



Cite this: *Green Chem.*, 2024, **26**, 11832

Advances in catalytic chemical recycling of synthetic textiles

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Synthetic fibres cover most of the textile market, but their value chain is almost entirely linear. Common raw materials are non-renewable and oil-derived while requiring large amounts of (toxic) chemicals and energy for their processing into final products. In addition, synthetic textiles are usually non-bio-degradable polymers; therefore, sustainable approaches for their depolymerisation into reusable monomers have not been implemented yet. As a result, most post-consumer synthetic textile waste ends up being landfilled, dispersed in the environment or incinerated, thus contributing significantly to global pollution. A possible solution to this issue is the design and use of advanced catalysts for their chemical recycling. This manuscript reviews the most significant approaches that appeared in the literature in the time span of 2015–2024, covering the selective depolymerisation process of synthetic waste textile to added-value reusable monomers using chemical catalysts. Unselective processes, for example, to produce fuel mixtures, biocatalytic methods and depolymerisation of polyolefins are not covered. The general aspects of the catalytic depolymerisation of synthetic polymers are briefly discussed, and the catalytic chemical recycling of synthetic textiles is detailed by the polymer type. While contributing to the overall achievement of the sustainable development goals, chemical recycling of synthetic textile waste may represent a useful strategy toward the circularity of the textile sector, which is almost unexplored.

Received 23rd September 2024,
Accepted 23rd October 2024

DOI: 10.1039/d4gc04768k

rsc.li/greenchem

1. Introduction

Synthetic textiles are widespread in our everyday life. They are present in clothes, shoes, home furnishings (carpets, curtains,

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Fig. 1 Possible classification of main fibre types with representative examples. Adapted from ref. 4. Fibres covered by the present review are circled in red.

couches), accessories (towels, cushions), safetywear, workwear and sportswear. They are also present in technical equipment and in the seats of cars, trains and aircraft. As opposed to textiles from natural fibres, which are derived from living organisms (animals, plants), synthetic textiles are made of artificial organic polymers manufactured by chemical (co)polymerisation processes, mostly using non-renewable, fossil-sourced monomers. Common raw materials are crude oil, natural gas, and aromatic hydrocarbons, which are processed into olefins, glycols, acids and nitrile intermediates.¹ Other man-made fibres exist, for instance, those obtained by the chemical treatment of naturally occurring polymers (e.g. cellulose) and inorganic materials, however, with more limited use. The classi-

fication and nomenclature of man-made fibres are available from The International Bureau for Standardisation of Man-made Fibres² and schematically reported in Fig. 1. The use of (generic) textile fibre names and related labelling is defined by the EU Regulation 1007/201.^{3,4} Classification of synthetic polymers is usually based on the chain linkage, either polyesters, polyamides, polyurethanes, acrylics or polyolefins, which are the most popular ones (Table 1).

Synthetic fibres are relatively inexpensive and versatile. Compared to natural fibres, they have significantly lower prices (Fig. 2)⁵ and environmental costs due to their manufacturing process (Fig. 3), whereby cotton poses severe concerns in terms of land and water consumption. Synthetic fibres are



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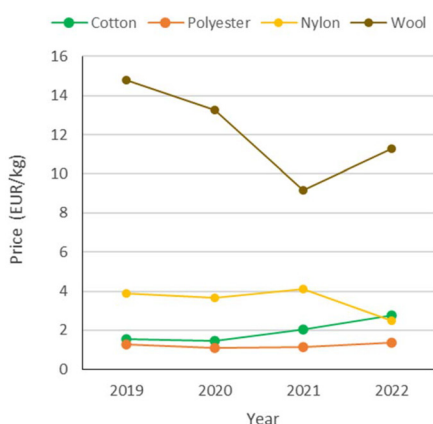
Science and Inorganic Systems" and in "Organic Chemistry" (2018). She is an investigator in several Industrial and European Projects and Research Contracts, co-editor of 1 Book with ISBN, and co-author of 38 articles in ISI journals, and she has 4 patents. Research interests: heterogeneous catalysis, green chemistry and sustainable processes.



Table 1 Classification of synthetic fibres based on the polymer chain linkage, with sketches and representative examples

Polymer	Polyesters	Polyamides	Polyurethanes	Acrylics	Polyolefin
Linkage					
Examples ^a	PET PBT PTT	Nylon 6 Nylon 6,6	EL	PAN MAC	PE PP

^a Abbreviations: PET, poly(ethylene terephthalate); PBT, poly(butylene terephthalate); PTT, poly(trimethylene terephthalate); nylon 6, polycaprolactam; nylon 6,6, poly(hexamethylene adipamide); EL, elastane; PAN, polyacrylonitrile; MAC, modacrylic; PE, polyethylene; PP, polypropylene.

**Fig. 2** Price trends of representative natural and synthetic fibres: cotton, polyester, nylon (polyamide) and wool. Data from ref. 5.

	CO ₂ emissions	Land use	Water use	Mineral resource use	Fossil resource use	Chemicals use
Polyesters	M	M	L	M	M	M
Polyamides	H	L	M	L	H	H
Polyurethanes	M	M	M	M	M	M
Acrylics	M	M	M	M	M	M
Cotton	L	H	H	H	H	M

Fig. 3 Schematic comparison of the environmental impacts of the manufacturing of identical amounts of selected fabrics: H, high; M, moderate; L, low. Based on estimates from ref. 4.

provided with specific properties, including strength, resistance to shrinking, elasticity and quick drying, which allow the production of a variety of durable fabrics. The large market potential associated with it has resulted in the so-called “fast-fashion” business model, *i.e.* the “cheap and mass manufacturing, frequent consumption and use of short-lived garments of clothes replicating high-fashion design”.⁶ Thus, clothes made of synthetic textiles have become quick consumer goods, which are rapidly produced as the demand is growing.

All of the above justifies the current fibre market as being dominated by synthetic materials. It is estimated that the global fibre production was around 109 million tonnes in 2020, with a 62% share of synthetic ones well above that of plant-sourced (polyesters 57 Mton, polyamides 5 Mton, cotton

26 Mton), as represented in Fig. 4.^{7,8} The manufacturing of synthetic fibres steadily increased over the last forty years. It surpassed that of cotton in late 1990s, with an average annual growth rate of around 7% during the period 2009–2019 (cotton 1.5%), which is expected to increase further to *ca.* 81 million tonnes in 2025, as shown in Fig. 5.

Unfortunately, besides their unquestionable benefits, synthetic fibres also have drawbacks. Unlike natural fibres and analogously to other artificial polymers (*i.e.* plastics), most synthetic fibres are not biodegradable,⁹ meant as “susceptible to lower their molar masses by an enzymatic process resulting from the action of cells”.¹⁰ Hence, they considerably contribute to the amount of domestic and industrial waste that may accumulate in the environment if not processed. With around 300 million tonnes per year, plastics are a major portion of the non-biodegradable waste globally generated,¹¹ where synthetic textiles provide one of the highest shares per industrial sector (*ca.* 14%, Fig. 6).^{12,13} Most synthetic textiles produced (*ca.* 60 Mton) ends up being trashed (*ca.* 40 Mton), which represents an unexploited resource potentially reusable in production streams. The amount and composition of textile waste may significantly vary with source and location. For instance, around

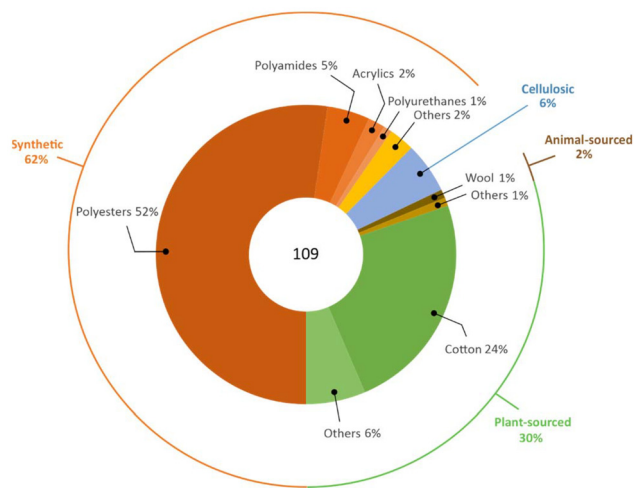
**Fig. 4** Global production of textile fibres in 2020 (million tonnes). Data from ref. 7 and 8.



Fig. 5 Global fibres production in the period 1975–2020. Data from ref. 7 and 8.

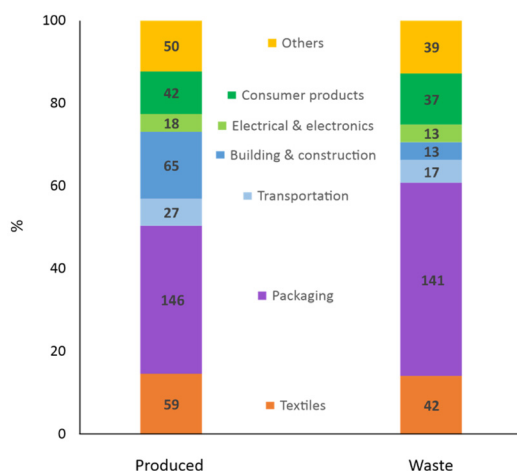


Fig. 6 Global primary plastics production (left) and waste generation (right) in million tonnes per industrial use sector. Data of 2015 from ref. 13.

13% (w/w) of the total municipal solid waste in the USA in 2017 were plastic materials,¹⁴ while plastics represented *ca.* 10% (w/w) of the separate collection of municipal solid waste in Italy in 2021.¹⁵ Batch-automated sorting tests of post-consumer textile waste collected from the Greater Copenhagen area in 2021 revealed the amount of synthetic fibres to be at least 35% (w/w) in both wearable and non-wearable materials,¹⁶ as shown in Fig. 7.

