important as it highlighted the possibility of a *one-system-multiple-reaction* concept, which is highly relevant for mixed plastics. The efficiency of this catalytic system was also further improved when incorporating additives such as Zr(OAc)<sub>4</sub> or changing the Co/Mn metal combination to V.<sup>77</sup> Despite the efficiency of the Co/Mn/Br system, it is disadvantaged by its need for heavy metals, chlorinated solvents and relatively harsh conditions (temperatures up to 220 °C, pressures up to 70 bar).

To avoid usage of heavy metal catalysts, Pifer *et al.*<sup>80</sup> first introduced the possibility of performing uncatalysed oxidative degradation on different plastic polymers (Fig. 5c). In this metal-free system, N<sub>x</sub>O<sub>y</sub> species acts as the HAT agent at high temperature and pressure. Common municipal plastics including HDPE, LDPE, PP, PS, PMMA, PAA and nylon-6,6 were all shown to be successfully degraded into their respective oxidised products, though each plastic was reacted individually. PS was particularly interesting, giving rise to a number of nitrated aromatic acids on top of the benzoic acid product, implying the occurrence of certain nitration processes under

these reaction conditions. While the use of nitrogen oxides successfully mitigates the need for heavy metal catalysts, the reaction conditions are still relatively harsh, requiring the use of pressurised vessels.

More recently, milder organocatalytic procedures were developed, involving *N*-hydroxyl compounds such as *N,N,N'*-trihydroxyisocyanuric acid (THICA) or *N*-hydroxyphthalimides (NHPis) that have demonstrated efficacy in various oxidative processes including the oxidative degradation of plastics.<sup>81</sup> These organocatalysts are precursors to *N*-oxyl radicals, known for their effectiveness in HAT reactions, and can operate effectively in the absence of metallic additives. Together with the availability of N<sub>x</sub>O<sub>y</sub> *via* thermal decomposition of HNO<sub>3</sub>, the *N*-hydroxyl catalytic system can oxidatively upcycle PS to benzoic acid.<sup>65</sup> N<sub>x</sub>O<sub>y</sub> was proposed to be multifunctional as (1) it assists in the regeneration of the active *N*-oxyl radical species, (2) participates in a secondary pathway that enables accelerated formation of the oxo-radical species, and (3) assists in the homolytic O–O cleavage of the peroxy radical species (Fig. 5c). The combination of the *N*-hydroxyl catalyst and N<sub>x</sub>O<sub>y</sub> successfully lowered the required degradation conditions to 120 °C under open air conditions. Because of its milder conditions as compared to that used by Pifer, the undesirable production of multiple nitrated aromatic acids was suppressed to

selectively produce only 4-nitrobenzoic acid as the minor side-product besides benzoic acid, with both products easily isolated from the crude reaction by hot water recrystallisation. Furthermore, besides high tolerance towards common additives such as plasticisers, coloured dyes and large amounts of metallic contaminants (such as Fe), the process also demonstrated applicability for other common PS derivatives such as poly(4-methylstyrene) and polystyrene-sulfonic acid, effectively converting them to TPA and 4-sulfobenzoic acid respectively. Recently, the same catalytic system was shown to be effective in a two-step sequential process to convert PS into phthalic acid and phthalic anhydride *via* a polyindane intermediate obtained from PS backbone rearrangement.<sup>82</sup> This method was also shown to convert poly(4-methylstyrene) into aromatic diacids (terephthalic acid and isophthalic acid) and triacids (mellitic acid), showing the potential to be used on upcycling mixtures of PS and related polymers which can be difficult to separate from each other.

Finally, photocatalytic reactions provide a relatively milder pathway for plastic degradation, involving only the need for irradiation without external heating. While most photoredox oxidative degradation was demonstrated using PS, VO(acac)<sub>2</sub> was shown to catalytically degrade a wide range of common plastic types, including LDPE, HDPE, PS, PP, PVC and other co-polymers into organic acids under photoirradiation.<sup>78</sup> To understand the mechanism behind the photocatalytic conversion, the authors monitored the reaction progress by UV-vis spectroscopy using PS as the polymer feedstock. Thorough analysis of the time-lapse spectra suggested an oxidation of V species from V<sup>IV</sup> to V<sup>V</sup>, along with the presence of V<sup>V</sup> peroxide intermediates. A time-dependent electron paramagnetic resonance spectroscopy analysis also displayed the signal for the V<sup>IV</sup> species, which completely disappeared after 5 h of photoirradiation, further supporting the conversion of the catalyst from V<sup>IV</sup> to V<sup>V</sup> species. As shown in Fig. 5d, the generation of the V<sup>V</sup> peroxide intermediates can be photoexcited by ligand-to-metal charge transfer followed by a HAT reaction to generate a radical on the polymer backbone. This radical can then be trapped by the V<sup>IV</sup> intermediate, which undergoes photoexcitation again to facilitate C–C cleavage of the polymer backbone. Eventually, the polymer will be deconstructed into the corresponding carboxylic acids.

Mixed polyolefin plastics (PE and PP) can also be converted into long-chain aliphatic carboxylic acids *via* a two-step combinatory process of pyrolysis and catalytic oxidation. To achieve this, Xu *et al.*<sup>79</sup> applied a two-step combinatory process using a temperature-controlled reactor setup (Fig. 5e). First, polymer fragmentation *via* pyrolysis was performed in Reactor A with the reactor base maintained at 360 °C. A cooling system was employed around the reactor body, which allows for an ~25 °C environment at the reactor lid. Because of the controlled temperature gradient, the authors could collect hydrocarbon fragments of desired molecular weights on the inner reactor walls. After which, the obtained hydrocarbon fragments were subjected to oxidation in Reactor B to yield the aliphatic carboxylic acids or esters of corresponding molecular weights, which

could either be used as starting materials in polyesterification or subjected to saponification to produce surfactants with twice the market value of virgin plastics.<sup>79</sup>

