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Unlocking the potential of plastic recycling processes with the integration of membrane technology: a focus on PET valorisation

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Plastics have revolutionised modern society; however, they have also caused environmental damage *via* waste accumulation. To balance their advantages with the negative impact on the environment, it is necessary to enhance recycling procedures, since the existing techniques are ineffective, require significant energy consumption, and yield recycled items of poor quality. One promising pathway is to utilise membranes. While membrane technology has been extensively studied in wastewater treatment and organic solvent recovery, its direct application to plastic recycling, particularly using real plastic feedstocks like PET, remains underexplored. This paper aims to address the gap by exploring different plastic recycling technologies, followed by an analysis of the current state of the art and then the prospective integration of membrane technology into plastic recycling processes with an emphasis on PET as a case study. This study provides a forward-looking discussion on some exciting opportunities including using conventional membrane processes and emerging membrane processes. These include separating impurities and additives from the recycling stream, recovery of valuable catalysts, and treating wastewater produced during PET recycling processes. This study also investigates emerging membrane research in PET recycling *via* use of mixed matrix membranes (MMMs) and implementation of hybrid membrane systems (HMSs). This study also aims to identify current challenges and future opportunities for membrane technology implementation in the field of plastic recycling, considering its impact on recycled product quality, energy consumption, and economic feasibility. Membrane technology is demonstrated to have the potential to transform the plastic recycling industry, and contribute to a more sustainable and circular economy.

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Green foundation

1. **Advances in Green Chemistry:** The perspective advances green chemistry by integrating membrane technology into plastic recycling, achieving precise molecular separations that reduce energy consumption by up to 90% compared with traditional methods. This technology enables the production of high-purity, food-grade recyclates from waste plastics, promoting a circular economy and significantly lowering the carbon footprint of the plastics industry.
2. **Significance of the Study:** With only 9% of the nearly 460 million tonnes of plastic produced annually being recycled, the study addresses a critical environmental issue. It provides an interdisciplinary solution that combines materials science and chemical engineering to enhance recycling efficiency, aligning with global sustainability goals and legislative targets for increased recycled content in products.
3. **Future Directions:** The field is set to advance with innovations in membrane materials and systems, such as mixed matrix membranes, to process diverse plastic types and scale up for industrial applications. The review identifies key research opportunities and challenges, guiding efforts to develop more efficient and sustainable recycling technologies, thereby shaping the future of green chemistry science.

Introduction

The prevalence of plastics in society has been ever-increasing in the modern world.[1–3] Polymer constituents are generally low cost and easy to process, enabling plastic use in a variety of industries.[4] Plastic production rose to around 460 million tonnes (Mt) in 2019 (Fig. 1).[5] However, most of these materials are produced from petrochemical sources,[3] and their

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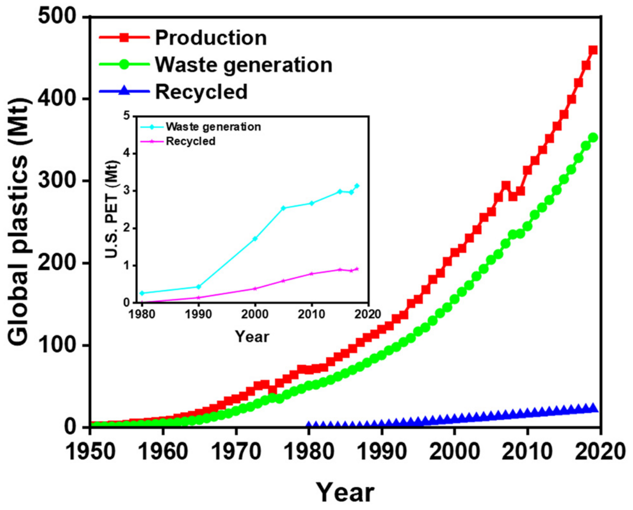

Fig. 1 Global plastics production, waste generation, and recycling, alongside U.S. PET waste generation and recycling.[5,26,35]

durability has led to environmental challenges related to their disposal.[4] Plastic waste is a significant environmental concern because it is non-biodegradable (taking ~450 years to degrade[3]) and detrimental to ecosystems and living organisms.[1,6]

Plastic waste is a diverse category that includes an extensive range of polymers. A few notable examples are polyethylene terephthalate (PET), low-density polyethylene (LDPE), high-density polyethylene (HDPE), polypropylene (PP), polystyrene (PS), polyvinyl chloride (PVC), polylactic acid (PLA), polycarbonate (PC), acrylonitrile butadiene styrene (ABS), thermoplastic/thermoset polyurethane (PU), and nylon. However, this review focuses on PET as a key case study due to its significant contribution to recycling efforts and its well-documented valorisation routes. PET is a model polymer that, despite accounting for approximately 8% of global plastics production in 2019,[1] represents a remarkable 55% of all recycled plastics globally in 2017.[7] This contrast highlights the significant role PET plays in the recycling industry and makes it an ideal candidate for exploring recycling processes and strategies. Knowledge gained from its recycling processes can be used to inform membrane integration strategies for other plastics. PET possesses unique and well-characterised properties. It is colourless and can be transparent or translucent, depending on crystallite size and content, allowing for visibility of the products it contains while possessing high strength and durability, making it impact resistant.[8–10] PET is chemically resistant and inert, so it can withstand a wide range of chemicals and does not react with other substances.[11] PET possesses a high melting point (260 °C) which makes it a suitable choice for applications involving elevated temperatures,[12] and it is also appropriate for use where moisture resistance is crucial, as its minimal water absorption and excellent barrier properties limit permeation of gases and liquids.[13] Furthermore, PET is normally considered nontoxic and typically does not require plasticisers. As a result, it is unlikely to release potentially dangerous substances into the environment,[1,11] and is an excellent candidate for recycling.[6,14]