Despite the remarkable CO₂ emissions, use of energy, resources and chemicals associated with textile production (see Fig. 3),^{4,6} only a minor portion of the textile waste is recovered for reuse or recycling.^{17,18} It has been estimated that most post-consumer clothing fibres end up being dispersed in the habitat, landfilled or incinerated (overall *ca.* 88% w/w), whereas a modest 12% is reused in similar and lower-value goods or recycled into substantially different applications after

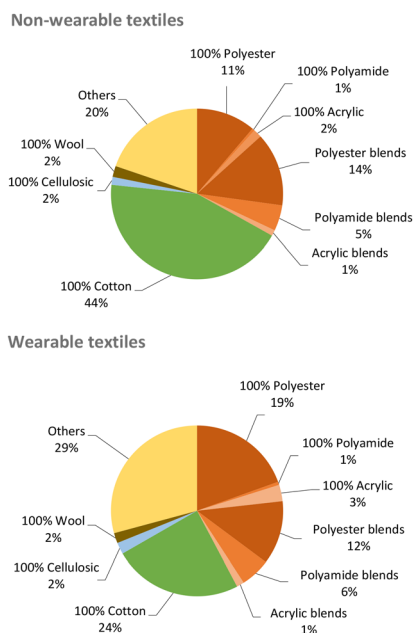


Fig. 7 Material composition of non-wearable (top) and wearable (bottom) textile waste collected from the Greater Copenhagen area in 2021. Data from ref. 16.

mechanical or chemical treatment,¹⁹ as summarised in Fig. 8.²⁰ Downcycling of waste clothes to insulation materials, mattress stuffing or cleaning cloths is around 11%, whilst *ca.* 1% are reused alike. Only a smaller portion of the recovered wearable textiles currently undergoes selective depolymerisation,²¹ which is usually referred as chemical recycling (*vide infra*). Whereas (mechanical) recycling of synthetic polymer waste in general (non-fibre plastic, *e.g.* packaging, bottles) has achieved technological maturity and significant volumes, to date, recycling of synthetic textiles shows comparably lower share and size. According to a study by Plastics Europe, recycling of the EU post-consumer houseware, leisure and sports plastic goods is around 7% of the amount produced (36% landfilled, 57% incinerated), whereas plastic packaging materials are recycled by 46%.²² The reason for this shortfall is attributable to a combination of factors, which include^{23,24}

- Waste textiles often consist of composite or mixed, hardly separable materials made up of several additives and dyes,
- Waste clothes may contain trims, hard parts and fasteners (*e.g.* buttons, zippers),
- Sorting technologies are inadequately developed,
- Chemical recycling methods for synthetic polymers are still in their infancy and mostly operating at the lab or pilot scale,
- Focused policies and regulations on textile recycling are lacking,
- Public awareness and motivation are insufficient,
- Collection options for textile waste are unsatisfactory.

The above considerations partly explain why a circular economy for synthetic textiles has not yet been developed,



Fig. 8 Global raw materials source and fate for clothes in 2015. Data from ref. 20.

whereby textile recycling facilities are basically limited to cotton. Particularly, separate collection and sorting, which is usually performed through a sequence of automated infrared spectroscopy and X-ray fluorescence steps,^{25,26} is deemed crucial, as it is estimated they may provide up to a 13-fold increase in waste plastic recycling rates.²² As a consequence, the synthetic textile industry ranks second in the contribution to global pollution.²⁷ Synthetic fibres in apparel have been identified as the major factor responsible for microplastic contamination of oceans^{28,29} simply as a result of cloth washings.³⁰

To address these challenges, focused strategies and policies for textile recycling have been released both at the European and National levels.^{31,32} The Circular Economy Action Plan, as a part of the European Green Deal, identifies plastics and textiles as priority fields of intervention.³³ The EU Strategy for Sustainable and Circular Textiles,³⁴ and other specific measures for plastic recycling,³⁵ have been launched in response to the EC Waste Framework Directive, which declares that the Member States shall set up separate collections for textiles by 1 January 2025.^{36,37} The European Environmental Agency in 2019³⁸ and the Organisation for Economic Co-operation and Development (OECD) in 2020³⁹ have indicated that recycling synthetic fibres is key to achieving a viable circular system for textiles. Textile Recycling Hubs have been proposed both in Europe⁴⁰ and Italy,⁴¹ the latter as a part of the National Recovery and Resilience Plan funded by the Next Generation EU programme,⁴² aimed at strengthening the recovery, mechanical and chemical recycling of textiles. Both the German Federal Ministry for Economic Cooperation and Development⁴³ and the Nordic Region Cooperation⁴⁴ have proposed their own agendas for textile fibre recycling. The European Clothing Action Plan project,⁴⁵ funded under the EU LIFE scheme, aims to drive fashion and textile sectors toward circularity, focusing, *inter alia*, on diverting clothing waste from landfills and incineration through resource-efficient design and recycling. It was highlighted that, besides targeting the minimisation of environmental impact and depletion of natural resources, the transition toward a circular economy model for synthetic textiles represents a significant opportunity in terms of new jobs and manufacturing.^{46,47}

The next section summarises the main strategies in use for recycling synthetic textiles with a focus on selective depolymer-

isations using chemo-catalytic methods. A number of reviews have been previously published on textile recycling, dealing with natural fibres,^{48,49} general aspects of recycling techniques,^{50,51} biocatalytic methods⁵² engineering,^{53,54} or economic issues.^{55,56}

2. General concepts of catalytic chemical recycling of synthetic textiles

Various strategies are in place for the recycling of synthetic polymers, including textiles. A generally accepted classification includes zero-order recycling (*i.e.* reuse), mechanical recycling, chemical recycling, and energy recovery (*i.e.* combustion).^{57,58} Mechanical recycling refers to those methods in which the polymeric structure of the material is not modified, while changing the appearance and the use, which is usually achieved by grinding, melting and assembling of new parts and products, possibly using additives or blending with fresh polymers.^{59,60} Chemical recycling is less unambiguously defined, whereby the lack of a clear nomenclature and the use of very broad terms may lead to occasional inconsistencies,^{61,62} and to a misunderstanding of the role of chemical recycling in a circular economy perspective.^{63,64} Among the various definitions proposed, the one we believe is the most accurate and representative of the circularity concept is that provided by Plastics Europe: “Chemical recycling converts, *e.g.* polymeric waste by changing its chemical structure to produce substances that are used as products or as raw materials for the manufacturing of products. Products exclude those used as fuels or means to generate energy”.²² From this definition, it is clear that all those unselective, thermal conversion processes (300–1000 °C), leading to small molecules or energy vectors (CO, CO₂, CH₄, H₂), light hydrocarbons, diesel oil or fuel mixtures, are not strictly considered as “chemical recycling”.^{65,66} These processes include pyrolysis,^{67,68} catalytic (hydro)cracking^{69,70} and gasification.⁷¹ By contrast, chemolysis techniques, which use chemical reagents to achieve selective depolymerisation, allow obtaining reprocessible monomers, building blocks or finished chemicals, thus providing valuable products for the process industry and ultimately converting plastic waste into useful materials.^{72,73} These processes are usually performed using a solvent (*i.e.* hydrolysis, alcoholysis, glycolysis, aminolysis),^{74,75} or dihydrogen (*i.e.* hydrogenolysis),⁷⁶ as chain linkage cleaving agents. A variety of products, including alcohols, acids, amines and esters can be obtained, depending on the starting polymer and the cleaving agent used. Chemolytic processes may benefit from the use of a catalyst, as it will be illustrated in the following paragraphs. Other techniques are sometimes referred as “chemical” recycling, though they are not. These include dissolution/precipitation, where a solvent is used to extract and purify a synthetic organic polymer from contaminants or other materials.^{77,78}

Chemical recycling is to be considered complementary to mechanical recycling: not all synthetic polymer waste can be mechanically recycled and mechanical recycling cannot be



reiterated indefinitely because of the gradual quality loss of the recycled material due to oxidation, hydrolysis or mechanical degradation.^{79,80} Chemical recycling *via* chemolysis allows the treatment of difficult-to-recycle plastic waste and those synthetic polymers that are not recyclable otherwise.⁸¹ By converting a synthetic polymer into chemicals, chemical recycling may enable a true circular economy for plastics, where plastic wastes represent secondary raw materials, from which myriads of products can be re-manufactured, including solvents, (pharma)building blocks, dyes, additives and monomers, thus to replace the virgin fossil-based ones, as sketched in Fig. 9.⁸² For this reason, the term “plastic refinery” has been proposed to identify an alternative facility to conventional petrol-based plants.⁸³ Noteworthy, “closed-loop” catalytic systems have also been developed, whereby easily depolymerisable polymers can be efficiently re-polymerised under mild conditions.^{84,85} Thus, chemical recycling of synthetic polymers is likely one of the clearest application examples of the circular chemistry concept.⁸⁶ Besides the significant benefits in terms of the reduction of landfills, incineration and consumption of non-renewable sources, chemical recycling of synthetic textiles may significantly contribute to the reduction of CO₂ emissions with respect to the production of virgin materials.^{87,88} In the case of polyesters, it has been estimated that alcoholysis provides a 60–70% greenhouse gas (GHG) emissions saving, compared to the conventional manufacturing sequence.⁸⁹ Consequently, chemical recycling technologies are considered indispensable in view of a transition to a climate-neutral economy. European plastics manufacturers plan to invest 7.2 billion euros in chemical recycling by 2030, whilst the production of recycled plastics *via* chemical recycling is estimated to increase from 0.1 million tonnes in 2020 to 3.4 million tonnes in 2030.²²

However, current chemical recycling *via* chemolysis suffers from several limitations, particularly concerning synthetic textiles. One reason for this is that the related technologies are poorly developed, and the research in the field is relatively young.⁹⁰ To date, most chemolytic depolymerisation processes

rely on organic (noxious) solvents, harsh reaction conditions, high pressures of gas, need for soluble promoters, corrosion issues, limited selectivity, and generation of a considerable amount of (salt) waste, which ultimately result in costly downstream purifications and high energy inputs.^{91,92} Applicability to specific waste streams may also be an issue.⁹³ Overall, chemolytic methods have to face economic and environmental sustainability indicators, whereby technological and financial viability, integration with the local market, and impact on human health and habitat are key parameters to be taken into account.⁹⁴ So far, chemical recycling is rarely competitive with mechanical recycling, which is more developed and hence favoured.^{95,96} Advancements in chemolysis processes are mostly expected in relation to improved selectivity and more friendly reaction conditions.