In addition to purely chemical approaches, combining chemical and bio-catalysis through cascade reactions can bring about complex, multistep chemical transformations that are challenging to achieve through either process alone.<sup>49</sup> Harnessing biocatalytic processes to achieve product diversification, which typically occur under mild, aqueous conditions, can result in environmental benefits compared to a purely chemical approach. Du *et al.* employed a hydrolase to first depolymerise PET, whereupon the ethylene glycol produced can be converted to glycolate electrochemically under ambient conditions, with an overall product yield of 92.6%.<sup>85</sup> Compared with traditional electrochemical technology, this approach offers a more cost effective and lower carbon pathway by removing the heavy reliance on alkaline chemical pretreatment of PET. While the effectiveness of enzymatic degradation of plastics other than polyesters remains controversial,<sup>86</sup> chemical depolymerisation can first occur on mixed plastics, followed by bio-funnelling of the unseparated product mixture through a biological system to produce high-value chemicals. For instance, this approach was demonstrated by Sullivan *et al.*,<sup>83</sup> who combined NHPI with a Co/Mn co-catalyst to first degrade HDPE, PS, and PET, both individually and as a mixture, into smaller molecules which were bio-funnelled using *Pseudomonas putida* to produce β-ketoadipate or PHA. This tandem chemical and biological funnelling process was particularly interesting as it omits the need for specific product separation after oxidative degradation of such mixed plastic feedstock. Despite its potential, it should also be noted that biocatalytic systems can be sensitive to the reaction composition and environment (*e.g.* pH). Additives, plasticisers, contaminants and even the plastic degradation products can be antagonistic to biodegradation, either through influencing feedstock susceptibility to breakdown<sup>87</sup> (*e.g. via* polymer packing and crystallinity) or potentially directly influencing biocatalytic activity. For the latter, PET degradation products such as mono(2-hydroxyethyl) terephthalate can inhibit PET hydrolases' activity,<sup>88</sup> while amphiphilic molecules such as long-chain aliphatic acids that possess surfactant-like properties can potentially disrupt cell membranes.

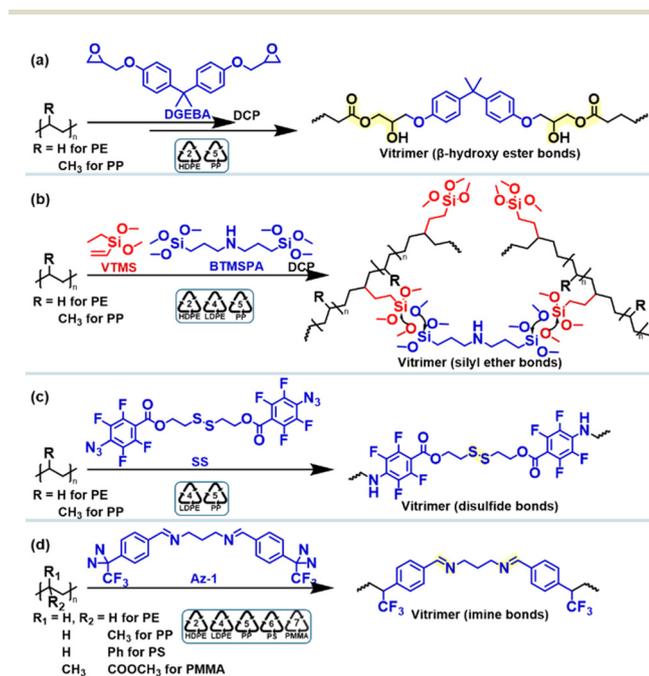
### 3.2 Chemical upcycling to functional materials

Besides chemical upcycling of plastic mixtures to small molecule chemicals, post-use plastics can also be converted into high-value functional materials such as vitrimers, membranes and different carbonised materials. Similarly, many previous studies have focused on homogeneous plastic streams, allowing for targeted chemical modifications based on the individual plastic's reactivity. In this aspect, mixed plastics presents unique challenges due to differences in chemical reactivities and physical incompatibilities, making it difficult to develop a single chemical process capable of treating mixed plastics efficiently. However, recent advancements demonstrate promising strategies for overcoming these hurdles.

Vitrimerers are a class of crosslinked polymers that combine the mechanical robustness of thermosets and the reprocessability of thermoplastics. They are formed by the inclusion of reversible cross-linkers into polymers, resulting in a cross-linked polymeric material with dynamic covalent bonding. Although crosslinking of different polymers might pose a problem due to varying chemical reactivity, a vitrimer made from a mixed plastic feedstock such as those from polyolefin mixtures (HDPE, LDPE and PP) would technically be feasible due to the parent polymers' similar chemical properties. Indeed, several groups have separately demonstrated the formation of vitrimer materials using polyolefin mixtures (PE and PP) as the feedstock, most commonly *via* radical-based activation approaches.<sup>89–91</sup> The method generates a radical on the inert polyolefin backbone using dicumyl peroxide (DCP), which then facilitates the attachment of crosslinking handles. After which, different crosslinking reactions through transesterification (Fig. 6a), silyl ether (Fig. 6b) and siloxane chemistry can be performed. A different approach, using reactive nitrene intermediates that can insert into the polyolefin backbone directly, was shown to crosslink polyolefins *via* thermal activation. A perfluorophenylazide-based crosslinker with dynamic disulfide bonds that can form reactive singlet nitrene species (Fig. 6c) was able to crosslink PP and LDPE to form vitrimers that displayed enhanced mechanical strength but lower Young's modulus.<sup>92</sup> Furthermore, the materials were

able to be reprocessed for several cycles, exhibiting thermo-plastic-like properties. Due to their similarity to the parent polymer, these vitrimers could also be used as compatibilisers to enhance the properties of a mixed plastic blend<sup>89</sup> (*vide infra*). Importantly, this process has been successfully applied to a mixture of thermoplastic olefins without prior sorting, demonstrating the possibility of using a mixed plastic feedstock for vitrimer production. Additionally, Png *et al.* extended this concept to a broader plastic mix, incorporating not only the typical polyolefin mixture (PE, PP), but also PS and PMMA, two other polymers with markedly different chemical reactivities. In this case, the crosslinking handle was attached *via* carbene-based chemistry to a carbene moiety, followed by crosslinking with an imine-based crosslinker (Fig. 6d). While the exact location of the crosslinking is unknown, the vitrimer produced exhibited enhanced mechanical properties with thermal stability, as well as reprocessability. This work further highlights the possibilities of converting mixed plastic feedstock into reprocessable polymers with potential applications in structural materials, adhesives, electronics and energy storage. Other than the production of liquid or gaseous hydrocarbons, pyrolysis of mixed plastics can also be exploited to selectively produce solid carbonised materials. This process of carbonisation allows for various carbon-rich products like carbon nanotubes, nanosheets and graphite to be produced for various applications such as CO<sub>2</sub> capture or energy storage. Like pyrolysis, carbonisation is applicable to a variety of plastics, having the potential for valorisation of a mixed waste plastic stream. Several groups<sup>94–96</sup> demonstrated the direct carbonisation of carbon-rich mixed waste plastics, followed by chemical activation *via* KOH impregnation to produce microporous carbon materials with CO<sub>2</sub>-affined functional groups. The resulting materials exhibited good stability, reusability and excellent CO<sub>2</sub> selectivity, with CO<sub>2</sub> absorption capabilities even outperforming the commercial carbon capture material, AC Norit R2030CO<sub>2</sub>,<sup>94</sup> by about 30%, highlighting the potential to chemically upcycle mixed waste plastics to carbon capture materials. Capitalising on the success of CO<sub>2</sub> capture materials, the group further demonstrated the excellent performance of these porous carbon materials as supercapacitor electrode materials, synthesised using the same method of carbonisation followed by KOH activation.<sup>97</sup> The porous structure within the material enhances ion transport while preventing nanosheet aggregation, boosting the capacitance and energy density. These structural advantages led to its exceptional supercapacitor performance, achieving specific capacitances of 207 F g<sup>-1</sup> (aqueous medium) and 120 F g<sup>-1</sup> (organic medium) at 0.2 A g<sup>-1</sup> and maintaining 72.5% and 79.2% capacitance retention in aqueous and organic electrolytes, respectively, even at high current densities. Compared to traditional thermally activated carbon and other carbon materials, these porous carbon materials derived from waste plastics exhibited high electrochemical performance, highlighting their potential for energy storage applications.