PET is a highly versatile material used in numerous industries owing to its distinctive properties. PET is widespread in the packaging industry, specifically the packaging of foods, drinks, and cosmetic products,[6,11] and it is also used to manufacture textile fibres and fabrics.[15,16] PET is a key material in the medical industry, having applications in surgical implants, medical equipment, and drug delivery systems.[17–20] In the electronics industry, components such as circuit boards and electrical housings are manufactured from PET,[21,22] and the automotive industry utilises the material in vehicle bumpers and dashboards.[23] Within the construction

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Laila Halim graduated with a Bachelor of Engineering (Hons) and Bachelor of Science from Monash University in 2020. She is currently undertaking a PhD within the department of Chemical and Biological Engineering at Monash University, working to utilise membrane technology for plastic recycling applications (under the supervision of Prof. Matthew Hill and Dr Leonie van 't Hag). Her key research interests reside within the sustainable engineering field including plastic pollution reduction, the circular economy, membrane-based separations, and green energy technologies.

industry, PET is employed to fabricate insulation, roofing sheets, and pipelines.[24,25] The engineering industry also employs PET in the production of gears, bearings, and electrical insulators.[23]

In 2018, only 29.1% of PET waste was recycled in the United States (Fig. 1),[26] leaving the overwhelming majority in landfills or oceans, according to the Environmental Protection Agency (EPA).[27] In addition to addressing the impact of plastic pollution on the marine ecosystem, developing a more circular carbon economy through recycling is crucial to mitigate the excessive greenhouse gas emissions associated with the production of plastics from petrochemical sources.[28] Despite the various PET recycling routes that have been developed in academia and that have penetrated industry, the use of PET plastic material remains overwhelmingly single-use across the globe. PET recycling techniques have obstacles, including the low quality and properties of recycled materials due to impurities and contaminants, high energy consumption, low efficiency, high cost, and waste of resources.[29–33] Moreover, the recycling rate of PET significantly deviates between countries due to the limited financial benefit it yields. Investigation of effective and economically viable methods of recycling PET is of crucial significance, as the production of PET from waste PET is currently not as profitable as PET sourced from fossil fuels, and only a handful of developing countries are willing to undertake this task.[1] Therefore, new recycling techniques or approaches for enhancing current techniques are needed to reduce plastic waste's negative effects on the ecosystem,[3] and harmonise production, consumption, and disposal.[34] One of the most promising approaches is membrane technology, which may play an increasingly important part in PET recycling methods (as well as the recycling of other plastics), as the demand for sustainable and eco-friendly recycling methods rises.

Membrane application perspectives

The emergence of membrane technology may ultimately spark a revolutionary transformation in the field of plastic recycling due to its ability to selectively separate and purify various components based on their size and molecular weight.[36,37] Moreover, the continuous progress in economic membrane preparation methods and the wide diversity of available membranes present considerable possibilities for the emergence of different membrane applications to improve plastic recycling processes.[38] Importantly, the technology could contribute to enhanced sustainability by reducing energy and water consumption.[39,40] One notable advantage of membrane technology lies in its easy integration potential within existing industrial processes. This feature not only makes it a scalable solution but also significantly contributes to its cost-effectiveness.[41] Considering these factors, membrane technology offers a broad range of advantages, including the ability to produce high-quality recycled products, the reduction of reliance on virgin materials, the improvement of plastic processing efficiency through selective separation capabilities such as the removal of impurities and contaminants, and the facilitation of recovery of valuable components from plastic waste. Therefore, the integration of membrane technology is a step towards achieving highly efficient and economically viable plastic recycling processes.[42] Moreover, membrane performance can be further advanced through various strategies such as utilising mixed matrix membranes (MMMs) and employing hybrid membrane systems (HMSs).[43] These potential uses of membrane technology have significant implications for future research in the field of plastic recycling. Nevertheless, the successful implementation of membrane technology first requires an in-depth understanding of valorisation routes. PET valorisation routes produce complex streams necessitating precise separations to optimise resource

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Xiaoheng Jin

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recovery and reduce waste. Membrane technologies inherently enhance these routes by providing modular, scalable solutions for contaminant removal, monomer purification, and by-product valorisation. For example, in some recycling processes, membranes could recover depolymerisation products while treating effluents, therefore decreasing energy requirements relative to conventional approaches. This synergy not only improves product quality but also fosters a circular economy by facilitating closed-loop processes. Thus, due to the technology's novelty in this domain and high potential impact, it is important to conduct a comprehensive study and analysis to determine the different potential aspects of membrane applications in PET recycling, thereby also guaranteeing the technology's utmost efficiency and environmental benefits.

Therefore, this paper aims to contribute to the ongoing discourse on improving plastic recycling by exploring the prospective integration of membrane technology into existing processes. Rather than providing a detailed review of past limited research, the focus has been directed towards offering a forward-looking perspective on how membranes can be applied to overcome existing challenges, such as separating impurities, reducing energy consumption, and improving the economic viability of recycling.

History of PET valorisation routes

The history of PET recycling demonstrates how society as a whole has become more aware of environmental concerns and the growing demand for more efficient and environmentally friendly PET recycling techniques. The history of PET recycling can be classified into distinct stages (Fig. 2). In the 1950s and 1960s, various methods of PET recycling were introduced such as methanolysis, hydrolysis, and glycolysis. Despite this, the PET bottle was only recycled beginning in the 1970s. Then, following earlier favourable outcomes, the implementation of recycling methods for recycling PET bottles was commercialised.[44] In the 1980s, the Resin Identification Code system, which comprises a triangular structure, was developed to assist with the identification and classification of plastic waste.[1] In the early 1990s, a newly developed recycling method called "bottle-to-bottle" arose, which made it possible to recycle PET bottles into fresh bottles. This breakthrough was a huge step forward for PET recycling, as it allowed for a more effective and sustainably oriented approach for recycling PET bottles. The United States provided an early stimulus for the expansion of the recycling industry by authorising the use of recycled PET in food-contact applications.[13] In the late 1990s and early 2000s, there was a notable upsurge in PET recycling, which was attributed partly to the development of the bottle-to-bottle recycling method and partly to the establishment of recycling programs and regulations in numerous countries. As a result, companies were beginning to give precedence to the implementation of recycled PET in their products. Moreover, these companies directed their efforts toward the development of novel technologies to enhance the efficacy and quality of PET recycling. They successfully implemented automated sorting systems, advanced washing systems, and introduced new catalysts like ionic liquids (ILs).[45,46]