Understandably, depolymerisation of plastics is hampered by the recalcitrance to chain scission due to thermodynamic and kinetic barriers, which depend on the polymer composition and structure.⁹⁷ To achieve efficient and selective depolymerisation at reasonable reaction temperatures (*e.g.* $\leq$ ceiling temperature⁹⁸), the polymer substrate should originate from low exergonic polymerisation reactions ($\Delta G_{\text{polymerisation}} \approx 0$).^{99,100} For instance, this is not the case for polyolefins because of the inherent strength of the C–C bonds. Therefore, polyolefins usually depolymerise with poor selectivity under harsh conditions.^{101,102} Moreover, even if thermodynamic requirements are favourable, high activation energies may slow down the depolymerisation kinetics, thus resulting in long reaction times for complete depolymerisation to occur at low temperatures. To circumvent this limitation, a possible solution is using a catalyst.^{103,104} The clear advantage of catalysis is decreasing the activation barrier, thereby minimizing energy inputs, while maximising reaction rates, monomer yields and selectivity.^{105,106} Representative examples may illustrate the macroscopic benefits. Compared to the catalyst-free process, the hydrolysis reaction of polylactic acid (PLA) has been reported to occur in 100% yield at a 30 °C lower temperature in the same reaction time using ZnO catalyst. In the case of PET, analogous results were obtained at a reaction temperature that is 60 °C lower.¹⁰⁷ In other words, under the same reaction temperature (*i.e.* 130 and 180 °C for PLA and PET, respectively), the monomer yield was around eight times higher over the ZnO catalyst compared to the uncatalysed reactions. In another example, the monomer yield was around four times higher for the glycolysis reaction of PET at 190 °C after 1 hour using titanium(IV) *n*-butoxide as the catalyst, with respect to the uncatalysed reaction at 220 °C.¹⁰⁸ In terms of the activation energy, the hydrolytic depolymerisation of nylon 6 to ϵ -caprolactam, in the temperature range 553–603 K, was *ca.* 9.26 kJ mol^{−1} lower using 3% phosphotungstic heteropoly acid catalyst, compared to the catalyst-free process.¹⁰⁹

Still, problems with chemo-catalytic recycling arise from the use of homogeneous catalysts,¹¹⁰ strong acid–base soluble activators,¹¹¹ poor efficiency of biological catalysts,^{112,113} and the need for toxic promoters or solvents.^{114,115} Heterogeneous catalysts are preferred at the industrial level due to the ease of



Fig. 9 Circular economy model based on molecular recycling of synthetic textile waste. A variety of products can be manufactured, including virgin textiles (green loop) or packaging materials (blue loop).



separation, recovery, reuse and integration into existing reactor equipment, including semi-batch and continuous flow set-ups, which may result in improved process intensification, long-term productivity, product purity and reduction of downstream processing.^{116,117} Limitations may arise in terms of catalyst activity and durability, the need for catalyst regeneration and leaching of metal species.¹¹⁸ Particularly for polymeric substrates, further drawbacks originate from their insolubility in common solvents, which results in restricted accessibility of the bulk polymer to a solid catalyst interface, where reactive interactions occur.¹¹⁹ Hence, high reaction temperatures and stirring rates may be needed using heterogeneous catalysts in order to enhance the diffusion rate of reactants within the polymer and the catalyst.^{120,121} Therefore, chemolytic depolymerisations *via* heterogeneous catalysis usually necessitate the design of more sophisticated systems, for instance, involving materials having large surface areas, porosities and stability.^{122,123} On the other hand, for the above-mentioned reasons, homogeneous catalysts for depolymerisation are usually significantly more active compared to heterogeneous, although high catalyst loadings (*e.g.* alkali hydroxides, ZnCl_2), sophisticated metal ligands (*e.g.* for pincer molecular complexes), toxic or corrosive species (*e.g.* Zn salts, H_2SO_4) may be required.^{124,125} In addition, issues related to catalyst/product separation, metal contamination, generation of considerable amounts of salt waste streams and non-reusability of the catalyst may contribute to increasing the process costs, thus making the system economically unattractive. Irrespective of the catalyst, whether homogeneous or heterogeneous, the mechanism of solvolytic depolymerisations usually involves the acidic/basic activation of the functional group chain linkages (*e.g.* ester) by the appropriate catalytically active species. This is, for instance, the case of the hydrolysis or glycolysis reaction of polyesters mediated by conventional, soluble Zn^{2+} salts, where the Zn ions act as Lewis acid centres toward the carbonyl ester bonds, which then undergo a nucleophilic attack by the solvent molecules, leading to chain scission.^{126,127} The mechanism of catalytic hydrogenative depolymerisation is less investigated, though it usually involves ruthenium centers due to the high affinity for $\text{C}=\text{O}$ bonds and is achieved using soluble molecular complexes, where the cleavage of the chain linkage occurs in a concerted manner by a heterolytic route.^{128,129}

In the case of synthetic textiles, the reported strategies for chemical recycling are analogous to those used for other plastic materials. However, additional hurdles come from the complexity of textile goods, particularly in relation to macroscopic aspects (*e.g.* size, mechanical resistance, blending with different fibre types, presence of additives, contaminants and hard parts),³¹ as outlined in section 1. Therefore, besides disassembling, sorting and selection, pre-treatments may be required to achieve effective depolymerisation. Typical is the case of dissolution in organic solvents or contact with acids. Pre-treatment methods are polymer-dependent; hence, they will be described case-by-case in the next section, *e.g.* polyurethanes in section 3.3.

The next sections review the most significant advancements recently achieved in the catalytic chemical recycling of synthetic textiles, broken down by polymer type. No polyolefins are considered due to the fuel product mixtures usually obtained.

3. Applications of catalytic chemical recycling of synthetic textiles

3.1. Polyesters

Polyesters, particularly polyethylene terephthalate (PET), are the most common synthetic yarns. PET is also widely used in the food and beverage packaging industry. The global production of PET is around 72 Mt per year (2019), and 65% is used in fabrics for clothes, sportswear and home furnishings.^{130,131} In addition to its ubiquity, PET is hardly biodegradable,^{132,133} which justifies PET being among the polymers most investigated for chemical recycling. Several reviews are available in the literature describing the strategies for PET depolymerisation in general.^{134,135} Most methods rely on the use of soluble catalysts (*e.g.* NaOH , ZnCl_2 or ionic liquids), either for hydrolysis, alcoholysis and glycolysis reactions under harsh conditions, or for hydrogenolysis (*e.g.* pincer Ru complexes), the former with drawbacks in terms of energy inputs, corrosion, downstream purification, and management of salt waste.¹³⁶ In a few instances, the use of insoluble catalysts has been described (*e.g.* ZnO , hydrotalcite).^{137,138} Solvolysis of polyesters may take advantage of strong Brønsted base or Lewis acid catalysts acting as activators of the carbonyl ester bond.¹³⁹ Two-step catalytic solvolysis-hydrogenation reactions in cascade have also been reported.^{140,141}

PET fibres are made of ethylene glycol and valued petroleum-derived terephthalic acid (TPA),^{142,143} but they are often blended with other natural materials, *e.g.* cotton, to obtain textiles for use in shirts and blouses. Common trade names include Trevira, Diolen, Dacron, and Coolmax. An overview has been published on the recycling (mechanical, thermomechanical, thermochemical, chemical, and biochemical) of PET from cellulose blended fabrics.¹⁴⁴ Several synthetic additives are used in the manufacturing of PET textiles, mostly as finishing to provide fire retardants, antistatic, antimicrobial, water or stain-repelling properties, or as colorants. Coatings may contribute up to 5–20% of the weight of the final textile product. Additives may not degrade or be removed easily,¹⁴⁵ while having toxic potential, as warned by the European Chemicals Agency (ECHA).¹⁴⁶ Recent studies on the recycling of PET-based textiles mainly concern sorting,¹⁴⁷ dissolution-separation techniques,^{148,149} removal of dyes,^{150,151} and the effect of additives on recycling.^{152,153} A few reports exist on the catalytic depolymerisation of PET-based textiles, which include glycolysis, hydrolysis and methanolysis reactions (Fig. 10). A summary of recent findings is reported in Table 2.