Graphite, comprising stacked layers of hexagonally arranged carbon atoms, is a carbon allotrope that has potential



**Fig. 6** Chemical upcycling of mixed polyolefins to vitrimers involving (a) dynamic  $\beta$ -hydroxyl ester bonds, adapted with permission under the CC-BY-3.0 license.<sup>90</sup> (b) Silyl ether exchange reaction, adapted with permission under the CC-BY 4.0 license.<sup>91</sup> Copyright 2024 American Chemical Society. (c) Dynamic disulfide bonds, adapted with permission.<sup>92</sup> Copyright 2025 Elsevier. (d) Dynamic imine bonds synthesised using carbene chemistry, adapted with permission.<sup>93</sup> Copyright 2024 Wiley.

applications in energy storage. A study using a mixture of carbon-rich PP, HDPE, PS and PET as feedstock for carbonisation was shown to provide a high-yield, high-quality and uniform synthetic graphite by incorporating graphene oxide (GO) as an additive.<sup>98</sup> The addition of GO allowed for improved solid carbon yield by assisting in radical combination reactions, exerting stabilising and diffusional confinement effects on the polymer radicals. Furthermore, GO also enhanced the quality of the graphitic material, endowing it with excellent crystallite sizes and nanostructures, which are desirable for energy storage materials. Other groups<sup>99,100</sup> further capitalised on the carbonisation of mixed plastic feedstock (namely PE, PP and PS) to produce CNT, which is one of the strongest materials in terms of tensile strength and elastic modulus. Furthermore, the full recyclability of produced CNT fibres was demonstrated without any loss in material properties, unlike the original plastic feedstock.<sup>101</sup> The process typically requires bimetallic Ni–Fe catalysts that dehydrogenate plastic polymers. After that, CNTs were obtained with yields of up to 55.6 wt%,<sup>100</sup> along with a significant amount of hydrogen as a by-product that could also be utilised as fuel. Finally, Abdelhameed *et al.*<sup>102</sup> introduced a hydrothermal process in HNO<sub>3</sub> for upcycling a mixture of PP and PE into blue-emissive carbon dots with a 96% yield. The carbon dots from this “top-down” synthesis approach exhibit good solubility in both aqueous and organic media, excitation-dependent emission and excellent thermal and photostability.

Materials from the upcycling of plastics also show considerable promise for environmental remediation applications. Their inert backbone structures, along with suitable chemical modifications, enable them to be repurposed as membranes or adsorbents for the removal of hazardous substances such as heavy metals, oil and organic dyes, which are increasingly becoming global environmental concerns.<sup>103</sup> For example, Saleem *et al.*<sup>104</sup> demonstrated the upcycling of mixed polyolefin (HDPE, LDPE and PP) into a bimodal super-oleophilic sorbent with an average of 600 cavities per cm<sup>2</sup> filled with 0.5 to 5 μm sized pores. Like sponges, these pore-filled cavities can expand twenty times their thickness with an oil uptake capacity of 70–140 g g<sup>-1</sup>, a result far superior to commercial sorbents such as Corksorb, 3M-HP-255, 3M-156 or Chemtex-BP-9 W. Furthermore, they can be used like a sponge to recover up to 97% of the oil, demonstrating its recyclability. Most importantly, the cavity-forming agent used was NaCl, a cheap and water-soluble additive easily removed after the upcycling process. In a further study,<sup>105</sup> the same authors demonstrated that pre-oxidation of the polyolefin mixture using KMnO<sub>4</sub> before pore induction could introduce oxygen moieties into the membrane, allowing for improved hydrophilicity and increased permeability. These studies offer a practical and sustainable solution for oil spill cleanup and other organic pollutant removal from water. Yue *et al.*<sup>106</sup> developed a universal process for converting different blended waste plastic mixtures consisting of PP, HDPE, LDPE and PS into flexible and hydrophilic “blending plastic-g-growing substrates” (BP-GS) with excellent porosity and uniformity. Polyolefin elastomers were

used as the reactive compatibiliser to thermally crosslink superabsorbent polymers with benzoyl peroxide. The inclusion of superabsorbent polymer crosslinks gave the material its water-absorbing capacity of up to 1188%, governed by a chemical absorption process. These materials have shown promise as growing substrates for vertical greening and slope restoration to support plant growth and environmental restoration. To further illustrate the versatility of mixed plastic blends as scaffolds for different absorbent materials, Yue *et al.*<sup>107</sup> utilised a PP, LDPE and PVC blend for the synthesis of zwitterionic adsorbents capable of absorbing both cationic and anionic heavy metals at a wide pH range of 3–9. Like the previous material, incompatibility between immiscible plastics was mitigated *via* a crosslinking process using sodium alginate and chitosan. These additives endowed the material with adsorption capacities for heavy metals such as Pb<sup>2+</sup>, Cd<sup>2+</sup>, Ba<sup>2+</sup>, and Cr<sup>4+</sup> *via* different processes such as electrostatic interaction, cation exchange and coordination interaction with functionalities containing Cl, N or O groups. Finally, the material also demonstrated excellent reusability for several adsorption–desorption cycles, making it a promising, low-cost solution for industrial wastewater treatment. Metal–organic frameworks (MOFs) are a unique class of porous materials comprising organic linkers bridging metal nodes, with excellent bottom-up designability for a vast range of applications such as gas capture, separation and catalysis. With industrial-scale MOF production underway, there is growing interest in producing these versatile materials from alternative feedstocks such as plastic waste.<sup>109–112</sup> Recently, Lim and coworkers demonstrated aromatic C–H activation as a new strategy to convert the unreactive terephthalate segment of PET into 2-hydroxyterephthalic acid (30–50% conversions), even in the presence of PET mixtures containing contaminants such as PE, PS and PEF.<sup>108</sup> Using real-life PET waste, the authors showed that the product mixture of 2-hydroxyterephthalic acid and unreacted terephthalic acid can be used to produce multivariate MOFs with mixed organic linkers (Fig. 7). The hydroxyl-