In the 2010s, technological developments, such as the implementation of new technologies that remove impurities and contaminants from recycled PET, increased both the efficiency and quality of the PET recycling process. On the other hand, a rise in demand for recycled PET has been observed concurrently with the expansion of the packaging

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Leonie van 't Hag

Dr Leonie van 't Hag obtained her MSc degree in Physical Chemistry from Wageningen University & Research (NL) and PhD from the Department of Chemical Engineering at The University of Melbourne (AU) in 2017. Leonie was a Postdoctoral Research Fellow at ETH Zurich (CH) for three years before starting as a Lecturer at Monash University in late 2019. The Hybrid Assembly Group focuses on the sustainable production and processing of soft materials. We do this by investigating the structure–property relationships for food, packaging and biomedical applications using advanced scattering and microscopy techniques. Dr van 't Hag is now a tenured Senior Lecturer (equivalent to early Associate Professor) and her work is placed at the interface of fundamental science and industrial applications that contribute to achieving sustainable development goals.

Zongli Xie

Prof. Zongli Xie is a Research Group Leader and Principal Research Scientist at CSIRO Manufacturing, Australia. Her research is in advancing membrane/catalysis materials and emerging technologies for water, environment and energy. She has over 160 journal publications, 5 patents and >30 confidential commercial technical reports. She is also the Vice President of the Membrane Society of Australasia (MSA), an Adjunct Professor in Victoria University, and an associate editor of Separation Science and Technology, Desalination and Water Treatment, and Results in Engineering. She was elected as an ATSE fellow in 2024.

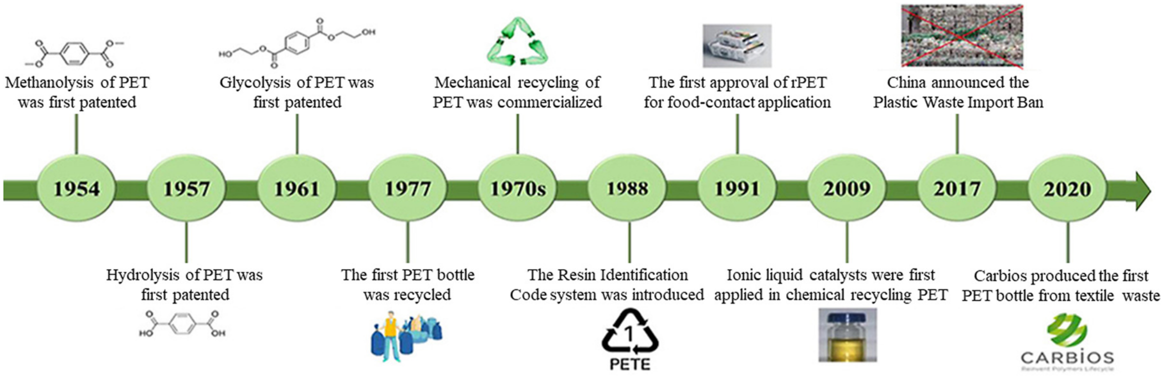

Fig. 2 The development history of PET and PET recycling (reproduced with permission.[1] Copyright 2018, Elsevier).

industry, prompting companies to incorporate greater quantities of recycled material into their products. Therefore, the percentage of PET materials being recycled has been rising over time.[1] A sustained trend of increasing PET recycling was observed up to 2017, but this was followed by a decrease in recycled PET amount as a result of changes in the regulations of key importing countries, which restricted importation of a significant portion of plastic waste. As a result, many of the world's leading PET plastic exporters have enhanced their own PET recycling infrastructure.[47]

Today, an increasing emphasis on sustainability and the circular economy is still promoting the growth of PET recycling, with the progression of technologies such as chemical recycling as well as the establishment of new markets for recycled PET.[48] The acceptance of PET-recycled products has long been established, and many countries and companies are actively advocating and engaging in recycling initiatives. For example, in 2020, Carbios produced the first PET bottle made entirely from textile waste with the same properties as conventional bottles, thanks to PET enzymatic depolymerisation. There is an increasing industry movement towards incorporating more recycled PET, such as into product packaging in the coming years.[1]

Matthew R. Hill

Prof. Matthew R. Hill received his B.Sc. in 2002 and completed his PhD in Chemistry in 2006 at the University of New South Wales. Following this, he was a postdoctoral researcher at CSIRO from 2006 to 2009. Transitioning to the role of research scientist at CSIRO from 2009 to 2011, he simultaneously held positions as an adjunct lecturer at the University of Melbourne starting in 2008. Additionally, he undertook roles as a visiting scientist at the University of California, Berkeley, in 2008, and later at the University of Colorado, Boulder, in 2011. In 2011, he joined the University of Adelaide as an adjunct lecturer. During 2011–2014, he served as a senior research scientist at CSIRO, advancing to the position of principal research scientist from 2014 to 2019. From 2019 to 2024 he held the position of senior principal research scientist at CSIRO. Concurrently, he has been associated with Monash University from 2016 to 2014, serving as deputy head in the Department of Chemical and Biological Engineering. Then, in 2024 he joined the Department of Materials Science and Engineering where he is serving as a head of Department.

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Prof. Benny D. Freeman received his B.S. in Chemical Engineering from NC State University in 1983 and his PhD from the University of California, Berkeley, in 1988. He was a NATO Postdoctoral Fellow at the Ecole Supérieure de Physique et de Chimie Industrielles of la Ville de Paris' Laboratoire Physico-Chimie Structurale et Macromoléculaire from 1988 to 1989. Afterwards, he taught chemical engineering at NC State from 1989 until 2001. Since then he has served as the William J. (Bill) Murray Jr. Endowed Chair in Engineering in the chemical engineering department at The University of Texas in Austin. He is also a Professorial Fellow at Monash University.