Glycolysis is usually performed using an excess of ethylene glycol (EG) to yield bis(2-hydroxyethyl)terephthalate (BHET),



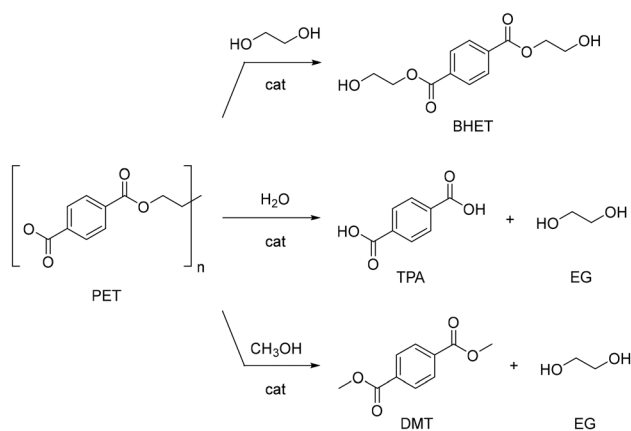


Fig. 10 General scheme of homo- and heterogeneously-catalysed glycolysis (top), hydrolysis (middle) and methanolysis (bottom) reactions of PET.

formally through a (reversible) transesterification reaction (Fig. 10, top). Drawbacks of the method may be the high reaction temperatures (≥ 190 °C) and the difficulty of purification of BHET. In an earlier paper, the glycolytic depolymerisation of PET fibre waste of 1.44 denier was achieved using 0.3–1% (w/w) sodium sulphate catalyst and 1 : 6 = PET : EG ratio at 197 °C. BHET was obtained in 60% yield after 8 h and crystallization from water.¹⁵⁴ In a further study, a two-step treatment of post-consumer polyester clothes, based on the addition of 40 wt% alkali solution (1 h, 100 °C), followed by glycolysis with 1% zinc acetate catalyst (PET : EG = 1 : 5 (w/w), 180 °C, 8 h), gave BHET in *ca.* 85% yield, after washing with water to remove cellulose, titania contaminants, EG, sodium and zinc salts.¹⁵⁵ Similarly, 80% BHET yield was obtained by the treatment of PET fabric waste with water and acetone, followed by glycolysis using 0.2 wt% zinc acetate catalyst, PET : EG = 1 : 2 (w/w), 198 °C, 4 h, and crystallisation.¹⁵⁶ More recently, the glycolysis of white and uncoloured post-industrial PET fibre wastes was reported over insoluble Mg–Al double oxide pellet catalyst (diameter 6 mm, height 8 mm, 1.5 wt%) to give BHET in *ca.* 80% yield after 2 h at 240 °C (PET : EG = 1 : 5). The catalyst could be reused three times with no significant decrease in catalytic activity, after which regeneration at 1000 °C was required.¹⁵⁷ Other solid catalysts used include 2 mol% Ag-

doped ZnO nanoparticles for the depolymerisation of red, blue, and green PET textile waste threads under microwave irradiation. Around 90% BHET yield was obtained at full PET conversion after recrystallization (5 wt% catalyst to PET ratio, 300 W, 30 min, 200 °C, PET : EG = 1 : 44 w/w).¹⁵⁸ Under similar conditions, 10 wt% In-doped ZnO-carbon nitride nanocomposite catalyst gave BHET in 90% yield after extraction with chloroform from the water solution washings.¹⁵⁹ The heterogeneous catalyst could be reused with *ca.* 5% BHET yield loss upon each recycling.

The hydrolytic depolymerisation of PET produces EG and water-insoluble TPA (Fig. 10, middle). Hydrolysis reactions clearly benefit the use of water as a friendly and economic reagent; however, harsh conditions are needed to achieve complete depolymerisation without catalysts. For instance, in the case of textiles, PET fibres with different viscosities and containing titanium dioxide to reduce luster were depolymerised to TPA in 92 wt% yield under 250 °C, 40 bar pressure, PET : H₂O ratio 1 : 10, and hydrolysis time of 30 min after reaching the steady-state conditions.¹⁶⁰ By contrast, alkali-catalysed hydrolysis of viscose/PET fabrics (70/30) (5 wt% NaOH, 24 h) allowed producing pure TPA in *ca.* 99% yield under 90 °C reaction temperature, with concurrent cellulose recovery.^{161,162} TPA was isolated after filtration of the aqueous reaction mixture and neutralization with H₂SO₄. After the fabric treatment, the intrinsic viscosity of the recovered cellulose was decreased by up to 35%. An in-depth techno-economic and process upscaling analysis was included, indicating the need to reduce the consumption of (costly) NaOH, increase the hydrolysis rate by adopting appropriate catalysts, and improve the downstream separation and purification technologies from the resulting EG/Na₂SO₄ stream. Taking advantage of the fact that TPA acts as a linker in metal–organic frameworks (MOF), in a recent work, TPA was extracted by reactive crystallization from mixed-TPA fibres (cotton, dyes) in the form of Ga-TPA MOF (10% NaOH, 200 °C, 200 min, Ga(NO₃)₃, 210 °C, 5 h).¹⁶³ Pure TPA was then obtained by decomposition of the MOF by nitric acid treatment.

In a recent paper, the methanolysis reaction of PET fibres to give dimethyl terephthalate (DMT) and EG was described using triethylamine as an organocatalyst (Fig. 10, bottom).¹⁶⁴ Running the reaction with an excess of methanol, 7% (v/v) NEt₃ and 200 °C resulted in an 82% DMT yield, which was

Table 2 Representative literature results for the lab-scale catalytic chemical recycling of PET textiles^a

Method	Catalyst	Catalyst type	Reaction conditions	Pre-treatment	Product (yield)	Ref.
Glycolysis	Zn acetate	Homogeneous	180 °C, PET : EG = 1 : 5	40% NaOH, 100 °C	BHET (85%)	155
Glycolysis	Zn acetate	Homogeneous	198 °C, PET : EG = 1 : 2	Water/acetone	BHET (80%)	156
Glycolysis	Mg–Al double oxides	Heterogeneous	240 °C, PET : EG = 1 : 5	n.a.	BHET (80%)	157
Glycolysis	Ag-doped ZnO NP	Heterogeneous	200 °C, PET : EG = 1 : 44	Water	BHET (90%)	158
Glycolysis	In-doped ZnO/C ₃ N ₄	Heterogeneous	200 °C, PET : EG = 1 : 22	200 °C	BHET (90%)	159
Hydrolysis	None		250 °C, 40 bar, PET : H ₂ O = 1 : 10	n.a.	TPA (92%)	160
Hydrolysis ^b	NaOH	Homogeneous	90 °C, PET : H ₂ O = 1 : 100	Water 60 °C	TPA (99%)	161
Methanolysis	Triethylamine	Homogeneous	200 °C, PET : MeOH = 1 : 12	n.a.	DMT (82%)	164

^a Abbreviations: NP, nanoparticles; C₃N₄, carbon nitride; MeOH, methanol. ^b Viscose/PET blend.



brought to 88% using a 1 : 1 (v : v) methanol : toluene mixture. No solvolysis took place without a catalyst under the same condition. More than 91% of the initial solvent/catalyst volume was recovered after five depolymerization cycles. Importantly, a life cycle assessment (LCA) analysis showed the process of avoiding the emission of 1.88 kg of CO₂ equivalent compared to the conventional DMT manufacturing route. DMT could be crystallized in 56% yield from the methanolysis reaction of dye-free polycotton (50% polyester/cotton).

Generally, the choice of the solvolytic method may be driven by the comparison of environmental, technical and cost factors.^{165,166} For instance, EG is not warned by the REACH regulation and may be produced from renewable sources; however, its high boiling point may result in difficulties in product separation and in the management of the excesses. By contrast, methanol, although toxic, is a better nucleophile than other alcohols; hence, using methanol often results in faster reaction kinetics and higher depolymerisation yields. Of course, water is the cheapest and safer option; still, shortcomings may arise from the poor solubility of reaction products (e.g. TPA), need for strongly basic conditions (e.g. NaOH) to achieve full depolymerisation, and instability of catalysts in the presence of nascent carboxylic acid generated upon hydrolysis.¹⁰⁷ In addition, it must be considered that all products obtained from the glycolysis, hydrolysis and methanolysis reaction of waste PET fabrics (i.e. TPA, BHET, DMT) are usable intermediates for manufacturing the virgin polymer or other plastic materials for diverse applications (polyesters, polyurethanes, epoxy resins),^{167,168} as well as acrylics, dyes or anti-bacterial drugs.^{169,170}

Finally, it is worth mentioning that the conversion of unspecified PET-based clothes to a mixture of benzene, toluene and xylene was reported over Ru@Nb₂O₅ catalyst, with a 46%, 46% and 8% selectivity, respectively, at an overall 81.6% hydrocarbon yield.¹⁷¹ The process conducted using water, a PET : catalyst ratio = 1 : 1 (w/w), 280 °C and 16 h, consists of a sequence of tandem hydrolysis, reforming, hydrogenolysis and decarboxylation steps, where the reforming of EG obtained by hydrolysis provides *in situ* generated H₂.

Industrial-scale technologies for the chemical recycling of synthetic textiles are still at an early market stage, whereby a few operate and others are under development.¹⁷² Because of their commercial nature, most of the related processes lack significant information concerning the reagents and catalysts (if any) used, energy inputs, nature of manageable streams, including contaminants, and the environmental performance. In the case of PET, the vast majority of recycled material comes from post-consumer bottles (ca. 99%), whereas only a minor amount is due to recycled textile waste or fabric processing residues.¹⁷³ Table 3 summarises the industrial processes claimed for the catalytic chemical recycling of synthetic textiles, broken down by polymer type, wherein at least basic details are available. Additional information on complementary technologies for synthetic textile recycling can be retrieved from the literature.^{7,92} Ioniqa, a spinoff from the Eindhoven University of Technology, has developed an efficient depolymerisation strategy based on glycolysis at 197 °C accelerated by an ionic liquid metal catalyst, namely bmimFeCl₄ (bmim = butylmethylimidazolium).^{174,175} Notably, linking the catalyst to magnetic particles enables catalyst recovery and reuse. Although the method uses PET bottles as feedstock, it is planned to be performed on PET fibres.¹⁷⁶ Jeplan has developed a “clothing-to-clothing” BRING Technology™ in which discarded PET clothing and fibres are converted to BHET by glycolysis, then re-polymerised to PET yarns.¹⁷⁷ The depolymerisation step is brought about by heating the input stream with an equivalent weight amount of BHET, followed by glycolysis at 200 °C using EG and sodium methylate as a catalyst.¹⁷⁸ The final BHET monomer is isolated after treating the product with activated carbon and ion-exchange resins. Japan-based Teijin Company announced to have developed a depolymerisation method for coloured polyester fibres to give BHET, arguably by glycolysis, using a new, undisclosed catalyst.¹⁷⁹ Italian Garbo is implementing a medium-scale production facility of BHET (55–65 ton per day) from PET waste, including fabrics, likely taking advantage of the Zn acetate catalyst.^{180,181} Similarly, Poseidon Plastics is producing BHET by glycolysis from several forms of waste PET plastic, including textiles and