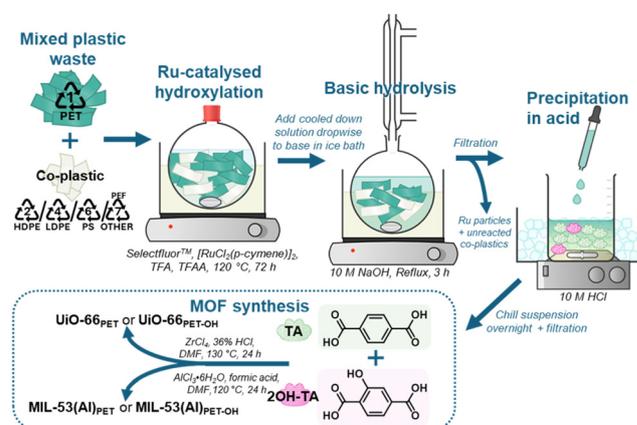


Fig. 7 Chemical upcycling of PET in mixed plastic waste to hydroxylated MOFs, adapted with permission.<sup>108</sup> Copyright 2025 American Chemical Society.

ated MOFs may be useful for applications including hydrogen adsorption, methane storage and increased conductivity for solid electrolytes.

### 3.3 Assessment

Although still largely restricted to lab scales, the chemical upcycling approaches discussed herein can be applied to heterogeneous plastic feedstock, including real-life plastic waste of different types. In one form of upcycling, plastics can be oxidatively depolymerised (section 3.1) to form valuable oxygenated aromatics such as benzoic acid, even in the presence of contaminants. Compared to thermolytic approaches for recycling such as pyrolysis, oxidative depolymerisation can occur under milder and less stringent conditions (with the possibility of an open-air system), even using photo-irradiation at near ambient temperatures, potentially suggesting the possibility of harvesting solar irradiation to drive upcycling processes. The other form of plastic upcycling involves utilising mixed plastic feedstock into functional materials with unique properties (section 3.2), as summarised in Table 4. Leveraging the robustness of recalcitrant plastics, they can be crosslinked with a properly designed “chemical bridge” and dynamic covalent bonding to form vitrimers, a class of materials that possess high strength and reprocessability desired for upcycled applications. Apart from that, the carbon-rich mixed plastic feedstock could also be carbonised selectively to form solid carbonised materials over the traditionally more favoured oil/wax, allowing potential applications in CO<sub>2</sub> capture and energy storage. Finally, mixed plastics can be upcycled into membranes which exhibit excellent properties that can surpass commercial sorbents for specialised applications, as well as mixed linker-containing multivariate MOFs.

Whilst promising, the full environmental impact and viability of these upcycling processes remain understudied. This can prompt life-cycle assessment (LCA), focusing on the environmental impact of technologies and assessing all stages of the life cycle, from cradle-to-grave. This can be done through environmental metrics such as carbon footprint, global warming potential (GWP), resource usage and effects on human and ecological health. Early LCA analyses of plastic upcycling processes are optimistic. In Li *et al.*'s assessment of pyrolysis oil hydroformylation for high-value chemical production on a large scale, they demonstrated that their multi-step separation process, which produced high-purity alcohol and diols, shows favourable economics at production scales of up to 100 000 tons of plastic per year, offering potential net profits of \$100 million annually and a payback period of just three years. Environmentally, the approach also reduces greenhouse gas emissions by 50% compared to plastic incineration and by 60% compared to traditional petroleum-based chemical production. The technology by Zhang *et al.*<sup>113</sup> to upcycle waste plastics (PP, PET, and PE) into methane using single-atom catalysis has also suggested overall positive environmental benefits with an energy efficiency of 92%, producing 896.1 kg of methane, 187.2 kg of water, and 60.9 kg of coke from 1 ton of waste plastic feedstock. This gave a GWP of just

0.26 kg and 0.4 kg of CO<sub>2</sub> equivalent per kg of methane under self-heating conditions and electrical heating, respectively, which is significantly lower than that of typical natural gas production.

## 4. Repurposing mixed plastics in polymer blends

### 4.1 Compatibilisation of mixed plastics

Mechanical recycling is the most employed recycling method for plastic waste and commonly involves a series of physical processes such as shredding, washing, drying, extrusion and granulation.<sup>114</sup> However, existing mechanical recycling processes require relatively clean and often single plastic types and can result in deterioration of material properties. The process is further complicated when dealing with mixed plastics, requiring considerations for the differences in polymer miscibility and mechanical and thermal properties. The presence of contaminants can greatly affect the quality of the recycled plastics, resulting in large variances in fracture toughness and shear or thermal stabilities.<sup>115</sup> On the other hand, there is potential for repurposing mixed plastics to produce polymeric blend materials. Polymer blends refer to physical mixtures comprising two or more polymers and often exhibit differing physical properties from those of individual polymers. Thus, polymer blending offers a facile approach for tailoring the physical properties of polymeric materials to suit their targeted industrial applications, without the need for complex synthesis or chemical modifications of the polymers. On that account, polymeric blend materials are widely used in automotive, electrical, construction and packaging applications due to their excellent electrical insulation, strength, durability and barrier properties against environmental conditions, realisable by the tunability of their polymer blend compositions. The demand for polymeric blend materials has been increasing with the global polymer blend market projected to reach USD 6.6 billion by 2028 at a compounded annual growth rate (CAGR) of 6.7% from 2023 to 2028.<sup>116</sup> Yet, the primary challenge is the unfavourable physical interaction between dissimilar polymer chains that leads to blend immiscibility and compromised mechanical properties of the material. As such, certain external additives or unconventional processing techniques can be employed to improve the compatibility of different plastic polymers, generating blend materials with sufficiently good mechanical properties from mixed plastic streams. Successful compatibilisation of an immiscible polymer blend is often associated with increased interfacial adhesion, decreased interfacial tension, reduced coalescence of droplet morphology during processing, finer droplet morphologies or improved crack resistance at the interface.<sup>117</sup> In light of mixed plastic waste mechanical recycling, this section of the review will discuss the use of copolymer compatibilisers, nanofiller compatibilisers and unconventional processing techniques for compatibilisation of immiscible commodity polymer blends. The use of ionomers and

Janus nanoparticles as compatibilisers to improve polymer recycling has been discussed in other reviews.<sup>118,119</sup> Table 5 summarises the effects of various compatibilisers or processing techniques on the material properties of their compatibilised polymer blends.