Development of PET valorisation routes

Valorisation is the process of turning waste into more useful materials or products. There are four distinct routes of plastic valorisation, as defined by ASTM D5033:

- primary recycling, which involves the mechanical reprocessing of waste materials into products with equivalent properties;
- secondary recycling, which involves the mechanical reprocessing of waste materials into products requiring lower properties;
- tertiary recycling, which involves the recovery of valuable chemical constituents such as monomers or additives;
- quaternary recycling, which involves the recovery of energy.[2,31,49,50]

The various routes differ in terms of their process and relative merits, depending on the feedstock and requisite process conditions (Fig. 3 and 4). The development of all valorisation routes has been promoted by the intensifying requirement for sustainable materials and the necessity to decrease waste.[51] Therefore, it is vital to possess awareness regarding PET valorisation routes to drive further innovation in the recycling industry.

Primary recycling

The primary recycling route refers to the physical degradation processes of converting waste plastic into a product that retains the same properties as the original product.[2,31] The key distinguishing feature of this recycling route lies in its emphasis on pre-consumer feedstock. This feedstock primarily comprises clean and uncontaminated pure polymers obtained from industrial by-products or discarded materials.[6,54–56] While primary recycling is often known as "closed-loop recycling",[31,53] it is technically feasible for other recycling routes to also establish a closed loop. Primary recycling is typically carried out through a process known as re-extrusion.[31,53]

Re-extrusion is a frequently employed process in plastic manufacturing facilities to directly recover waste. As shown in Fig. 5, the process involves shredding pure polymer scrap into flakes, followed by melting and reprocessing of these flakes, frequently accompanied by addition of virgin plastic to improve recycled plastic properties. However, there is degradation in product quality upon repeated extrusion of the same materials due to minor amounts of chemical breakdown during extrusion. Because of the degradation during re-extrusion, a depolymerisation process, which would avoid such degradation, may have some advantages over re-extrusion.[6,53]

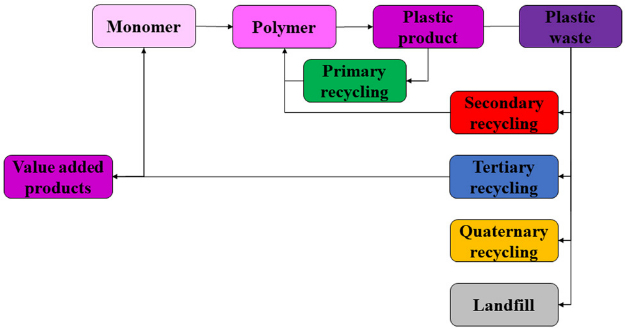

Fig. 3 PET valorisation routes, associated plastic quality, and their position within the processing line (adapted under terms of the CC-BY license.[52] Copyright 2013, Scientific Research Publishing Inc.).

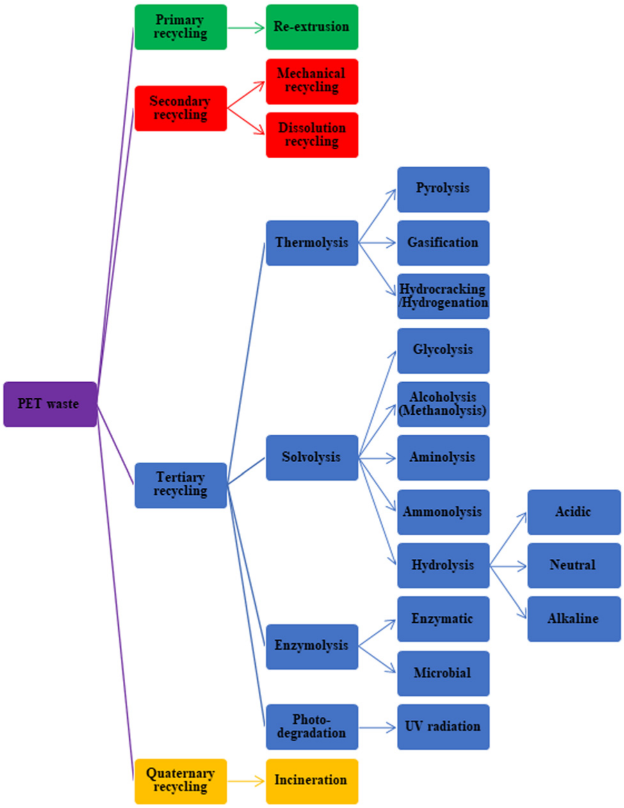

Fig. 4 PET valorisation routes (adapted under terms of the CC-BY license.[53] Copyright 2022, MDPI).

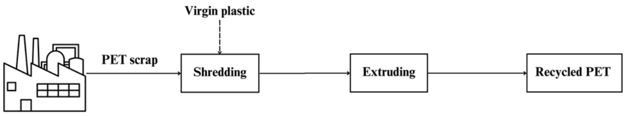

Fig. 5 Proposed schematic of the re-extrusion recycling process (based on provided information in ref. 6).

