ESRA

Green and Sustainable Chemistry Letters

Chemical Recycling and Upcycling of Plastic Waste: Pathways, Mechanisms and Life-Cycle Evidence

A Narrative Review

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ARTICLE INFO

Prepared 20th October 2026, www.esrapublications.com

Revised & Published 5th November 2026

ABSTRACT

Only about 9% of the plastic waste generated up to 2015 was recycled [1], and mechanical recycling alone cannot absorb contaminated, multilayer, degraded or heteroatom-rich streams. Chemical recycling and upcycling aim to recover monomers, fuels, chemical intermediates or higher-value materials from such streams. This review compares four families of routes: Solvolytic and enzymatic depolymerization, thermal and catalytic pyrolysis, catalytic hydrogenolysis and hydrocracking, and selective oxidation or photochemical conversion. For each family polymer chemistry has been linked (cleavable bonds, contaminants) to reaction mechanism, operating window and product specification, using schemes and a polymer–route compatibility matrix. Quantitative evidence from comparative techno-economic and life-cycle studies, which indicates that route ranking depends strongly on feedstock, product quality, system boundaries and the displaced counterfactual has been summerised. Finally, a reporting checklist (mass and carbon balances, product quality, energy and hydrogen demand, residues, counterfactual) and a route-selection logic is proposed. The review is narrative rather than systematic; sources were selected from recent reviews and landmark primary studies, and all references were checked against publisher or index records.

Keywords: Plastic waste; chemical recycling; upcycling; depolymerization; pyrolysis; hydrogenolysis; life-cycle assessment.

1. Introduction

Global plastic production reached about 8.3 billion tonnes by 2015, of which roughly 6.3 billion tonnes had become waste; of that waste about 9% had been recycled, 12% incinerated and 79% landfilled or released to the environment [1] (Figure 1). Land-based inputs alone were estimated to send 4.8–12.7 million tonnes of plastic waste to the ocean in 2010 [2]. Because plastics are also energy- and greenhouse-gas-intensive to produce [3], keeping their carbon in use has both waste-management and climate rationales.

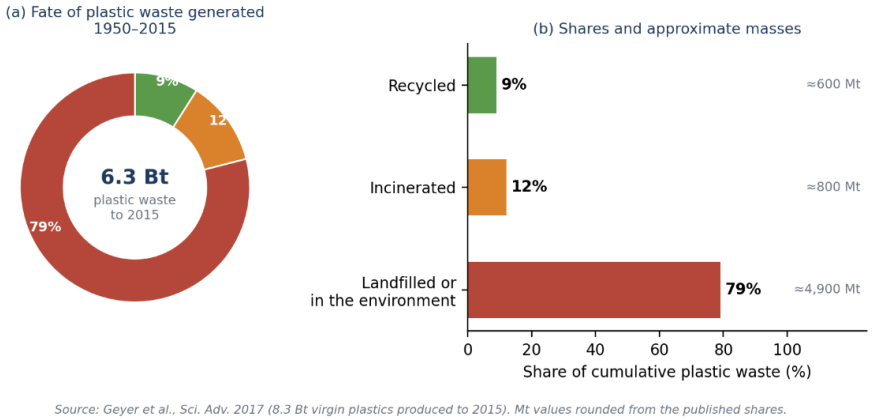

Figure 1. Fate of the 6.3 Bt of plastic waste generated between 1950 and 2015: (a). shares; (b). shares with approximate masses (rounded from the published percentages). Data: Geyer et al. [1].

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Mechanical recycling is the preferred first option where streams are clean, sorted and compatible, but repeated processing degrades polymer chains, and additives, inks, adhesives and mixed polymers limit recyclate quality [4]. Chemical routes are therefore proposed as complements for streams that mechanical recycling handles poorly, not as replacements [5,6]. Early classifications distinguish primary, secondary (mechanical), tertiary (chemical) and quaternary (energy recovery) recycling; contemporary reviews further separate **chemical recycling** (return to monomers or feedstocks for the same or similar polymers) from **upcycling** (conversion to products of higher value or different function) [7–9].