Copolymers are among the main classes of compatibilisers employed in immiscible polymer blend systems. Typically, they are polymeric surfactants comprising multiple segments of differing affinities to facilitate interactions with different phases of the immiscible polymer blend for an improved extent of blending.<sup>120</sup> For example, a nonpolar backbone (e.g. PE or PP) grafted with polar side groups (e.g. maleic acid, maleic anhydride, acrylic acids or methacrylate) efficiently interacts with both nonpolar and polar segments in a polymer blend. Copolymers can either be added as an external additive (known as a non-reactive compatibiliser) or created *in situ* at the blend interface with the use of functional compounds that covalently bond with polymer blend components (known as reactive compatibilisers). Primarily, these copolymer compatibilisers assist in proper dispersion by sterically hindering the coalescence of dispersed phase droplets in the polymer matrix. This results in a larger surface area of the dispersed phase, lowering the surface tension and thereby improving the blending between immiscible polymers. Furthermore, finer-sized dispersed phase droplets also act as heterogeneous nucleating sites for the continuous phase, improving the nucleation rate and crystal growth for the continuous phase matrix.<sup>121</sup> Thus, a higher copolymer concentration is often associated with an exponential decrease in interfacial tension and enhanced mechanical properties up to a maximum. After which, a higher copolymer loading would result in copolymer agglomeration, weakening the copolymer–matrix adhesion and decreasing the tensile strength of the blend.<sup>122,123</sup> The application of copolymer compatibilisers was observed across a variety of immiscible polymer mixtures such as PP/PE,<sup>120,124–126</sup> PE/PET,<sup>120,124–126</sup> PP/PET,<sup>121,127</sup> PS/PVC,<sup>128</sup> and PE/PMMA<sup>129</sup> blends that are commonly found from waste plastic streams.

Besides copolymers, a variety of nanofiller materials including carbon nanotubes (CNTs),<sup>130</sup> carbon black (CB),<sup>131,132</sup> graphene oxide (GO)<sup>133</sup> and organoclays (OC)<sup>134,135</sup> have also exhibited efficacy in compatibilising immiscible polymer blends *via* physical interactions. The generic design of nanofiller compatibilisers resembles their copolymer counterpart in that they possess segments of varying affinities to facilitate good interactions with both polar and nonpolar phases for enhanced interfacial adhesion, enabling enhanced properties for the recycled polymer blend. Some nanofillers like GO are inherently amphiphilic and do not require further functionalisation before use.<sup>133</sup> However, other hydrophobic nanofillers such as CNTs need to be functionalised with polar groups for improved interactions with the polar phases.<sup>130</sup> Unlike copolymers, the addition of nanofillers can greatly influence the blend morphology of the polymer mixture. This is because nanoparticles are more prone to occupying one phase of the immiscible polymer blend rather than being localised at the

interface. While localised, the high rigidity of nanofiller particles such as CB can alter the viscosity ratio of the dispersed and continuous phase, increasing the phase viscosity and stiffness of polymer matrices *via* hydrodynamic reinforcement.<sup>122,132</sup> Some nanofillers even possess distinctive properties that further improve compatibilisation or impart value-added properties to the recycled blend materials. For example, OC, clay modified by replacing its cations with cationic organic surfactants, contains galleries that can host and promote intercalation of immiscible polymer chains, facilitating better interfacial adhesion in PP/PS blends.<sup>122</sup> Meanwhile, CNTs form network structures that bridge polymer matrices, facilitating stress transfer under load to prevent crack initiation and propagation along the interface. This nanobridge effect results in increased fracture toughness and electrical conductivity of HDPE/PA6 blends.<sup>130</sup> Conductive fillers like CB also confer electrical conductivity to PS/PMMA blends.<sup>131</sup>

To further mediate the interactions between immiscible mixed polymer blends, some unconventional processing techniques such as solid-state shear pulverisation (S<sup>3</sup>P) and microfibrillar composite (MFC) fabrication employ mechanochemistry to improve the properties of immiscible polymer blends. S<sup>3</sup>P facilitates excellent mixing of polymers and dispersion of nanofiller additives. It is based on twin-screw extrusion with continuous cooling below the melting temperatures of semicrystalline polymers and the glass transition temperatures of amorphous polymers to repeatedly fragment and fuse polymers under compressive and high shear conditions.<sup>146</sup> Effective dispersion of nanofiller additives in polymer matrices further enhances the mechanical properties of polymer blends.<sup>147</sup> S<sup>3</sup>P is a highly scalable process due to solvent-free conditions, no requirements for external additives and high product output. However, optimisation of S<sup>3</sup>P process parameters is non-trivial due to the differing responses of different polymers towards S<sup>3</sup>P compressive and shear conditions, which significantly impacts the product structure and properties. For instance, tough and semicrystalline polymers with high heat capacity and thermal diffusivity, such as HDPE, linear LDPE, PP, PA6 and PPS, were deemed most suitable for S<sup>3</sup>P as they engage better with chilled S<sup>3</sup>P screws.<sup>148</sup> Conversely, MFC fabrication involves melt extrusion of an immiscible polymer blend containing polymer phases of different melting temperatures, followed by cold drawing of the extrudate (fibrillation) to highly orientate the polymer components, and final thermal treatment melts the lower melting component to transform the blend into an isotropic matrix reinforced with microfibrils of the higher melting component.<sup>144</sup> The resultant MFC has enhanced elastic modulus and tensile strength of immiscible polymer blends.<sup>144,145</sup> It is a simple and inexpensive technique that can be used in conjunction with compatibilisers but is limited to only blends with polymers of similar processing temperatures, such as PET/PP<sup>144</sup> and PET/LDPE<sup>145</sup> blends, to avoid degradation. Compression moulding was deemed to be more suitable for MFC due to significant relaxation of reinforcing fibrils during

**Table 5** Properties of polymer blends compatibilised with compatibilising additives or unconventional processing techniques