Table 3 Industrial processes for the catalytic chemical recycling of synthetic textile streams to reprocessable products^a

Company	Input material	Method	Product	Catalyst	Catalyst type	Ref.
Ioniqa	PET	Glycolysis	BHET	[bmim][FeCl ₄]	Heterogeneous	174
Jeplan	PET	Glycolysis	BHET	Sodium methoxide	Homogeneous	177
Teijin	PET	Glycolysis	BHET	n.a.		179
Garbo	PET	Glycolysis	BHET	n.a.		181
Poseidon Plastics	PET	Glycolysis	BHET	Zn acetate–urea	Homogeneous	182
Axens	PET	Glycolysis	BHET	n.a.		185
NextChem	PET	Hydrolysis	TPA, EG	Alkali hydroxides	Homogeneous	187
Tyton BioSciences	PET and blends	Hydrolysis	TPA, EG	NaOH	Homogeneous	189
DePoly	PET	Hydrolysis	TPA, EG	n.a.		190
Mistra Future Fashion	Polycotton	Hydrolysis	TPA	NaOH, BTBAC	Homogeneous	191
Rittec	PET	Hydrolysis	TPA, EG	NaOH	Homogeneous	193
Eastman	PET	Methanolysis	DMT	n.a.		194
Loop Industries	PET	Methanolysis	DMT, TPA, EG	KOH, NaOH	Homogeneous	196
Aquafil	PA and composites	Hydrolysis	CPL	Acid	Homogeneous	230

^a Selective depolymerisation methods, see definition in section 2. Enzymatic methods and methods leading to fuel products are not covered.



carpets, at the Teesside facility (UK).¹⁸² Typical conditions involve the use of a zinc acetate–urea catalyst system ($(\text{NH}_2\text{CONH}_2)_n\text{Zn}(\text{CH}_3\text{CO}_2)_2$, around 190 °C reaction temperature, EG to PET ratio 4. BHET purification follows, which includes recrystallisation from alcohols and decolouration over cation exchange resins.^{183,184} Axens runs a demonstration plant based on the Rewind® PET glycolysis process for PET fibres.¹⁸⁵ No information on catalyst usage is available. As a part of the EU Horizon DEMETO project,¹⁸⁶ NextChem completed a demonstration plant in Italy for the depolymerisation of PET from textiles, co-licensed with the Swiss start-up Gr3n, using a microwave-assisted alkaline hydrolysis technology (NaOH, KOH or LiOH catalyst).^{187,188} US Tyton BioSciences has developed a hydrolysis method to recover TPA from PET waste textiles or PET/cotton blends, using 0.5% (w/v) NaOH at 180 °C to increase the reaction rate.¹⁸⁹ TPA is isolated by adjusting the pH to 2–4. EG can be recovered by conventional distillation. Swiss start-up DePoly company operates a low-temperature chemical recycling process for polyester textiles into TPA and EG, hence inferred *via* hydrolysis.¹⁹⁰ No information on catalyst use is provided. Mistra Future Fashion claims the Blend Re:Wind hydrolysis process for polycotton fibres, based on treatment with 10% NaOH, benzyltributylammonium chloride (BTBAC) and H_2SO_4 , in collaboration with the Chalmers University of Technology in Göteborg.^{191,192} The RevolTex® hydrolysis process has been developed by Rittec, in cooperation with the Braunschweig Technical University, for the depolymerisation of waste polyester textiles into TPA using concentrated NaOH solutions.¹⁹³ Eastman reported the chemical recycling of polyester plastic waste, including carpets and clothing, *via* glycolysis or methanolysis using an undisclosed catalyst.¹⁹⁴ The latter process was estimated to result in 29% lower greenhouse gas emissions compared to conventional DMT production from fossil sources.¹⁹⁵ Loop Industries' depolymerisation technology converts PET fibre waste into DMT, EG and TPA by treatment with 30–40% (w/w) KOH, methanol and dichloromethane at room temperature, followed by acidification with H_2SO_4 .^{196,197}

Other industrial processes are in place for PET textile waste recycling based on the dissolution/precipitation strategy, thereby not involving depolymerization. For example, Worn Again Technologies has patented a process to convert PET textiles, including cotton blends, into pellets.^{198,199} Another start-up company, Ambercycle, has developed a technology to purify and separate polyesters from different garments and is planning a demonstration plant to produce a metric ton of recycled material per day.^{200,201} Recycling polycotton fibres has been described as a treatment with sulfuric acid and water to yield cellulose powder and unreacted PET.²⁰²

3.2. Polyamides

Polyamides (PA), also known under the trade name nylons, are the second most common synthetic fibres. PA are largely used for sport- and underwear, hosiery, umbrellas, technical fabrics, ropes and carpets due to their suitable thermal and mechanical properties, including high strength, flexibility,

elasticity, toughness and low moisture absorption. It is estimated that textiles account for around 70% of global PA production (2018),²⁰³ whereas carpets contribute around 17%.²⁰⁴ Nylons are aliphatic condensation polymers formally derived from organic acids and amines, whereby homo-polymers resulting from amino-acids or lactams (PA *n*) or from dicarboxylic acids and diamines dyads (PA *n,n*) exist (*n* indicates the number of carbon atoms). The PA textiles market is dominated by PA 6 (*ca.* 55%) and PA 6,6 (*ca.* 35%) (Fig. 11).²⁰⁵

Similar to polyesters, depolymerisation of PA *via* chemical recycling is possible *via* hydrolysis, alcoholysis, glycolysis, aminolysis and hydrogenolysis reactions.^{206,207} By contrast, the superior resistance of PA results in more drastic reaction conditions. At the molecular level, this is attributable to the resonance stabilisation of the amide bond, which hampers the nucleophilic attack by lytic agents.²⁰⁸ In the case of PA 6,6, chemical recycling is further complicated by the strong intermolecular hydrogen bonds, resulting in a semi-crystalline polymer structure with higher tensile strength, melting point and lower solubility in common solvents.²⁰⁹ Most works addressing the depolymerisation of PA6 to monomers relate to the water-assisted conversion to ϵ -caprolactam (CPL) which, irrespective of the mechanism, is formally achieved through hydrolysis-cyclocondensation steps, usually at high reaction temperatures (240–600 °C), or aided by strong acidic or basic catalysts (H_3PO_4 , H_2SO_4 , HCl, NaOH, $\text{H}_3\text{PW}_{12}\text{O}_{40}$) (Fig. 12, top).^{210,211} In the case of PA 6,6, the process is limited by repolymerisation of the resulting acid and amine reaction mixture to provide polyamide resins.²¹² Despite the significant amount of literature related to these systems, few works describe the



Fig. 11 Structures of nylon 6 and nylon 6,6.

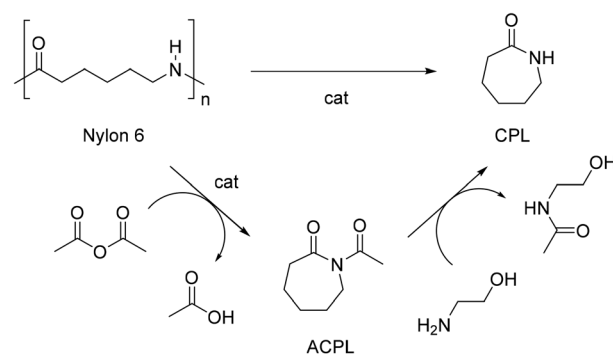


Fig. 12 Sketch of cyclodepolymerisation processes of nylon 6 to CPL. Top: conventional water-assisted or solvent-free reaction. Bottom: acetic anhydride fragmentation, followed by *trans*-acetylation.



depolymerisation of actual PA-based textiles, most likely due to the usual blending with other polymers (PET, polyolefins).²¹³ As a consequence, the recycling of nylon textiles is currently confined to dissolution–precipitation^{214,215} or energy recovery processes *via* incineration; however, it has serious drawbacks in terms of the emission of greenhouse gases (N₂O).²¹⁶ With the exception of unselective deconstruction processes (*e.g.* pyrolysis),²¹⁷ the catalytic depolymerisation studies of nylon-based textiles are briefly reviewed in the following paragraphs.

In an earlier study, the catalyst-free cyclodepolymerisation of nylon 6 carpets was reported in the presence of steam to give CPL in 90% yield, under 1.5 MPa and 340 °C (Fig. 12, top).^{218,219} More recently, the use of water under sub-critical conditions (*i.e.* below 374 °C and 22.1 MPa), with no external catalyst added, 300 °C reaction temperature, 60 min and 11 : 1 H₂O/nylon 6 mass ratio, resulted in *ca.* 91% CPL yield and 10 wt% solid residue.²²⁰ Nylon 6 waste was pulverised prior to use. A careful analysis of reaction by-products revealed the *n*-value of oligomers in the liquid phase to range from 0 to 5. Protons generated by water dissociation under the reaction conditions were ascribed to be the catalytically active species in that case. Friendlier reaction conditions were possible by the addition of catalysts. Shukla *et al.* reported the treatment of nylon 6 fibre waste using a concentrated aqueous solution of HCl (30%) or H₂SO₄ (15%) as an acid catalyst under reflux temperature, which resulted in low-molecular-weight compounds of various chain lengths and in 93% and 79% aminocaproic acid yield, respectively.²²¹ Nylon 6 processing wastes were also hydrolysed over a series of γ -Al₂O₃ supported solid acid catalysts and commercial zeolites.²²² Depolymerisations were performed under sub-critical conditions at a mass ratio of water/nylon 30 : 1, 345 °C, 30 min and 30 wt% catalyst loading. Zeolite H β -25 exhibited higher activity and better recyclability than Al₂O₃ catalysts. Up to 90% conversion to soluble oligomers and 63% CPL yield was observed over Zeolite H β -25. Kinetic studies indicated consecutive oligomerisation and cyclisation steps, showing the catalyst to be effective in accelerating the hydrolysis step, compared to the un-catalysed process. The preferential formation of linear oligomers was attributed to the microporous structure of the zeolite.