Recent reviews cover chemical recycling broadly [10–12], depolymerization [13], polyolefin upcycling [14], catalytic and biological conversion [15], and hazardous plastic waste [16]. The present article adds a mechanism-first comparison that links polymer structure to route choice and to the evidence needed for fair life-cycle comparison. It is a narrative synthesis, not a meta-analysis; numerical values are quoted only where they come from the cited sources and figures that are qualitative are labelled as such.

2. Recycling pathways and polymer–route compatibility

Figure 2 places chemical routes within the waste hierarchy: prevention and reuse come first, mechanical recycling next where quality allows, and chemical routes address the remaining streams, with energy recovery or disposal as the residual option. Which chemical route is plausible depends first on the bonds in the polymer backbone (Table 1).

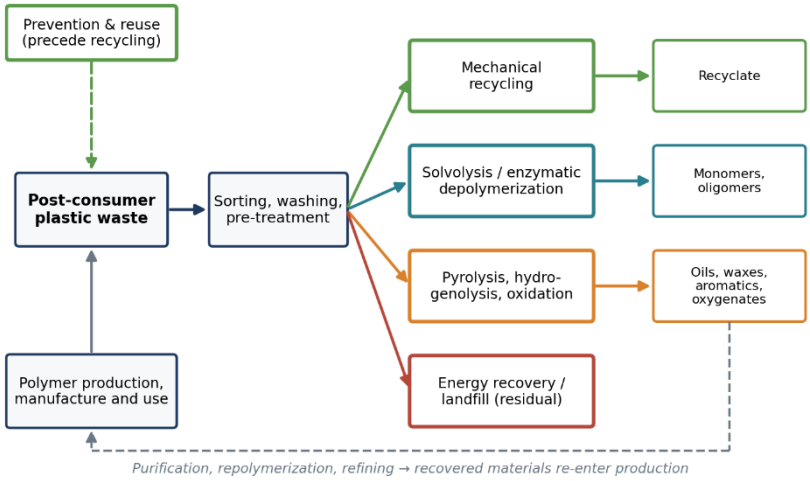

Figure 2. Pathways for post-consumer plastic waste. Route colours match later figures (green: mechanical; teal: depolymerization; orange: C–C-cleaving chemical routes; red: residual).

Table 1. Polymer families, cleavable bonds and typical chemical routes

Polymer family	Key chemical feature	Plausible chemical routes	Typical products and cautions
PET / polyesters	Ester C(=O)–O bonds	Glycolysis, methanolysis, hydrolysis, enzymatic hydrolysis	Monomers or oligomers (BHET, DMT, TPA); colour bodies, co-polymers and additives affect purity
Polyamides (nylon 6, 66)	Amide C(=O)–N bonds	Hydrolysis, alcoholysis, catalytic depolymerization	Lactams, diamines, diacids; nitrogen content complicates thermal routes
PE, PP	C–C / C–H backbone (no heteroatoms)	Pyrolysis, hydrogenolysis/hydrocracking, tandem catalysis, oxidation	Oils, waxes, alkanes, alkylaromatics, olefins or oxygenates; selectivity is the central problem
PS	Aromatic C–C backbone	Pyrolysis / catalytic depolymerization	Styrene-rich oils and aromatics
PVC	Chlorine; plasticizers and stabilizers	Dechlorination before or during conversion	HCl and chlorinated species; contaminant management is mandatory
Mixed / multilayer waste	Heterogeneous composition	Pyrolysis, gasification, oxidation with biological funnelling	Variable products; contaminants (halogens, N, O, metals, fillers) govern feasibility