Compatibiliser type or technique	Feedstock	Compatibiliser	Blend properties	Ref.
Copolymers – non-reactive	PE/PP	PE- <i>g</i> -PP, PE- <i>b</i> -PP	<ul style="list-style-type: none"> <li>• Grafted copolymers: increased tensile strength of blends with graft density and copolymer length</li> <li>• Multiblock copolymers: improved mechanical properties of blends, up to 600% strain at break due to increased entanglements and interfacial adhesion</li> </ul>	136 and 137
		EPR	<ul style="list-style-type: none"> <li>• Reduced dispersed phase size, improved interfacial adhesion, improved notched impact strength and ductility</li> <li>• No significant improvement in mechanical properties</li> </ul>	138
	PET/PE	PET- <i>b</i> -PE	<ul style="list-style-type: none"> <li>• Reduced dispersed phase size.</li> <li>• 30 times increase in strain at break and three orders of magnitude increase in peel strength</li> </ul>	124
		SEBS	<ul style="list-style-type: none"> <li>• Stable uniform morphology, improved PET crystallisation rate and interfacial adhesion</li> <li>• Tripled in elongation at break and doubled in impact strength</li> </ul>	120 and 126
	PE/PMMA	PMMA- <i>b</i> -PE- <i>b</i> -PMMA	<ul style="list-style-type: none"> <li>• Improved elastic modulus (up to 80%), hardness (up to 42%) and scratch resistance</li> <li>• Triblock copolymer with the highest molar-mass end blocks was most effective due to the higher degree of entanglements with the PMMA dispersed phase that enhance stress transfer</li> </ul>	129
Copolymers – reactive	PE/PP	HDPE- <i>g</i> -MAH, PP- <i>g</i> -MAH	<ul style="list-style-type: none"> <li>• Higher yield strength, storage modulus, loss modulus, complex viscosity and phase homogeneity of compatibilised blends</li> </ul>	139
	PET/PE	PE- <i>co</i> -MAA	<ul style="list-style-type: none"> <li>• Good morphological distribution of PE in PET</li> <li>• Enhanced tensile strength by five times with marginal increase in ductility</li> </ul>	120
	PET/PP	PP- <i>g</i> -MAH	<ul style="list-style-type: none"> <li>• Better mechanical and thermal properties</li> <li>• Enhanced mechanical properties and fine dispersed morphology</li> </ul>	121
		PP- <i>g</i> -GMA	<ul style="list-style-type: none"> <li>• Improved nucleation, resulting in increased tensile strength, elongation at break, Young's modulus and crystallisation temperature</li> <li>• Tensile behaviour changed from fragile to ductile with a 10% increase in tensile strength, 10- to 20-fold improvement in elongation at break, and no effect on stiffness. Low-GMA graft content was more efficient in blend emulsification</li> </ul>	127
	PE/PP/PS PS/PVC	SEBS/EPR P( <i>S-b</i> -MMA)	<ul style="list-style-type: none"> <li>• Reduced dispersed phase size and enhanced impact resistance</li> <li>• Increased interface area, finer morphology and doubled impact resistance</li> </ul>	140 and 128
Nanofiller particles	PE/PA6	CNTs	<ul style="list-style-type: none"> <li>• Improved peel adhesion strength, fracture toughness and ductility of the blend material</li> <li>• Selective distribution of carbonyl and hydroxyl-functionalised CNTs at the interface improves the extent of blending and the bridge effect of CNTs prevents crack propagation along the interface</li> </ul>	130
	PS/PMMA	CB particles	<ul style="list-style-type: none"> <li>• Improved storage modulus by more than 10 times and complex viscosity by more than 5 times</li> </ul>	131 and 132
	PA/PPO	GO particles	<ul style="list-style-type: none"> <li>• Electrical conductivity observed from <math>10^{-12}</math> S cm<sup>-1</sup> to <math>10^{-5}</math> S cm<sup>-1</sup></li> <li>• Dramatically reduced dispersed phase droplet size and increased ductility</li> </ul>	133
	PP/PS	OC	<ul style="list-style-type: none"> <li>• Enhanced mechanical strength and thermal stability</li> <li>• Reduced dispersed phase size and improved the homogeneity of PP and PS</li> <li>• More than 3 times increase in peel strength</li> </ul>	122
Solid-state shear pulverisation (S <sup>3</sup> P)	PS/PMMA	S/MMA gradient copolymer	<ul style="list-style-type: none"> <li>• Fine dispersion phase size with quasi-nanostructured morphology and irregular dispersed phase domain sizes of ~100 nm or less</li> <li>• Stable morphology under short-residence-time single-screw extrusion but requires gradient copolymers to suppress coarsening over long-term high-temperature static annealing</li> </ul>	141
	PS/PMMA, PS/PE	—	<ul style="list-style-type: none"> <li>• Stable dispersed phase domain size under static annealing</li> <li>• Blend compatibilisation attributed to <i>in situ</i> block copolymer formation at interfaces or highly mixed regions during pulverisation following chain scission and radical combination</li> </ul>	142
	PP/PS	—	<ul style="list-style-type: none"> <li>• Considerably smaller dispersed phase size with less coarsening upon annealing compared to melt-mixed blends</li> </ul>	143
Microfibrillar composite fabrication (MFC)	PP/PET	E-GMA	<ul style="list-style-type: none"> <li>• Improved flexural modulus and flexural strength, only dependent on E-GMA loading for compression moulding</li> <li>• Significant relaxation of reinforcing fibrils during injection moulding was detrimental for fibril orientation and the mechanical properties of the composite</li> </ul>	144
	PET/PE	—	<ul style="list-style-type: none"> <li>• Marked improvement in mechanical properties with 10 times higher elastic modulus, at least 2 times higher tensile strength and a 50% increase in impact strength compared to pristine LDPE</li> </ul>	145

Table 6 Comparison of compatibilisation methods

	Advantages	Disadvantages
Copolymers	<ul style="list-style-type: none"> <li>Designed to localise at the blend interface to reduce interfacial tension</li> </ul>	<ul style="list-style-type: none"> <li>Copolymer concentration at the interface can be depleted by micellar formation</li> <li>Copolymer agglomeration at higher loading reduces the tensile strength of the blend</li> <li>Reactive compatibilisers impractical due to slow reaction rates during melt processing</li> <li>May not improve the mechanical properties of the immiscible blend</li> </ul>
Nanofiller particles	<ul style="list-style-type: none"> <li>Extremely high aspect ratio promotes high filler–matrix interactions, minimising interfacial tension</li> <li>Dual role as fillers improves mechanical properties, scratch resistance, barrier properties, and fire resistance of polymer matrices at low loadings</li> </ul>	<ul style="list-style-type: none"> <li>Challenging to localise NPs at the interface, mostly locate in one phase of the polymer blend</li> <li>Nanofiller agglomeration at higher loading</li> </ul>
Solid-state shear pulverisation	<ul style="list-style-type: none"> <li>Repeated fragmentation and fusing effectively disperse polymers and filler additives for enhanced mechanical properties</li> <li>Solvent-free, high output, scalable and continuous</li> <li>Can mix polymers of different viscosities and be used with or without compatibiliser additives</li> </ul>	<ul style="list-style-type: none"> <li>Chain scission occurs during repeated fragmentation and fusing of polymers</li> </ul>
Microfibrillar composite fabrication (MFC)	<ul style="list-style-type: none"> <li>Polymer reinforcement using polymer improves mechanical integrity with no need for external additives</li> <li>Easy processability of the blend, can be used with or without compatibiliser additives</li> <li>Simple and inexpensive</li> </ul>	<ul style="list-style-type: none"> <li>Polymer phases must be of different melting temperatures to form reinforcing fibrils</li> <li>Polymers must be of similar processing temperatures to avoid degradation</li> </ul>

injection moulding that was detrimental for fibril orientation and thus the mechanical properties of the composite.<sup>144</sup>

Table 6 sums up the advantages and disadvantages of compatibilisation methods discussed in this section.