Solvent-free processes were also reported for the conversion of nylons to CPL. Around 80% yield, with 99% pure CPL was obtained by contacting coloured waste and scrap PA6 textiles over 4 wt% solid NaOH at a high temperature of 320 °C required to melt NaOH and PA6.²²³ A number of by-products obtained by decolourization were identified in the vapour phase. An intricate depolymerisation mechanism was postulated involving hydrolysis with adventitious water, decarboxylation and hydrogen transfer steps. Differently, solid catalysts with lower basic strength, and based on γ -Al₂O₃ supported metal catalysts (Ni, Cu, Fe, Co), required much higher temperatures to achieve cyclodepolymerization.²²⁴ Thus, using stockings with diverse compositions (nylons, cotton and elastane) as a feedstock, a modest increase in CPL yield from 54% to 69%, without and with Cu@Al₂O₃ catalyst, respectively, was

obtained at 500 °C reaction temperature. Based on nylon content, the effective CPL yield was 95.3 wt%. Recently, solvent-free processes promoted by homogeneous lanthanide-based catalysts were reported. In the first example, commercially available tris-amido complexes of the general formula Ln(N(TMS)₂)₃ (Ln = lanthanide) afforded CPL in 90% yield and 95% purity at 240 °C reaction temperature, showing the catalytic activity to be higher for lanthanides with larger ionic radius.²²⁵ The process was adaptable to post-consumer nylon 6 and blends (1 : 1 wt) of nylon 6 with PE, PP and PET where the CPL yields were 93%, 91% and 77%, respectively, and the unreacted PE and PP were recovered at the end of the reaction. Kinetic and theoretical investigations suggested a mechanism involving a first deprotonation step of the nylon NH bond, followed by a chain-end backbiting step in which CPL units are sequentially removed from the chain ends. In a second example from the same group, the use of metallocene-lanthanide complex catalysts resulted in the formation of CPL in 81% yield, with a high TOF of 810 h^{−1}, under 240 °C and 0.2% catalyst loading.²²⁶ The catalysts can be reused without significant deactivation. The process required fine grinding and washing of polymers with 1 M KOH solution, careful drying, and the use of an oxygen-free environment under a 10^{−3} Torr vacuum. Under these conditions, actual feedstock (fishing nets, carpets, yarns, t-shirts, medical gloves) was converted in 65%–99% CPL yield.

Depolymerisation of nylon 6 commodities was also reported using acetic anhydride as a lytic agent and 4-dimethylaminopyridine (DMAP) as an organocatalyst (5% mol) (Fig. 12).²²⁷ In a typical reaction, a mixture of *N*-acetylcaprolactam (ACPL) and acyclic acetylimides by-products were obtained in around 60 : 40 ratio under microwave irradiation at 260 °C. Under these conditions, ACPL was obtained in 20%–78% yield by treatment of nylon 6-based stripes, threads, hammocks, and patches. Free CPL could be obtained by *trans*-acetylation of ACPL with an equimolar amount of ethanolamine at 80 °C.

In a different approach, nylon 6,6, and the PP mixture thereof, was effectively depolymerised through a transamidation reaction to give a mixture of amide dimers and monomers (Fig. 13).²²⁸ A 20% and 69% yield was obtained, respectively (together with a stoichiometric amount of adipamide), by contacting with a *ca.* 18-fold excess acetamide, under 200 °C, 3 bar NH₃ and Nb₂O₅ catalyst. Nb₂O₅ is a well-established and water-tolerant solid acid catalyst, which has been used in a variety of applications, including dehydration, esterification, and hydrolysis.²²⁹ Herein, a mechanism was proposed where the Lewis acid Nb₂O₅ activates the amide bond of Nylon, which then undergoes the nucleophilic attack of ammonia.

On the industrial scale, chemical recycling of nylon 6, or composites with glass, PE or polyurethanes from waste carpets and clothing textiles, is operated by Aquafil,^{230,231} *via* hydrolytic depolymerisation to caprolactam at 200–400 °C, 0.2–20 bar, using a soluble acid catalyst (Econyl process)^{232,233} (Table 3). Asahi Kasei claimed a chemical recycling process using microwaves to depolymerize polyamide 6,6 from waste





Fig. 13 Sketch of transamidation reaction of nylon 6,6 over heterogeneous Nb_2O_5 catalyst.

airbags and automobile parts into hexamethylenediamine and adipic acid monomers.²³⁴

3.3. Polyurethanes

Polyurethanes (PU) are an important class of specialty polymers with a vast array of applications and a global market volume expected to reach 28 Mton in 2025.²³⁵ PU refers to all those polymers that contain urethane linkages ($-\text{HN}-\text{COO}-$) in their backbone, regardless of the rest of the molecule. Typically, PU is synthesized by condensation of a di-isocyanate with a polyol, either a monomer or a polymer. Consequently, by varying the type of PU precursors, it is possible to tailor the properties of the final material to result in superior performances in terms of elasticity, durability, and insulation.²³⁶ PU is classified into two major categories: foams (soft and rigid) and CASE (coatings, adhesives, sealants, elastomers).²³⁷ PU elastomers are used in textiles because of their elasticity. The most common PU elastomers are commercialized with the trade name elastane (EL), Spandex or Lycra®, whose global production was 1.24 Mton in 2022.²³⁸ EL is composed of at least 85 wt% of segmented polyurethane elastomers and possesses high elasticity, showing elongations up to 100–600% of its original length.²³⁹ From a molecular viewpoint, EL is a polyurethane–polyurea block copolymer, resulting from the condensation of 4,4-methylenediisocyanate (MDI) and poly-tetrahydrofuran (polyTHF) as polymeric diol. In addition, EL often contains short diamines, mostly ethylenediamine, used as chain extenders. The latter is commonly obtained from oil, whereas polyols may be bio-sourced. For example, the Lycra company markets a 70 wt% bio-elastane based on corn starch.²⁴⁰ PU is increasingly used in the textile industry because of additional properties. It is worth mentioning that waterborne polyurethane is used as coatings to provide textiles with resistance to abrasion and waterproofing. PU coatings are also used on PET to produce synthetic leather and on PA to reinforce light-weight work wear.²⁴¹ EL may be used as an additive to enhance the stretchability and shaping behaviour of other textiles, such as cotton. Additionally, the current landscape is progressively enriched with complex yarns containing EL blended fibres. Therefore, garments rarely contain 100% EL yarn; instead, EL ranges between 1 and 10%, but it can reach up to 49%.²⁴² For instance, Alcantara® is an aesthetic and sensor-like leather material for interiors made of PET and PU fibres (*ca.* 70 : 30).²⁴³ Despite the current relatively low market (Fig. 4), EL is quickly diffusing in the textile sector, wherein it is expected to contribute with around 30% of all recovered materials by 2030.²⁴⁴ Unfortunately, when it comes to re-

cycling, the elastic properties provided by EL represent a major hurdle.²⁴⁵ Indeed, even small amounts of EL may hamper textile recycling. It has been estimated that almost 20% of the non-recyclable textiles in EU contain elastane.²⁴⁶ Therefore, the valorisation of EL plays a major role in facilitating garment recycling and, more generally, enhancing the circularity of the textile industry.

The valorisation of EL-containing blends is achieved by two distinct strategies, namely the separation of components by dissolution, possibly followed by depolymerisation (Fig. 14, path A) or the selective deconstruction of one fibre, while keeping the other component intact (Fig. 14, path B). Although not categorised as a chemical recycling method (see section 2), the dissolution-based methodology may remove EL, which is usually selectively solubilized. Typical solvents used for this aim are the REACH-restricted dimethyl formamide (DMF), dimethyl sulfoxide (DMSO), tetrahydrofuran (THF), or potentially green solvent as cyrene, tetrahydrofurfuryl alcohol, and γ -valerolactone.^{247,248} Moreover, the reusability of the recovered EL is not always effective due to fibre degradation upon extraction. Innovative solutions have been investigated for this approach, including the usage of deep eutectic solvents or ionic liquids.²⁴⁹

The second strategy exploits the suitability of a given depolymerisation process for a single (or multiple) component of the blended yarn, which then may undergo upcycling to reusable materials (Fig. 14, path B). For example, Zhang *et al.* claimed the extraction and depolymerisation of PET into terephthalic acid from a 95 : 5% PET : Spandex blend using a dichloromethane/ethanol solvent mixture and an excess of KOH at 30 °C for 30 min.²⁵⁰ Hummel and Sanchez *et al.* described an aminolysis-based technique to selectively degrade EL from cotton (*vide infra*). Although conceptually feasible, it must be noted that the deconstruction of yarns is particularly challenging due to the variety of blended textiles that would require specific processing. To date, catalytic systems enabling the selective cleavage of mixed EL fibres have not been adequately developed, as described in the following section.