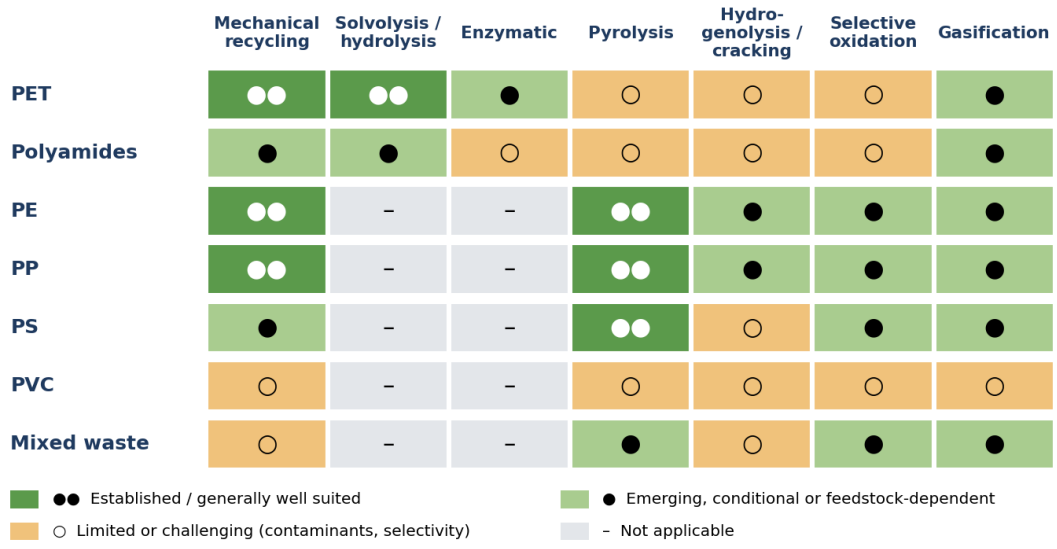

Figure 3. Qualitative polymer–route compatibility matrix. Cell shading reflects chemical plausibility and typical maturity as discussed in the reviews cited in the text [10, 11, 13–15]; it is an interpretive guide, not a performance ranking.

2.1 Depolymerization of condensation polymers

Polyesters, polyamides and similar condensation polymers contain heteroatom linkages that nucleophiles can cleave under comparatively mild conditions, which allows selective recovery of monomers or oligomers [13]. For PET, the ester carbonyl is attacked by ethylene glycol (glycolysis, giving bis(2-hydroxyethyl) terephthalate, BHET), methanol (methanolysis, giving dimethyl terephthalate, DMT) or water (hydrolysis, giving terephthalic acid, TPA) (Figure 4). Typical reported operating windows are about 180–250 °C for glycolysis and 180–280 °C for methanolysis, with hydrolysis spanning mild alkaline to high-temperature neutral conditions [5, 10]. Recovered intermediates must reach polymerization-grade purity to support closed-loop use; alternatively, PET-derived intermediates can be combined with bio-based monomers to make higher-value polymers in open-loop upcycling [17]. Polymers can also be designed for chemical recycling to monomer, which relaxes purification demands [18].

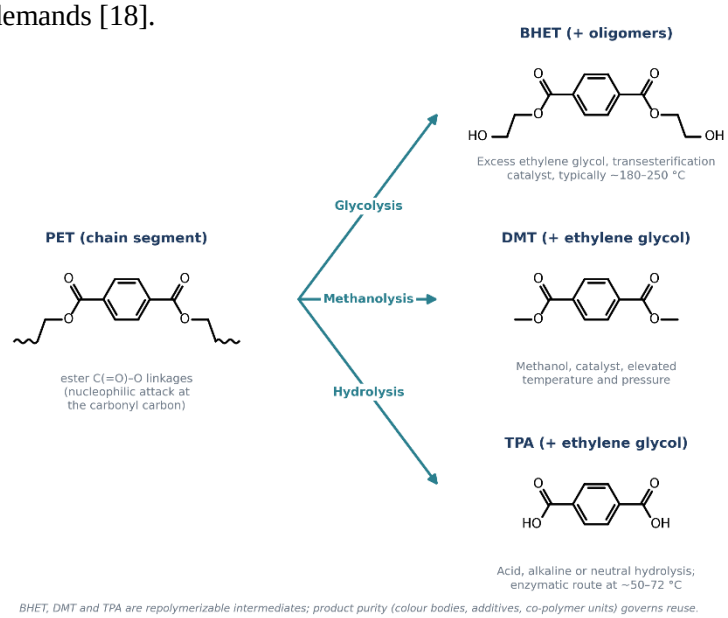

Figure 4. Depolymerization routes for PET. Nucleophilic attack at the ester carbonyl yields BHET, DMT or TPA depending on the reagent; temperatures are typical literature ranges [5, 10].