Though few, there have been some promising reports on the use of mixed municipal waste plastics to produce polymer composites. As organic and inorganic additives are conventionally incorporated into plastics to support their intended function in domestic and industrial activities, the effect of these residual additives on the outcome of recycled polymer blends is paramount to the practicality of this approach. In that light, Singh *et al.*<sup>149</sup> prepared polymer blends from both fresh mixed municipal plastics and landfill-mined plastic waste containing PE, PP, PES, PVC, PS and others. It was found that calcium carbonate, a filler to enhance processability and mechanical properties, and titanium dioxide, a pigment for ultraviolet stability, were the dominant residues in the waste plastics. Interestingly, despite the immiscibility between polymer phases and the absence of compatibiliser additives, the mechanical properties of the waste blends were superior to those of virgin blends, attributed to the filler and reinforcing role of the residues. Thus, production of polymer blends directly from municipal waste plastics can be promising when supported by the incorporation of suitable compatibiliser additives, as discussed earlier in this section.

#### 4.2 Assessment

All in all, mechanical recycling technologies for mixed plastics have only found limited success in blending municipal plastic waste to yield mechanically recycled plastic products with adequate mechanical properties. This is evident from the fact that most successes have been achieved only on selected bivalent

polymer blends of known composition and physical properties, which means that there is still a long way from achieving quality products from a generic municipal plastic waste stream. For polymer blending, successful compatibilisation of an immiscible polymer blend is the key factor determining the material properties of the blend material. While different plastic polymers are usually not miscible due to innate differences in their chemical structures, these could be resolved by incorporating compatibilisers such as copolymers and nanofillers. Together with unique processing techniques such as S<sup>3</sup>P or MFC, we have seen successful recycling of different commodity plastic polymer mixtures, producing polymer blends with similar or (sometimes) enhanced and unique material properties to their parent polymers. Notably, the mechanical properties of blends produced from municipal plastic waste were reported to be superior to blends produced from virgin plastic blends due to the reinforcing effect of certain filler residues.<sup>149</sup> Thus, there is potential to generate polymeric blend materials directly from municipal plastic waste with the use of suitable compatibilising additives and fabrication processes. However, it is noteworthy that dirt and organic matter contamination in municipal mixed plastic waste might require special handling including blend processing equipment capable of withstanding organic matter charring and toxic gas release that may occur at high operating temperatures, requiring more frequent equipment upkeep and maintenance.

A critical bottleneck in compatibilised blending of municipal plastic waste is the stochasticity of its plastic composition. The waste plastic composition is crucial because ideal blend processing parameters and additives are highly dependent on the types of plastics present due to considerations of thermal stability or phase immiscibility. In addition, diverse variation in the ratio of plastic types across waste batches makes it chal-

lenging to reproduce product blends with consistent material properties. This could be circumvented by thorough quantification of each waste plastic batch and the inclusion of other waste plastics to tune the blend composition before the melt extrusion stage.<sup>150</sup> Yet, extensive characterisation of each waste batch will require consideration of the recoverable economic value of the produced polymeric blend materials. Alternatively, machine learning and AI-assisted tools have the potential to be used for the prediction of product blend properties and optimisation of polymeric blending conditions and additives to yield valuable materials from waste plastic mixtures of diversely different plastic compositions, thereby reducing the resources required to address the complexity and stochasticity of municipal plastic waste compositions. For example, Zhang *et al.* designed a simple machine learning model with at least 75% accuracy for the prediction of polymer-blend compatibility using PMMA/PVC blends, which revealed that miscibility is contingent on various factors.<sup>151</sup> Though PMMA and PVC are not entirely immiscible, such machine learning models could be adapted to work for immiscible plastic blends.

In addition, it is important to consider the end-of-life of the compatibilised polymer blends formed from mixed plastics. Comprising a number of different polymers, fillers and compatibilisers, the post-use treatment of these blends could prove to be significantly more complex than sorting the plastic feedstock mixture prior to blending. Theoretically, the compatibilised polymeric blends could be reprocessed at their end-of-life using similar conditions as those used in the first iteration of blending, albeit with the need for additional compatibilisers. However, reprocessing is only viable for end-of-life polymeric blends with non-significant degradation to their blend components during use. The degradation behaviour of polymer blends is highly complex and largely depends on antagonistic or synergistic interactions between blend components. In the case of PP/PS blends with and without an SBS compatibiliser, the SBS compatibiliser exhibited a significant stabilising effect on thermomechanical degradation but increased the kinetics and carbonyl content in photodegradation.<sup>152,153</sup> Thus, stability testing of polymeric blends with various compatibilisers should be studied to minimise blend degradation during use and better preserve them for more reprocessing iterations. It is also noteworthy that, because most compatibilisers localise at the interface between polymers, removing the compatibiliser from a polymeric blend is very difficult and seldom done. Therefore, upon polymeric blending, individual polymers cannot be recovered from the blend for separate recycling or upcycling, further necessitating the development of new polymeric blend recycling or upcycling methods. At the time of writing, this was a significant research gap in need of simple, sustainable and cost-effective solutions.

## 5. Summary and perspectives

An effective and practical post-use plastic economy built on recycling and upcycling hinges on the quality of accessible

plastic feedstock. As such, strategies to deal with the unavoidable contamination and mixtures that make up plastic feedstock, both from post-industrial and municipal streams, become integral to the entire value chain. To complement prior sorting and cleaning, designing recycling and upcycling processes for greater tolerance towards mixed plastics and contaminants can not only offer process simplification but also potentially lower costs. In this review, we showcased the possibilities and highlighted a number of key considerations and approaches towards utilisation of mixed/contaminated plastics in chemical recycling, upcycling and composite formation. Amongst these approaches, chemical recycling of mixed plastics is the most technologically mature. Despite these recent advances, many other considerations remain, with large scope for further improvement and technological advancement. This section highlights some of the most pertinent issues that need to be considered and addressed, though it should be noted that their solutions may not currently exist at the time of writing.