PU comprises complex polymers that may contain different bonds in their backbone (*e.g.* urethane, esters, ethers, urea). Depolymerisation reactions of PU may affect any of these bonds. Herein, we will focus only on those processes resulting in the cleavage of the urethane chain linkage (Fig. 15). Indeed, most works performed with commercial and model polyurethanes that target urethane linkages could ideally be extended to EL chemistry. In this section, the generic chemistry of PU depolymerization will be considered. The most





Fig. 14 Schematic of elastane blended fibres and chemical recycling pathways: (A) separation by dissolution and (B) separation by selective deconstruction.

common depolymerisation strategy is based on a transcarbamoylation reaction, consisting of the transfer of a carbamoyl moiety (R-NH-CO) from a “donor”, usually PU, to an acceptor group. Based on the type of acceptor, the process can be referred to as alcoholysis (glycolysis), aminolysis or acidolysis (Fig. 15a–c).²⁵¹ In addition, hydrolysis and hydrogenolysis processes can be used to cleave the urethane bonds. Notably, these latter reactions are useful in “closing the loop” for PU production by providing reprocessable diamines and polyols.

Among these methods, glycolysis is the most used chemical recycling method for PU and the only one used in the industry, though applied to flexible foams (e.g. by EniChem,²⁵² Troy polymers,²⁵³ Rampf²⁵⁴), slabs, shoe soles waste, and not specifically designed for textiles, and often without catalysts.²⁵⁵ An exhaustive review of industrial glycolysis of polyurethanes can be found in the literature.²⁵⁶ In a typical PU glycolysis reaction, an excess of ethylene glycol is used in the

temperature range of 150–300 °C to provide low-molecular weight functionalised carbamates and a polyol.²⁵⁷ Recently, Xu and co-workers reported a two-step process for the glycolysis of a polyester-spandex blended fabric (94:6 ratio), where spandex is first dissolved in DMF at 75 °C, whereas insoluble PET is recovered to undergo conventional methanolysis depolymerisation (Fig. 10).²⁵⁸ Spandex is then depolymerised using an excess of EG and K_2CO_3 as a catalyst and 200 °C reaction temperature to give 4,4'-diaminodiphenylmethane (DMA) (10 wt% yield), 2-imidazolidinone and sticky polyTHF as main products. The drawbacks of the method are the use of DMF and the cost of product mixture separation, as usual for glycolysis processes.

Alcoholysis, e.g. using super-critical methanol,²⁵⁹ may be a friendlier option for PU depolymerisation. Skrydstrup's group reported a solvolysis process whereby elastane was simultaneously extracted and depolymerized from blended textiles



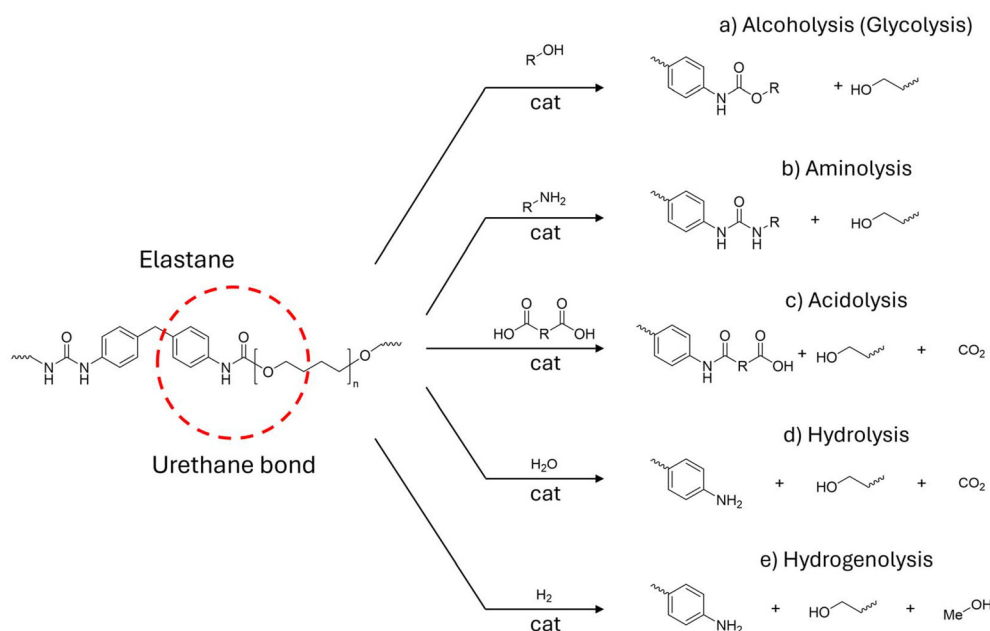


Fig. 15 Scheme of the major possible EL chemical depolymerization reactions involving the urethane bond using homogeneous-phase catalysts: (a) glycolysis, (b) aminolysis, (c) alcoholysis, (d) hydrolysis, and (e) hydrogenolysis.

using *tert*-amyl alcohol and KOH catalyst (0.19 wt%) at 225 °C for 4.5 h.^{260,261} The approach was highly versatile and adaptable to a wide variety of textiles, including pure elastane, 8–27 wt% elastane blends, and PU-coated textiles (PET/PU and PA/PU, gloves, jackets) (Table 4). Noteworthy, up to 98% overall mass recovery was possible with the tested fibres.

In another approach, primary and secondary amines or ammonia were used to perform the transcarbamoylation of PU-based materials *via* aminolysis using a series of aminic organocatalysts at 160 °C (Fig. 15b).²⁶² Compared to alcohols, amines are stronger nucleophiles, hence more effective in transcarbamoylation reactions, for instance at lower reaction temperatures. Sanchez and Hummel's group reported the aminolysis of EL from cotton-EL blended yarns using diethylamine and 1,4 diazobicyclo-non-5-ene catalyst in DMF or DMSO at 80 °C.²⁶³ Yet, aminolysis has been scarcely investigated in PU depolymerisation so far, likely due to hazards related to the use of amines.

Table 4 Solvolysis of elastane from mixed fabrics and PU coated fabrics^a

Garment type	Blends type	DMA yield (wt%)	Polyol yield (wt%)	Leftover solid (wt%)
Elastane fibres	EL 100%	13	74	n.a.
Blended tissue	Cotton-EL 14%	3.9	77	75
Synthetic leather	PET/EL coated	8.2	21	0.65
Post dissolution extract	EL 100%	12	72	n.a.
Workwear: glove	PA-EL coated	6.5	24	61
Synthetic leather jacket	PA-EL coated	9.0	30	61
Nylon tights	PA-EL 73%–27%	9.8	77	75

^a Isolated yield. Data from ref. 260.

In the acidolysis process, a dicarboxylic acid, such as maleic or fumaric acids, is used to cleave PU into an amide, a polyol and CO_2 (Fig. 15c).²⁶⁴ To date, acidolysis is claimed at a pilot scale by RAMPFT and Anace; however, process details are not available.^{265,266} Acidolysis is mostly used for the recovery of polyols, whereas amide moieties cannot be reprocessed easily. To the best of our knowledge, acidolysis has not yet been applied to EL or textiles but to PU foams.

PU hydrolysis was first developed by Ford Motor in the 70s.²⁶⁷ It consists of the cleavage of the urethane bond using hot water or steam to give diamines, polyols, and carbon dioxide (Fig. 15d).²⁶⁸ To be efficient, hydrolysis requires high temperatures (150–350 °C), high pressure (up to 50 atm), and the presence of an acid or base.²⁶⁹ Despite being the first PU depolymerization technique developed, hydrolysis has not yet been industrialized due to the costs associated with energy inputs and the biphasic separation of water and polyols.

All the above depolymerisation methods involve the management of considerable amounts of solvents or the usage of strong acids or bases, which results in poor control over product selectivity. Recently, the reductive catalytic cleavage of the urethane linkage, *i.e.* hydrogenolysis (Fig. 15d), has emerged as a promising strategy for the chemical upcycling of PU.⁷⁶ The process affords diamines and polyols alongside methanol. The approach has the double benefit of: (i) producing reprocessable chemicals, (ii) avoiding excesses of reactants and using H_2 as a clean reducing agent. PU hydrogenolysis typically involves homogeneous organo-metallic hydrogenation catalysts and a base co-catalyst. Kumar and co-workers reported the depolymerization of polyurethane resins using a Ru pincer complex at 135 °C, in the presence of 60 bar of H_2 and KO^tBu as a base.²⁷⁰ Similarly, Skrydstrup's group



described the reductive deconstruction of commercial end-of-life polyurethanes using the manganese complex $\text{Mn}^{\text{Ph}}\text{MACHO}$, isopropyl alcohol, KOH as a base, at 180 °C and 50 bar of H_2 .²⁷¹ DMA-based PU foams were depolymerised with a diamine yield of up to 90 mol%. Schaub *et al.* reported the hydrogenation of MDI-based polyurethane using a Ru complex and KO^tBu (4 mmol%) as catalysts in THF at 120 °C, under 50 bar of H_2 , to yield 76% MDI and 85% 1,6-hexane diol.²⁷² In these studies, the solubility of the polymer is indicated as a key to enabling the optimal conversion, whilst a synergistic effect of solvolysis–hydrogenolysis has been highlighted for efficient depolymerisation.

3.4. Polyacrylics

Unlike polyesters and polyamides, polyacrylates are thermosetting materials that cannot reshaped by melting. Acrylics find applications in various fields, including textiles. acrylic fibres, particularly polyacrylonitrile (PAN) and modacrylic (MAC), are mostly used in home furnishings, *e.g.* blankets, and in wool-like clothing, *e.g.* sweaters and socks, due to their favourable strength, density, softness, heat retention and flexibility properties.²⁷³ PAN is a homopolymer of acrylonitrile, whereas MAC is a more complex co-polymer of PAN with vinyl chloride or vinylidene chloride monomers (Fig. 16).²⁷⁴ Due to the strong C–C bonds, these polymers are particularly resistant to depolymerisation and biological degradation.²⁷⁵ Polyacrylics are produced in around 2 Mton per year, and less than 10% is recycled.²⁷⁶ In most cases, polyacrylate fabrics are not composed of pure acrylic fibres, but rather mixtures with other materials, such as polyesters, polyamides, cotton and wool, and are usually pigmented.