2.2 C–C-cleaving routes for polyolefins

Polyethylene and polypropylene lack cleavable heteroatom bonds, so conversion requires breaking C–C bonds by thermal, catalytic or oxidative means (Figure 5). In **pyrolysis**, random homolysis produces radicals that undergo hydrogen transfer and β-scission, giving broad mixtures of gas, oil and wax; catalysts (for example zeolites) can narrow the distribution and lower temperature [5, 10]. In **hydrogenolysis**, supported metals cleave C–C bonds in the presence of H_2 at lower temperatures. Pt/$SrTiO_3$ converted polyethylene to lubricant- and wax-range liquids at 300 °C under 170 psi H_2 [19], while a tandem hydrogenolysis/aromatization over Pt/γ-Al_2O_3 at 280 °C, without added H_2 or solvent, gave up to about 80 wt% of long-chain alkylaromatics and related liquids [20]. A different tandem approach used ethylene together with isomerization and olefin metathesis catalysts to deconstruct polyethylene to propylene [21]. **Selective oxidation** converts polyolefins to oxygenates such as carboxylic acids; combined with biological funnelling it can transform mixed PS/HDPE/PET streams into single products [22]. Catalyst accessibility to polymer chains, mass transfer in viscous melts and catalyst deactivation by contaminants are recurring constraints [14, 15].

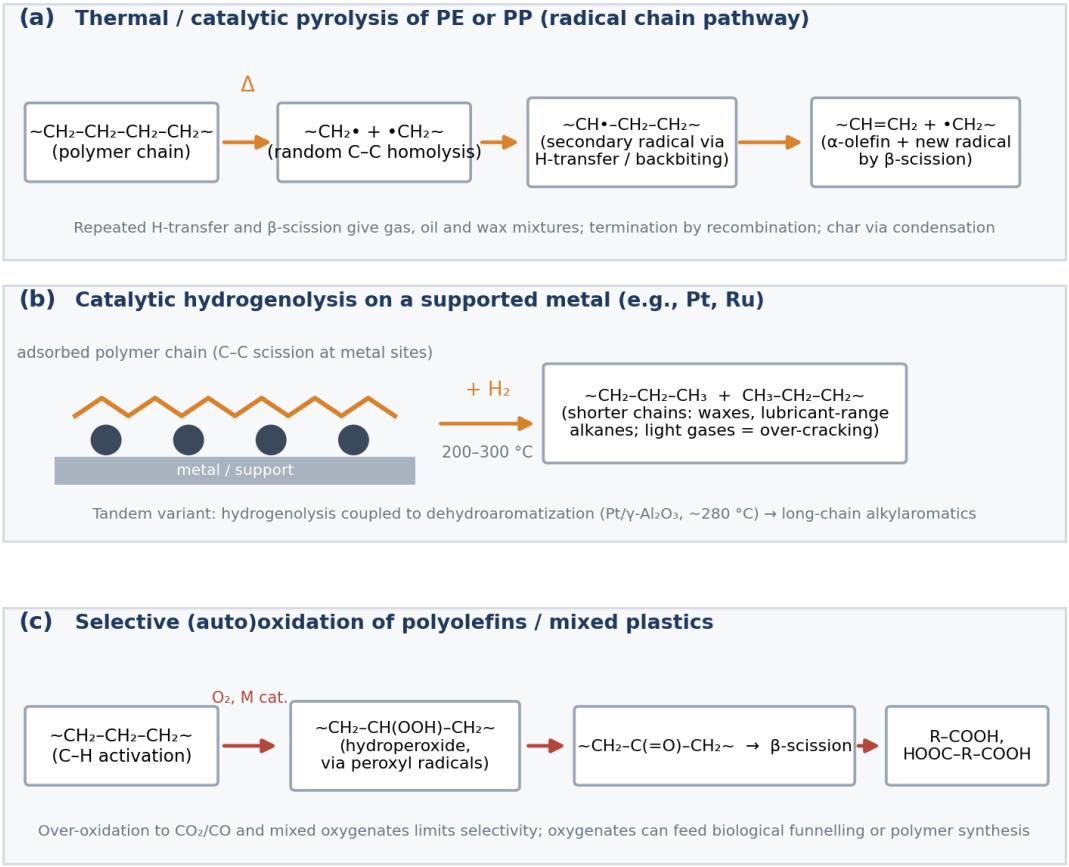

Figure 5. Simplified mechanisms for polyolefin conversion: (a) radical pyrolysis; (b) hydrogenolysis on a supported metal, including the tandem aromatization variant [20]; (c) selective oxidation to oxygenates. Schematic only; real systems form product distributions.