Secondary recycling

Secondary recycling is similar to primary recycling whereby PET stays in polymeric form throughout the process. However, the physical degradation processes are performed on post-consumer waste.[55] Moreover, the secondary recycling route typically leads to a final product with diminished properties compared with the initial product.[2,53] Consequently, this category of recycling is commonly referred to as "downgrading".[53] Secondary recycling does not involve altering the basic polymer chain chemistry,[54] thereby rendering it relatively

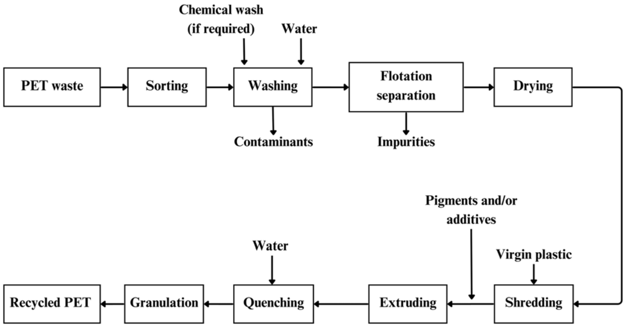

Fig. 6 Schematic of the mechanical recycling process (adapted with permission.[63] Copyright 2009, Elsevier).

energy-efficient and environmentally friendly. Two processes are frequently employed for secondary recycling: mechanical recycling and dissolution recycling.[57]

Mechanical

The mechanical recycling process is widely used in recycling moderately contaminated waste PET, resulting in the production of recycled products.[1–3,58] Fig. 6 depicts the mechanical recycling process. Despite maintaining the fundamental polymer chain chemistry, each cycle results in property deterioration due to a decrease in molecular weight resulting from chain scission.[59,60] In addition, the degradation is influenced by various chemical reactions such as hydrolysis, esterification, *trans*-esterification, hydroxylation, polyaddition, and thermo-oxidative reactions;[11] the presence of impurities further intensifies degradation.[11,55,61] Therefore, pre-treatment washing and drying are essential due to the presence of contaminants in post-consumer waste. Washing removes physical impurities, whereas instances involving substances such as glue require chemical washes, frequently involving the use of NaOH.[3,62,63] Often, highly contaminated items such as take-out containers are rejected from the process.[1] Moreover, PET flakes are subjected to melting and reprocessing to produce recycled PET pellets, typically through extrusion or injection moulding techniques,[6,11] with the potential addition of pigments and additives.[63]

Dissolution

Dissolution recycling is a less commonly employed process, but it results in a recycled product comparable in quality to virgin PET.[64,65] Fig. 7 presents a schematic representation of the dissolution recycling process. The dissolving process involves use of heat and a solvent (such as *N*-methyl-2-pyrrolidone (NMP) or benzyl alcohol)[66] to selectively dissolve PET waste flakes, resulting in the formation of a solution consisting of the original polymers and additives.[67,68] Following dissolution, additives and insoluble contaminants are separated *via* filtration.[3,65,67] The target PET polymer can be obtained by reprecipitation, achieved by introducing non-solvents/anti-solvents.[3,66] Afterwards, the polymer undergoes a series of filtration, washing, and drying steps,[66] ultimately leading to its final form as a powder or small grains appropriate for use in traditional plastic manufacturing processes.[65] To promote sustainability, the recovery of the solvent or non-solvent can be achieved by evaporative techniques.[69]

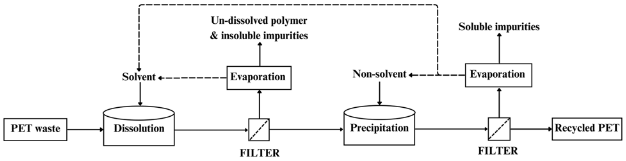

Fig. 7 Schematic of the dissolution recycling process (adapted under terms of the CC-BY license.[69] Copyright 2021, MDPI).

Tertiary recycling

Tertiary recycling routes can be employed to recycle waste which is highly contaminated, or composite or heterogeneous in nature, such that it is not suitable for primary or secondary recycling.[55] Tertiary recycling takes advantage of a range of chemical, biological, and radiation-based processes to break down plastic waste into its basic chemical constituents: the monomers.[54] On the other hand, the quality of the recycled product deteriorates when the same materials are extruded again through primary or secondary routes. The degradation is a result of the compromise of both the physical and chemical properties, mostly caused by contaminants. Furthermore, there is a slight occurrence of chemical degradation during extrusion. Therefore, the process of depolymerising polymers, purifying the individual monomers, and reassembling the material from its fundamental constituents could provide a significant benefit in tackling these problems.[70,71] Tertiary recycling processes involve several steps including pre-treatment, degradation of the original polymeric chain, and purification,[72] resulting in the formation of predominantly monomers, dimers, and oligomers that can be employed to produce valuable commodities, such as high-quality recycled plastics.[6,29,54,61]

Thermolysis

Thermolysis is an approach for plastic recycling that utilises thermal energy to disintegrate polymer chains into smaller gaseous and liquid constituents that can be repolymerised or used as fuels.[52] Thermolysis exhibits exceptional adaptability in managing a diverse array of (mixed) waste streams, and it leads to degradation and elimination of impurities and colourants. This approach involves a range of processes, including pyrolysis, gasification, and hydrocracking/hydrogenation.[55]

Pyrolysis

Pyrolysis refers to the thermal degradation of PET in an environment free of oxygen.[73,74] Fig. 8 depicts a schematic representation of the pyrolysis recycling process. The process is often conducted in fixed or fluidised bed reactors at 300 to 900 °C, using catalysts such as zeolites and silica–alumina.[73–78] This process results in hydrocarbon-rich liquid/

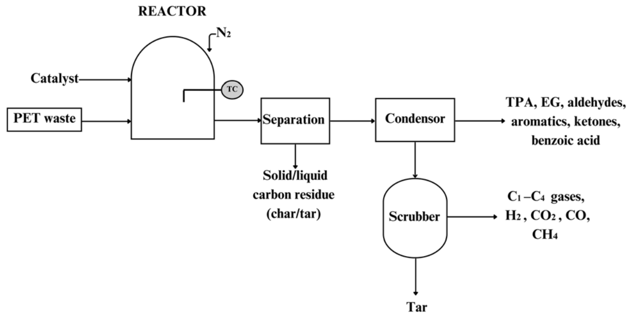

Fig. 8 Schematic of the pyrolysis recycling process (adapted under terms of the CC-BY license.[80] Copyright 2021, MDPI and CC-BY license.[83] Copyright 2022, Springer Nature).