Firstly, with rapid advances in biorefinery and considerations of polymer circularity, the diversity of commercial high-volume plastic types is expected to increase considerably. While drop-ins (*e.g.* bio-PE) are indistinguishable from their petroleum-derived counterparts, there are numerous alternatives whose presence in waste streams can likely enhance the complexity of waste streams and necessitate more selective processes to be developed. For instance, plastic bottles made of poly(ethylene furanoate) (PEF), a bio-derived polymer considered non-compostable,<sup>154</sup> may be disposed of together with their traditional PET counterparts. With both polymers susceptible to similar chemistries such as solvolysis during chemical recycling, the resulting depolymerised product mixture will need to be further purified before each can be re-polymerised. Alternatively, novel strategies can also be developed to selectively depolymerise one type of polyester over the other, though the preference for the eventual strategy will ultimately depend on process complexity and the associated economics.

To achieve large-scale practical adoption, the products from recycling and upcycling need to achieve cost parity with incumbent products/materials, with at least comparable performance in various applications. The scalability of a recycling or upcycling process depends on its efficiency, robustness, energy input and simplicity, ideally doing away with extensive and costly pretreatment or separation processes. Although municipal contaminants can poison the catalyst used for plastic valorisation and compromise the efficiency of catalytic systems,<sup>46</sup> intelligent process and catalyst design was found to enable tolerance and achieve favourable outcomes even in their presence.<sup>48,83,93,98,107,149</sup> Environmental and resource sustainability of the processes should also be considered, necessitating reduced reliance on precious metals and favouring heterogeneous or neat reactions to avoid the use of hazardous organic solvents.<sup>60</sup> Not only will these reduce their environmental impact but also minimise costs associated with treatment or disposal of the associated waste produced.

To this end, it is recommended that the 12 Principles of Green Chemistry<sup>155</sup> serve as key guidance for process development. In addition, as aforementioned, TEA and LCA should be employed to better assess the environmental footprint and economics of new recycling/upcycling processes developed, whilst identifying key sustainability and financial bottlenecks respectively. For example, a recent TEA and LCA conducted on a conceptual catalytic fast mixed plastic pyrolysis facility revealed that the recycling process favourably reduced fossil fuel depletion and often lowered acidification, ozone depletion and smog formation compared to the virgin manufacturing processes.<sup>156</sup> However, the economic viability of the process was largely dependent on the product composition, their yield and their minimum selling price with respect to virgin incumbents, demonstrating the importance of upcycling waste plastics to products of higher commercial value. Key economic factors driving up the product costs were identified to be the cost of mixed plastic feedstock, operation, and product separation. As such, employing a catalyst with higher aromatic selectivity improved both the environmental impact and economic viability of the process with less utilities and plastic feedstock consumed per unit of BTX produced, making it competitive with virgin BTX production. Supportive government regulations and legislation are also invaluable in further driving adoption and development of both recycling and upcycling technologies. Governmental support in the form of EU's Circular Economy Action Plan,<sup>157</sup> China's National Sword Policy<sup>158</sup> and India's Plastic Waste Management Rules<sup>159</sup> are crucial steps in this direction. Other international policies like the Extended Producer Responsibility (EPR) mandate that producers are responsible for handling post-consumer plastic packaging waste and can steer companies towards designing more recyclable and sustainable packaging and investing funds in post-consumer plastic waste collection and recycling infrastructures.<sup>160</sup>

Most importantly, the fate of the upcycled materials or composites must be well considered and designed from the outset. Although upcycling of post-consumer plastics gives opportunities to reduce usage of additives by conferring new properties onto common polymers,<sup>63</sup> they can potentially complicate the waste stream for both recycling and upcycling, leading to overall more detrimental outcomes. A similar consideration applies for polymer blends, as aforementioned. For upcycling, one may envisage a "cradle-to-cradle" sequential upcycling approach, where the original plastic is first upcycled into a new (polymeric) material, which can then be further chemically upcycled into other value-added products. This concept was recently demonstrated by the conversion of PE into PE-graft-PCL polymer electrolytes for energy storage, which was further upcycled at the material's end-of-life into industrially-relevant C<sub>4</sub>-C<sub>6</sub> dicarboxylic acids.<sup>67</sup> In another potential approach, enhanced chemical recyclability can be engineered into the upcycling strategy during post-synthetic polymer modifications. For instance, inserting a small amount of amide linkages into the PE backbone enables site-specific chemical degradability (similar to that of nylons) for recycling,

whilst simultaneously conferring new properties such as luminescence, enhanced tensile strength and surface hydrophobicity offers the possibility of new applications not accessible to the parent PEs.<sup>161</sup>

Finally, the advent of artificial intelligence, automation and high-throughput screening capabilities opens numerous new opportunities. In the sorting of mixed plastic feedstock, these technologies can enhance the accuracy of resin identification, leading to greater efficiency and improved sorting capabilities.<sup>162,163</sup> Additionally, they have the potential to shorten the innovation cycle by accelerating the discovery of new catalysts, chemistries and processes for both chemical recycling and upcycling of even mixed plastic mixtures. In totality, the need to address mixed plastic waste feedstock is essential for plastic recycling and upcycling technologies to reach technological maturity and large-scale implementation. We are optimistic for significant advancements to emerge in coming years, driven by the current strong research momentum in this area, promoting a more sustainable end-of-life for these versatile and ubiquitous materials.

## Conflicts of interest

The authors declare the following competing financial interest (s): Patent applications have been filed on some of the papers cited in this review.<sup>65,67</sup>

## Abbreviations

PE	Polyethylene
HDPE	High density polyethylene
LDPE	Low density polyethylene
PP	Polypropylene
PS	Polystyrene
PET	Poly(ethylene terephthalate)
rPET	Recycled poly(ethylene terephthalate)
PEF	Poly(ethylene furanoate)
PVC	Poly(ethylene chloride)
dPVC	Dechlorinated poly(ethylene chloride)
PLA	Poly(lactic acid)
PCL	Poly(caprolactone)
PMMA	Poly(methyl methacrylate)
PAA	Poly(acrylic acid)
PC	Polycarbonate
PU	Polyurethane
PES	Polyethersulfone
PPO	Polypropylene oxide
PHA	Polyhydroxyalkanoates
SEBS	Styrene-ethylene-butylene-styrene
EPR	Ethylene propylene rubber
GO	Graphene oxide
CNTs	Carbon nanotubes
CB	Carbon black
OC	Organoclay

MAH	Maleic anhydride
MAA	Maleic acid
GMA	Glycidyl methacrylate
EG	Ethylene glycol

## Data availability

No primary research results, software or code have been included and no new data were generated or analysed as part of this review.

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