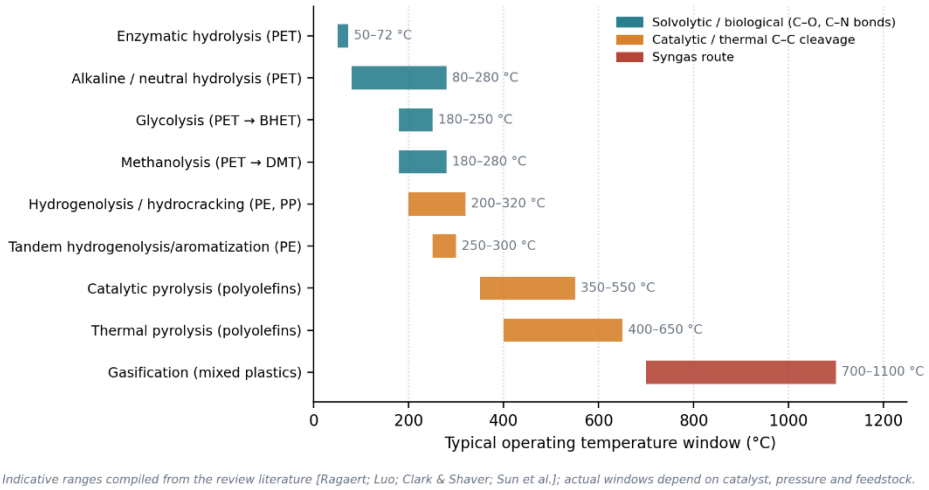

Figure 6. Indicative operating-temperature windows of the main chemical routes (compiled from reviews, not from a single study). Solvolytic and biological routes operate at lower temperature than C–C-cleaving and syngas routes, but they are restricted to heteroatom-containing polymers.

2.3 Biological and photochemical routes

Enzymatic hydrolysis is the most mature biological route. The PET-degrading bacterium Ideonella sakaiensis established that enzymes can depolymerize and assimilate PET [23]. An engineered leaf-compost cutinase achieved at least 90% depolymerization of pretreated post-consumer PET within about 10 hours at 72 °C and 200 g per kg of reaction mixture, and the recovered monomers were repolymerized into bottles [24]. Machine-learning-guided engineering has since produced PET hydrolase variants active at lower temperatures on post-consumer plastics [25]. Light-driven approaches use photocatalysts or radical chemistry to depolymerize polymers to monomers or small molecules under milder conditions, but remain largely at laboratory scale [26].

3. Comparative performance: What the evidence shows

Table 2 compiles representative primary studies, restricted to parameters stated in the cited abstracts and records. Laboratory conditions (pure, additive-free feedstock; batch reactors; high catalyst loading) are not equivalent to industrial operation, so such results indicate chemical potential rather than process performance.

Table 2. Representative primary studies and reported conditions

Study	Route / feedstock	Catalyst or system	Reported conditions	Key outcome
Tournier et al. 2020 [24]	Enzymatic hydrolysis; pretreated post-consumer PET	Engineered cutinase (LCC variant)	72 °C; 200 g kg^{-1} PET; about 10 h	≥90% depolymerization; monomers repolymerized to PET
Celik et al. 2019 [19]	Hydrogenolysis; PE	$Pt/SrTiO_3$	300 °C; 170 psi H_2; solvent-free; up to 96 h	Liquid products in lubricant/wax range with narrow distribution
Zhang et al. 2020 [20]	Tandem hydrogenolysis/aromatization; PE	$Pt/\gamma\text{-}Al_2O_3$	280 °C; no added H_2 or solvent	Up to about 80 wt% long-chain alkylaromatics and related liquids
Conk et al. 2022 [21]	Catalytic deconstruction with ethylene; PE	Tandem isomerization / olefin-metathesis catalysts	See original study	Propylene from polyethylene
Sullivan et al. 2022 [22]	Chemical oxidation + biological funnelling; mixed PS, HDPE, PET	Metal-catalysed autoxidation; engineered bacterium	See original study	Mixed plastics converted to single target products