wax mixtures as pyrolysis oil, which are commonly used to manufacture fuel or produce high-value chemicals.[55,73] With PET as the feedstock, the liquid/wax fraction primarily comprises terephthalic acid (TPA) and its derivatives, aldehydes, benzoic acid (BA) and its derivatives, aromatics and other oxygen-containing compounds such as ethylene glycol (EG) and ketones.[79–81] Fig. 9 depicts a primary thermal cleavage mechanism where PET becomes TPA. The process also generates solid carbonaceous residue and a gas fraction comprising C_1–C_4, methane (CH_4), hydrogen (H_2),[79] carbon monoxide (CO) and carbon dioxide (CO_2) (due to PET being oxygen-containing).[74,75,80] The presence of BA in the recycling stream presents challenges, including obstructing the piping system and having a detrimental effect on product oil fuel efficiency.[77] The system can be optimised by incorporating metal oxides into the procedure to mitigate acid production and promote a high yield of the valuable product benzene in the oil.[81,82]

Gasification

Gasification converts carbonaceous materials into mostly gases, and it can be implemented after pyrolysis of the char, tar, and gas products.[77] The gasification recycling process is shown in Fig. 10. Gasification is conducted at temperatures ranging from 800 to 1500 °C, using a gasification agent (steam, air, or oxygen (O_2)) in sub-stoichiometric amounts,[52,77,85] leading to incomplete combustion.[86] The process primarily produces syngas, accompanied by CO_2, CH_4, and other hydrocarbon gases. Steam gasification yields syngas with a high H_2 content, rendering it well-suited for various chemical synthesis purposes. However, this approach results in increased levels of tar. In contrast, air gasification yields syngas that contains a greater concentration of CO, and H_2 is often used in energy-related contexts as a viable alternative to natural gas.[77] The N_2 present in air dilutes the gaseous products, diminishing their calorific value.[85] The process of O_2 gasification is a more intricate and costly option; however, it includes the benefits of both steam and air agents.[77] Management of tar content is of utmost importance in PET gasification because of its propensity to condense at low temperatures and impede the functioning of gasification equipment or engine components when product syngas is used for energy applications.[77,87] To tackle this challenge, many approaches have been suggested, including careful catalyst and bed additive selection and implementation of downstream tar cracking processes.[87]

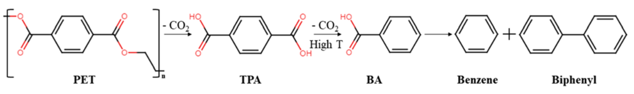

Fig. 9 Schematic representation of the primary thermal cleavage mechanism of PET pyrolysis.[84]

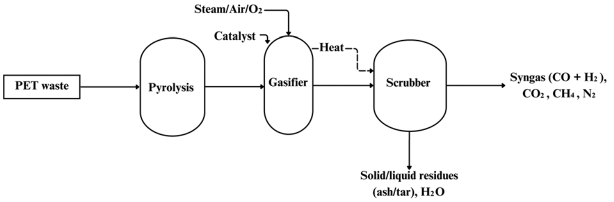

Fig. 10 Schematic of the gasification recycling process (adapted under terms of the CC-BY license.[88] Copyright 2022, MDPI).

Hydrocracking/hydrogenation

Hydrocracking/hydrogenation has similarities with pyrolysis. However, in hydrocracking, PET waste is subjected to heating in the presence of hydrogen (H_2).[51,86,89] Indeed, the hydrocracking process is often used to treat pyrolysis product oils and gases.[90] A hydrocracking/hydrogenation recycling process schematic is presented in Fig. 11. The process involves the breakdown of heavier PET molecules into lighter substances by the cleavage of carbon–carbon bonds, while simultaneously subjecting unsaturated by-products to hydrogenation. Therefore, hydrocracking yields a greater percentage of saturated products compared with pyrolysis, leading to the production of C_7–C_{19} as liquid products of superior quality.[91] A higher fraction of gases is produced from PET compared with other plastic types,[91] and consists of predominantly light C_1–C_5 hydrocarbon gases.[92] A coke-like solid carbon residue is also produced by the process.[92] The usual reaction conditions include temperatures ranging from 300 to 450 °C and pressures ranging from 2 to 15 MPa (20–150 bar) in the presence of pressurised cold H_2. These conditions are generally achieved using a batch-stirred autoclave reactor.[91] To facilitate the cracking activity, the process uses a bifunctional acidic catalyst, often a zeolite.[55]

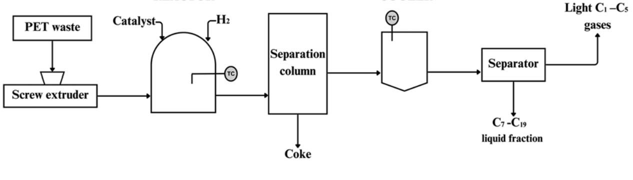

Fig. 11 Schematic of the hydrocracking/hydrogenation recycling process (adapted with permission.[92] Copyright 2005, Elsevier).

Solvolysis (depolymerisation or chemolysis)

Solvolysis is a highly efficient approach utilised in the recycling of plastics, wherein chemical compounds are employed to disintegrate the polymer into its constituent components.[55] Solvolysis exhibits significant advantages in the recycling of contaminated and heterogeneous plastics by enabling possible enhanced filtration and purification processes.[6] The solvolysis approach comprises a range of processes including glycolysis, alcoholysis (methanolysis), aminolysis, ammonolysis, and hydrolysis (acidic, neutral, and alkaline).[6,55]

Glycolysis

Glycolysis is a prominent recycling process that effectively depolymerises PET into its constituent components through a transesterification reaction, which is facilitated by an excess amount of glycols such as EG.[29] The schematic of the glycolysis recycling process is shown in Fig. 12. EG initiates a chemical attack on the ester bonds in PET waste, thereby inducing depolymerisation of the polymer (as shown in Fig. 13).[4] The chemical reaction occurs at elevated temperatures, typically within the range of 180 to 240 °C, and it can be facilitated by the presence of a catalyst such as zinc acetate.[29] The predominant product of glycolysis consists of bis(2-hydroxyethyl) terephthalate (BHET) monomers, accompanied by minor quantities of undepolymerised PET, dimers, low molecular weight oligomers, and potentially other impurities and colours.[3,29] It is crucial to prioritise the removal of undepolymerised PET, as these oligomers can undergo polycondensation. The purification processes may present a level of complexity owing to the potential occurrence of subsequent polymerisation reactions.[4,29] Consequently, vacuum distillation is not a viable option.[29] Instead, effective methodologies to separate the components in the depolymerised mixture include precipitation, agitation, filtration,[3] crystallisation, and membrane or molecular distillation.[80]