For the above-mentioned reasons, the mechanical recycling of acrylic-based textiles is unfavourable and expensive.²⁷⁷ In addition, acrylates are quite insoluble polymers (*e.g.* in DMF, dimethylacetamide, nitric acid, ethylene carbonate), which makes their recycling by dissolution/precipitation (with water) industrially unattractive. Depolymerisation is also troublesome. Thermal processes may lead to the formation of carbonaceous solid waste, as well as toxic gases (*e.g.* HCN, NH_3), whilst controlled depolymerisation processes are still at the very early development stage. Since PAN is the main precursor of carbon fibres, some studies have focused on the thermal upcycling of waste acrylic textiles for this aim.²⁷⁸ Dissolution in organic solvents still remains the most pursued recycling route to date.²⁷⁹

No studies have been reported on the catalytic depolymerisation of PAN and MAC waste. Recent findings are limited to

the treatment of the parent poly(methyl methacrylate) (PMMA), commonly used as a glass substitute (Plexiglas). Thus, depolymerization of PMMA was achieved at 290 °C, with >90% yield of methyl methacrylate (MMA), *via* reversible-deactivation radical polymerization by the incorporation of thermolytically labile end-groups (*N*-hydroxyphthalimide esters and trithiocarbonates), under solvent-free conditions.²⁸⁰ Depolymerization of PMMA and poly(*n*-butyl methacrylate) (PBMA), with terminal chlorine chain-end functionality, was reported over iron chloride at 170 °C, using tetra(ethylene glycol) dimethyl ether as solvent.²⁸¹ A conversion of >70% was achieved, where the monomer could be recovered by distillation.

Alternative to depolymerisation, upcycling of waste acrylics has been possible by chemical functionalisation. For instance, recycled PAN fibres from textiles were amino-functionalized by means of ethylenediamine and used for the adsorption of Hg (II) ions from contaminated water media, with 99% removal efficiency and an adsorption capacity of 1116 mg g^{-1} .²⁸²

4. Conclusions and prospects

Economic indicators suggest the synthetic textile market will considerably grow in the coming years, mostly as a consequence of the fast-fashion phenomenon and the ease of synthetic fibre manufacturing. In addition to the high turnover rate of clothes in stores, unsold and used disposable garments are also rapidly destroyed, mainly in pulping mills or incinerators. Fundamentally hydrocarbon-based and non-biodegradable synthetic yarns may also be low-quality, often containing significant amounts of toxic substances (DMF, heavy metals, phthalates, aromatic polycyclics), even exceeding the REACH limits.^{283,284} Therefore, fashion is considered the most polluting industry in the world after oil. This mainly affects developing countries (*e.g.* Ghana, Nigeria, Kenya, Tanzania, Chile), where about 70 tonnes of waste garments are estimated to be daily transferred in illegal dumps or ditches from the second-hand cloth markets of Accra.^{285,286} Strong commitments have been undertaken by policymakers, both at the European and global level, to increase sustainability and circularity of the textile sector by adopting strategies to improve recycling while minimising disposable fashion and making textile products more repairable, reusable, recyclable and free of hazardous substances.^{287,288} Current limitations are due to the lack of circular textile supply chains, which are mostly structured around recovery, selection and mechanical recycling.

Complementary to mechanical recycling, depolymerisation *via* chemical recycling is considered an industrially attractive option. Treatment of plastic waste to recover monomers, which are reprocessable into virgin yarns, other plastic goods or valuable chemicals, may help diminish the depletion of non-renewable oil feedstock, the economic and environmental impact of synthetic polymer production, and the accumulation of non-biodegradable materials in landfills. The strategy may



Fig. 16 Structures of PAN and MAC. X = H or Cl, *m* 15–65%.



ensure the supply of future raw materials for bottle-to-textile and textile-to-textile circular chains or the development of innovative value chains, which may expand the concept of plastic refinery. Drawbacks of depolymerisation include the massive use of solvents or chemicals, soluble promoters, corrosive conditions and the need for high reaction temperatures. Using a catalyst may significantly improve the processes by decreasing the energy inputs and reaction times while enabling selective depolymerisation to be driven into a variety of useful monomers under mild conditions. However, unlike other plastic materials, the complexity of synthetic textile waste poses further problems. These waste streams are intricate mixtures of additives, dyes, hard parts and different fibre-type blends. In addition to precise upstream sorting and separation, this would require demanding pre-treatments, such as grinding, dissolution or selective disintegration. Specialised catalysts with enhanced resistance and versatility may also be needed.¹⁰⁴

Depolymerisation of polyester, polyamide, polyurethane and polyacrylic textiles illustrated in the previous sections show that despite the growing interest in the catalytic chemical recycling of these polymer types, few studies directly focus on textiles, whereas the majority relate to pure polymers or model compounds. Depolymerization of PET fibres (section 3.1) is surely the most advanced, with catalytic depolymerization processes usually involving solvolysis. However, catalyst selectivity and durability are still obstacles to the handling of post-consumer products. Similarly, catalytic depolymerisation of PA has been mostly proven at the laboratory scale using homogeneous and heterogeneous catalysts (section 3.2). By contrast, PU catalytic depolymerization is still in its infancy, especially targeting elastane. Model polyurethanes are usually employed, and very few works focus on elastane (section 3.3). In addition, EL research suffers from a lack of standards and the high complexity and diversity of commercial EL on the market. Therefore, joint efforts between researchers and EL manufacturers are needed to standardize R&D benchmarks. Finally, depolymerisation of polyacrylics such as PAN and MAC has seldom been investigated (section 3.4).

All the above considerations explain why the few existing chemical recycling processes for synthetic textiles are currently demonstrated mostly on a small scale, whereas their implementation on a large scale has not yet been developed. To this aim, these technologies should be radically improved. Some challenges and priorities in this direction may be highlighted.

Innovative, preferably heterogeneous, catalysts must be designed, whose structure, morphology and composition may dramatically affect the fate of depolymerisation.⁸³ Particularly, granulometry and porosity of solid catalysts may control the surface interaction with (insoluble) synthetic polymers and diffusion within the catalyst body. Catalysts with nanometric size and macroporosity may be the preferred choice. Heterogeneous catalysts may show improved thermal and chemical resistance under the usual reaction conditions of solvolytic depolymerisation, and they are preferred by the industry due to the easier recovery and implementation into the existing reactor equipment. Further, bifunctional catalysts may

enable complex reaction sequences to efficiently occur in cascade and in one-pot, thus allowing otherwise inaccessible reaction pathways and products.⁹⁹ This is, for instance, the case of solvolysis-hydrogenation paths.^{289,290} Coupling heterogeneous catalysis with continuous flow reactor engineering can add substantial process intensification, which, to date, in the field of plastic material waste is limited to unselective cracking.²⁹¹ The use of earth-abundant or low-cost catalytic precursors should also be considered for the establishment of depolymerisation processes at large.

Given the complexity of (depolymerisation) treatments and downstream purifications inherent to the chemical recycling of synthetic textiles, careful technical, economic and environmental analyses shall be performed to assess the sustainability, feasibility and competitiveness of the overall processes.^{292,293} This may include, for example, either (Environmental) Life Cycle Assessment, Life Cycle Sustainability Assessment (ELCA, LCA and LCSA),^{294,295} or Life Cycle Cost (LCC) analyses.^{296,297}

Particularly for research in Chemical Sciences, together with an accurate catalyst and process design, upstream material design is desirable. This may include recyclable-by-design, for instance, in terms of separability, biodegradability or ease of chemical depolymerisation,^{72,298} and safe-by-design, for instance, fibres devoid of noxious monomeric components, additives or contaminants.^{299,300}

Traceability technologies and labelling and information declaration for textiles shall be improved and implemented. These may include textile fibre name, origin, composition, use of recycled materials, sorting and recycling options.³⁰¹

Finally, consumer awareness regarding textile usage, disposal, reuse and recycling should be increased. Consumers may not be aware of the synthetic content of their clothes, nor their fate or negative impact on the environment when discarded. Separation, collection and recycling options for textiles, particularly synthetic ones, should be made known to the public at large. Recycled garments, second-hand clothes or textiles made of recycled or secondary raw materials often suffer from negative perceptions, which may include safety and low-quality prejudices. Market acceptance of recycled materials may be improved by education and the commitment of brands to sustainability.

In conclusion, advancements in textile chemical recycling require a collective effort from all stakeholders across the value chain, including researchers, policymakers, consumers, textile producers, and recycling companies. To this respect, innovation in catalytic chemical recycling has the clear potential to provide new pathways toward a closed-loop circular economy for textiles, hence reinforcing the overall objectives of the European Green Deal³⁰² and supporting the achievement of the Ecological Transition and Sustainable Development Goals.^{303,304}

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.



Conflicts of interest

There are no conflicts to declare.

Acknowledgements

Thanks are due to the Circular and Sustainable Made in Italy Extended Partnership (MICS) funded by the European Union Next-Generation EU. (Piano Nazionale di Ripresa e Resilienza (PNRR) – Missione 4, Componente 2, Investimento 1.3 – D.D. 1551.11-10-2022, PE00000004, CUP B53C22004100001) for financial support. Thanks are also due to the LABSolve “Lactic acid based new green solvents: design, synthesis and use in catalysis” project (20229P7PPM, CUP B53D23015230006) funded by the European Union Next-Generation EU, PNRR, Missione 4 “Istruzione e Ricerca” – Componente C2 Investimento 1.1, “Fondo per il Programma Nazionale di Ricerca e Progetti di Rilevante Interesse Nazionale (PRIN)”.

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