Comparative assessments add system context (Figure 7). A technical, economic and environmental comparison of closed-loop options for PE, PET and PP found that mechanical recycling and PET glycolysis had the lowest costs (9–73% lower than competing technologies) and environmental impacts (7–88% lower), whereas dissolution, enzymatic hydrolysis and methanolysis delivered the best recyclate quality (2–27% higher) [27]. For mixed plastic waste, pyrolysis had about 50% lower climate-change impact and life-cycle energy use than energy recovery, but higher impacts in other categories than mechanical recycling, energy recovery and virgin production [28]. A review of life-cycle studies reported that inconsistent upstream assumptions accounted for 5–26% of the climate-change impact in the studies examined [29]. Integrated techno-economic and life-cycle analyses of thermochemical LDPE upcycling illustrate how hydrogen, energy and product-value assumptions drive outcomes [30], and earlier reviews already noted the heterogeneity of chemical-recycling LCAs [31].

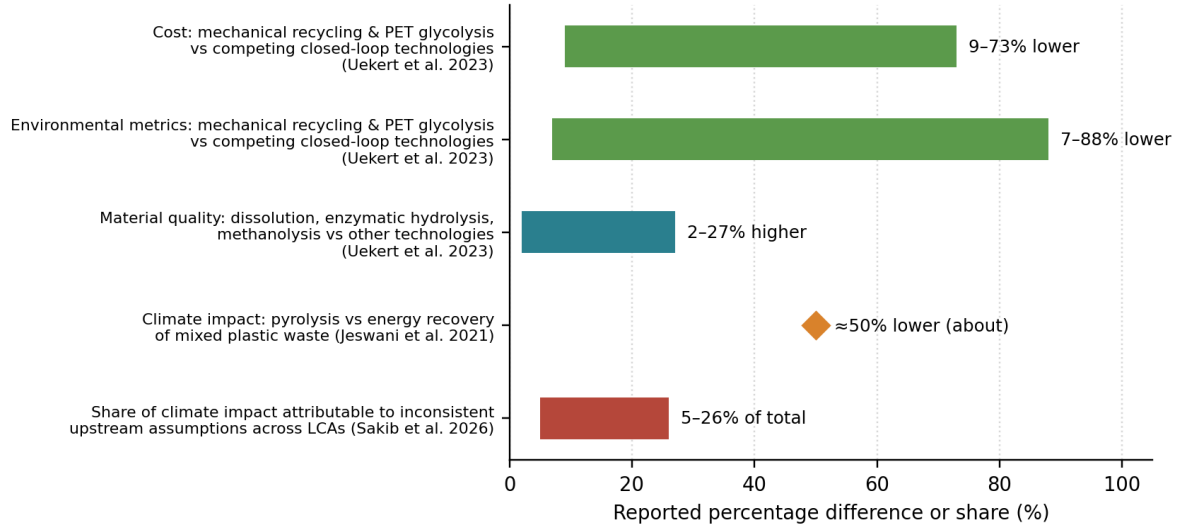

Figure 7. Quantitative ranges reported in selected comparative assessments [27–29]. The bars come from different feedstocks, system boundaries and impact categories and must not be read as a single ranking.

Three conclusions follow. First, no route is uniformly best: low cost and low impact favour the simplest, most selective routes where feedstock is suitable, while higher recyclate quality and tolerance of difficult feedstocks often cost more energy or reagents. Second, the displaced product matters: crediting a pyrolysis oil as replacing virgin naphtha gives a different result from crediting it as replacing fuel. Third, methodological transparency is as important as the technology, which motivates the reporting framework in Section 4.

4. Assessment framework and reporting checklist

A defensible comparison starts with a functional unit that reflects the service delivered (for example, one tonne of recovered product of specified quality), not merely one tonne of waste treated, and with recommendations for the system boundary, allocation and end-of-life modelling [32]. Figure 8 outlines the boundary and counterfactual elements, and Table 3 lists the metrics we recommend reporting.