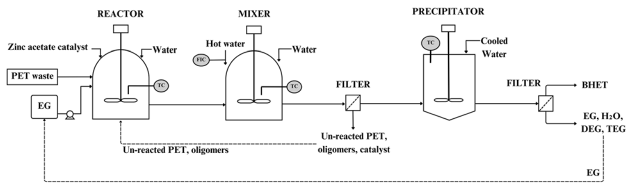

Fig. 12 Schematic of the glycolysis recycling process (adapted with permission.[93] Copyright 2013, Springer Nature).

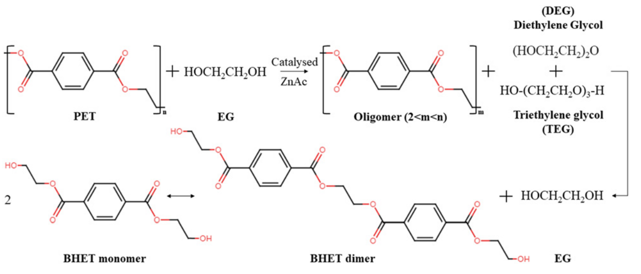

Fig. 13 Schematic representation of the PET glycolysis mechanism.[33]

Alcoholysis

Alcoholysis facilitates the depolymerisation of PET through a transesterification reaction involving the use of an alcohol, which is usually methanol.[29] The schematic depiction of the methanolysis recycling process is presented in Fig. 14. The carbonyl group in the PET polymer chain is subjected to attack by the alcohol, resulting in the formation of dimethyl terephthalate (DMT) and EG.[54] This process takes place at temperatures ranging from 180 to 280 °C and pressures ranging from 2 to 4 MPa, accompanied by a catalyst such as zinc acetate, cobalt acetate, lead dioxide, or magnesium acetate.[6,29] Fig. 15 illustrates a schematic representation detailing the mechanism of PET methanolysis. The resulting products can also consist of some by-products, including phthalate derivatives and a range of alcohols.[29] To ensure the process efficiency, it is essential to eliminate any residual water since it would deactivate the catalyst and form an undesirable azeotrope.[94] Moreover, to prevent further reaction and potential loss of DMT, it is essential to separate the catalyst from the resulting products.[6]

Aminolysis

Aminolysis involves depolymerisation of PET by reacting it with an amine.[29,58] Fig. 16 provides a schematic of an aminolysis recycling process. Amines, such as allylamines, hydrazines, polyamines, and morpholines, undergo a chemical reaction with the ester group in PET, resulting in the formation of monoamine and diamine derivatives of EG and TPA.[4,29,32] Fig. 17 illustrates a schematic representation of the mechanism of PET aminolysis. The reaction takes place within a moderate temperature range of 20–100 °C and is facilitated by the presence of a catalyst, which often includes glacial acetic acid, sodium acetate, or potassium sulphate.[29] Ethanolamine is a commonly observed amine used in aminolysis, resulting

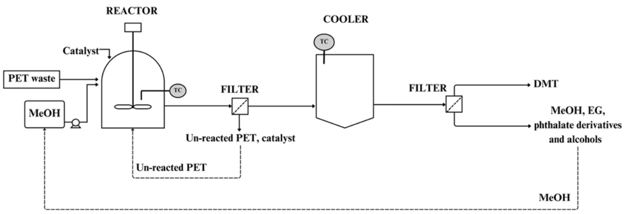

Fig. 14 Proposed schematic of the methanolysis recycling process (based on provided information in ref. 29).

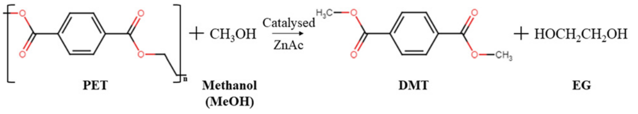

Fig. 15 Schematic representation of the mechanism of PET methanolysis.[4]

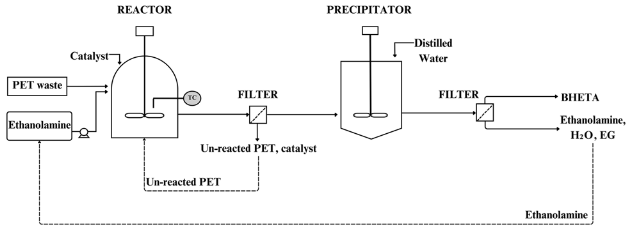

Fig. 16 Proposed schematic of the aminolysis recycling process (based on provided information in ref. 95 and 96).

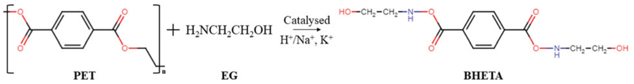

Fig. 17 Schematic representation of the mechanism of PET aminolysis.[6,95]

in bis(2-hydroxyethylene) terephthalamide (BHETA) production, and some EG.[6] The BHETA product can be precipitated thereafter by adding distilled water while agitating.[6,95]