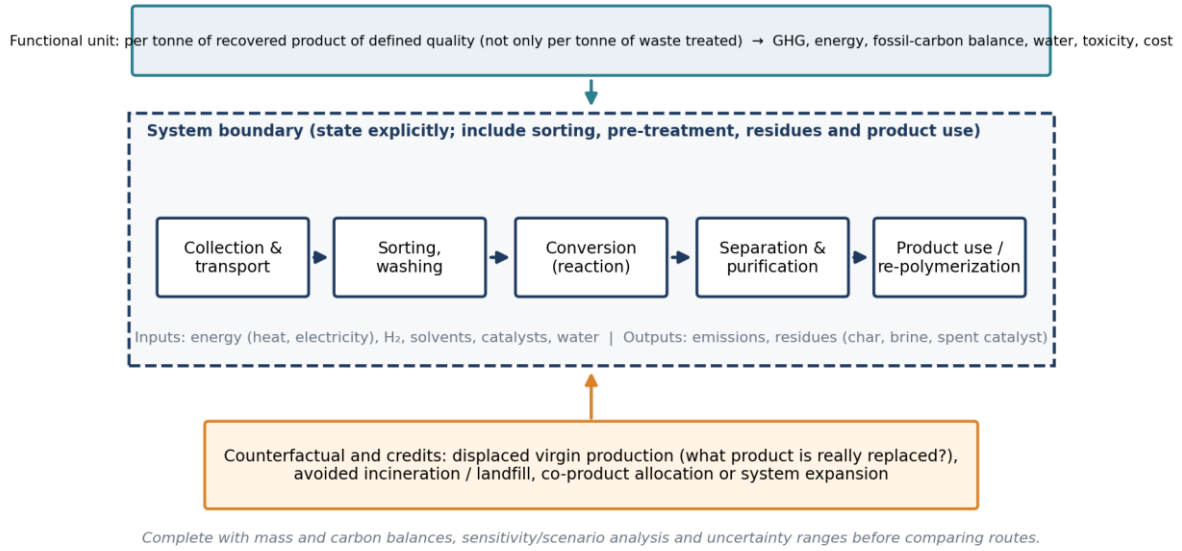

Figure 8. Assessment framework: functional unit, system boundary (collection to product use), inputs and outputs, and the counterfactual used for credits.

Table 3. Recommended metrics for reporting chemical recycling and upcycling studies

Metric	Definition / expression	Why it matters
Conversion and yield	Mass of feed converted; mass of target product ÷ mass of feed (wt%)	Shows what fraction becomes the intended product, not merely lower-molecular-weight material
Selectivity and product quality	Fraction of target species; specification (purity, boiling range, colour, contaminants)	Determines whether the product can be used without costly upgrading
Carbon efficiency	Carbon in useful product ÷ carbon in feed × 100%	Distinguishes true recycling from loss to gas, char or CO_2
Process mass intensity (PMI) and E-factor	PMI = total mass input ÷ mass of product; E-factor = mass of waste ÷ mass of product	Captures solvents, reagents and residues
Energy and hydrogen demand	Net heat, electricity and H_2 per tonne of product	Often the dominant driver of life-cycle impact
Residues and emissions	Char, ash, spent catalyst, brine, HCl and other emissions per tonne of feed	Reveals hidden burdens and disposal needs
Catalyst performance	Turnover, deactivation, recyclability, metal loading	Governs cost and scalability
Life-cycle and cost metrics	GHG, fossil-energy use, other categories; minimum selling price; sensitivity ranges	Enables comparison under stated counterfactuals

5. Technical and system challenges

Feedstock heterogeneity is the central obstacle. Halogens, nitrogen, oxygen, metals and fillers deactivate catalysts, corrode equipment and contaminate products [6]; pyrolysis oils intended for steam crackers therefore need decontamination strategies whose cost and benefit must be weighed [33]. Reviews of the field also highlight energy demand, scale-up, and the lack of harmonized definitions and metrics [11,34]. Table 4 pairs the main challenges with mitigation strategies.

Table 4. Main challenges and mitigation strategies

Challenge	Consequence	Mitigation strategies
Mixed or contaminated feed	Catalyst poisoning, off-spec products, fouling	Improved sorting; pre-treatment (washing, dechlorination); guard beds; tolerant catalysts
Low selectivity of C–C cleavage	Broad product distributions requiring upgrading	Tandem and shape-selective catalysts; tuned H_2 pressure and temperature; product-driven process design
High energy and hydrogen demand	Climate benefit eroded	Lower-temperature routes; heat integration; low-carbon electricity and H_2
Mass transfer in viscous polymer melts	Slow rates, limited catalyst access	Solvents or swelling agents; reactor design; interfacial catalysts [15]
Inconsistent LCA/TEA practice	Results not comparable	Common functional units, boundaries and counterfactuals; transparent sensitivity analysis [32]
Product market and quality	Recovered output has no buyer	Early engagement with downstream users; specification-driven development

6. Outlook and route-selection logic

Figure 9 summarizes a decision logic consistent with the evidence above: prevention and reuse first; mechanical recycling where quality allows; depolymerization for clean, mono-material condensation polymers; and C–C-cleaving routes for polyolefin-rich or mixed residues, with contaminant control and residual disposal as the final tier. Each branch should be confirmed by mass balance, life-cycle assessment and techno-economic analysis.

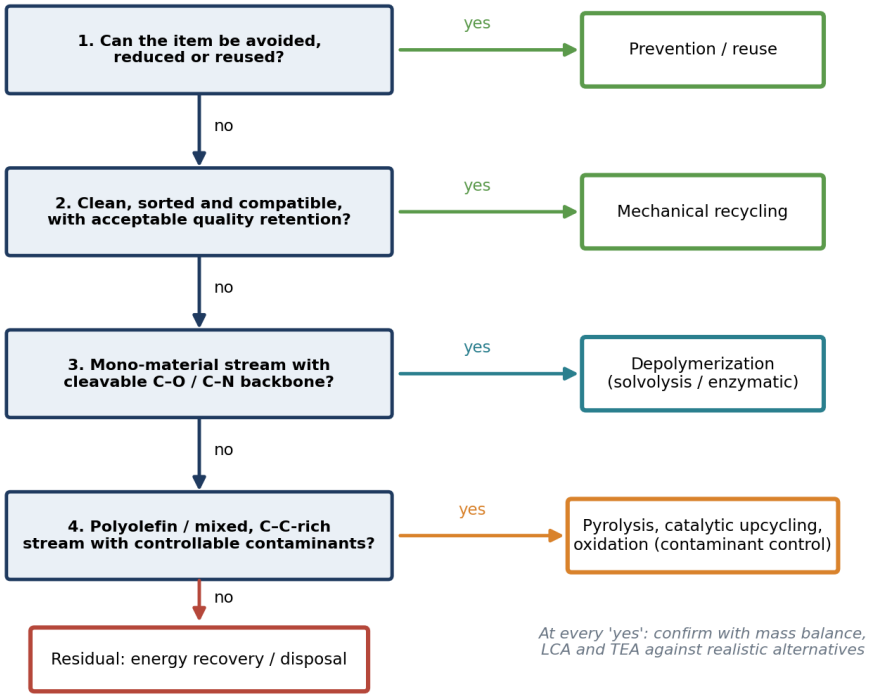

Figure 9. Route-selection logic (a decision aid, not a substitute for quantitative assessment).

Priorities for future work include:

- Catalysts and enzymes that tolerate realistic, contaminated feedstocks, with reporting of deactivation and regeneration.
- Selective C–C cleavage to defined products (for example olefins, lubricant base oils, aromatics) rather than broad mixtures.

- Polymers and additives designed for recyclability, including chemical recycling to monomer [18].
- Standardized reporting (Table 3), open datasets and harmonized LCA/TEA practice.
- Integration with low-carbon energy and hydrogen, and with collection and sorting infrastructure.

7. Conclusions

Chemical recycling and upcycling can extend plastic circularity to streams that mechanical recycling cannot serve, but their benefit is route-, feedstock- and system-specific. Heteroatom-containing polymers such as PET can be depolymerized selectively by solvolysis or enzymes; polyolefins require C–C cleavage, where selectivity and energy demand are the main hurdles; and mixed wastes remain the hardest case. Comparative studies show trade-offs between cost, environmental impact and product quality, and conclusions are sensitive to boundaries and counterfactuals. Progress therefore depends as much on rigorous, transparent assessment and on feedstock control as on new catalysts.

Declarations

Author contributions, funding and competing interests: to be completed by the authors.

Use of AI tools: AI-assisted tools were used for language editing, figure generation and reference checking. The author remain responsible for the content and confirms every citation against the original publication.

Data availability: no new experimental data were generated; figures redraw published values or are qualitative as labelled.

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