Ammonolysis

The process of ammonolysis is also employed to recycle PET, although it is less explored than other methods.[96] This is achieved by subjecting PET to a reaction with ammonia in the presence of EG solvent[94,96] to produce terephthalamide (TPD) and an additional quantity of EG.[94] The TPD product will easily precipitate as it is insoluble in EG.[96] This reaction occurs within a moderate temperature range of 70 to 180 °C and at pressures approximately equal to 2 MPa. The utilisation of a catalyst, such as zinc acetate, can expedite the reaction.[29] Fig. 18 offers a schematic representation illustrating the ammonolysis recycling process. Fig. 19 provides a schematic representation elucidating PET ammonolysis. Moreover, the TPD product has the potential to generate terephthalic acid nitrile, which can subsequently be transformed into 1,4-bis(aminomethyl) cyclohexane and/or *para*-xylylene diamine.[6,97] These intermediates have significant value in polyamide (PA) synthesis.[96]

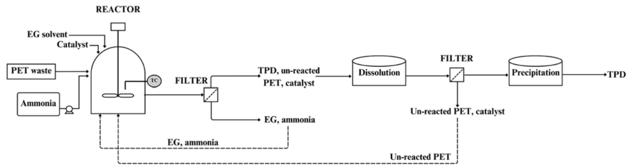

Fig. 18 Proposed schematic of the ammonolysis recycling process (based on information in ref. 96).

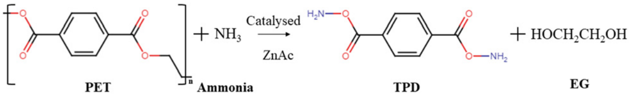

Fig. 19 Schematic representation of the mechanism of PET ammonolysis.[6]

Hydrolysis (acidic, neutral, and alkaline)

Hydrolysis is a widely recognised process for PET depolymerisation, and the one that also occurs in nature and leads to the formation of microplastics (MPs).[98] The process produces TPA and EG, which play crucial roles in the manufacturing of recycled PET.[4,61,94] This method involves the reaction of water with PET, resulting in the conversion of an ester group to a carboxyl group,[94] and it can be conducted under acidic, neutral, or basic conditions.[94,99] The expenses associated with purification of the TPA product can be comparatively high in the context of hydrolysis.[97,99] Moreover, the process tends to proceed at a slower rate compared with alternative recycling methods, primarily due to the comparatively weaker nucleophilic properties of water.[33]

Acidic. The process of acidic hydrolysis commonly employs concentrated sulfuric acid (H_2SO_4) or other mineral acids, such as nitric acid (HNO_3) or phosphoric acid (H_3PO_4).[4,94,97,99] If a concentrated solution of at least 87 wt% H_2SO_4 is utilised, rapid degradation of PET can be achieved at temperatures below 100 °C and atmospheric pressure.[6,97] The TPA product readily precipitates from the reaction mixture, but it can be re-dissolved in a solvent to separate it from the unreacted PET, and then reprecipitated.[100] Fig. 20 presents a schematic of the acidic hydrolysis recycling process, while Fig. 21 illustrates a schematic representation of the mechanism of PET acidic hydrolysis. The process is commonly extended to achieve high purity by utilising an alkaline solution such as sodium hydroxide (NaOH) to form TPA as a sodium salt, separating impurities and colourants, and then once again reprecipitating TPA utilising sulfuric acid.[97]

Neutral. The neutral hydrolysis process utilises hot water or steam at temperatures in the range of 200 to 300 °C, along with pressures ranging from 1 to 4 MPa.[4,94,99] This process is carried out in the presence of excess water, with a PET/water ratio (w/w) ranging from 1 : 2 to 1 : 12 to achieve complete PET depolymerisation.[55,101] To accelerate the process, catalysts such as alkali metal acetates, zeolites, or organophosphorus

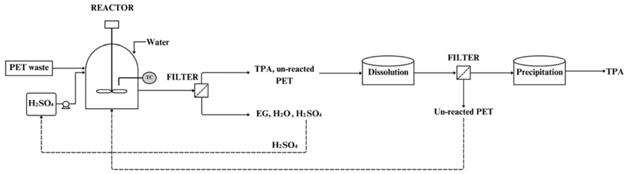

Fig. 20 Proposed schematic of the acidic hydrolysis recycling process (based on provided information in ref. 93 and 100).

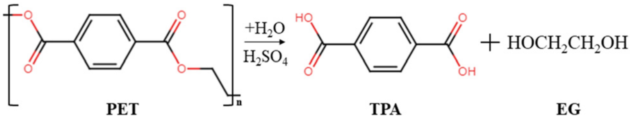

Fig. 21 Schematic representation of the mechanism of PET acidic hydrolysis.[6,93]

compounds can be employed.[55] Fig. 22 provides a schematic of the recycling process for neutral hydrolysis. Additionally, Fig. 23 depicts a schematic illustration of the mechanism involved in the neutral hydrolysis of PET. In contrast to acidic or alkaline conditions, neutral hydrolysis yields a reduced quantity of inorganic salts, thereby rendering it a more ecologically sustainable process.[6,99] Nevertheless, its main limitation lies in the retention of all impurities from PET in the TPA, resulting in a product of comparatively lower purity compared with the output of acid or alkaline hydrolysis.[6,94] The resolution of this matter can be accomplished by dissolving TPA in caprolactam or NaOH, followed by filtration to attain high-purity TPA.[6]

Alkaline. Alkaline hydrolysis is conducted by employing a 4–20 wt% aqueous solution of a strong base, such as NaOH or potassium hydroxide (KOH).[6] The reaction necessitates a temperature range of 210 to 250 °C and pressures between 1.4 to 2 MPa.[6,99] The reaction produces EG and either a disodium or dipotassium terephthalate salt. Consequently, an additional step is required to transform these salts into TPA by employing a strong acid, such as H_2SO_4 or HCl.[6,94] Fig. 24 demonstrates a schematic of the alkaline hydrolysis recycling process, whereas Fig. 25 outlines a schematic of the PET alkaline hydrolysis mechanism.

Enzymolysis

Enzymolysis is a biological degradation mechanism wherein plastics undergo enzymatic and microbial breakdown. This phenomenon can occur spontaneously within the natural surroundings or can be expedited with genetic engineering to produce and/or modify enzymes and microorganisms.[102–104] These processes offer a potentially viable solution for depolymerising waste petro-plastics into components suitable for recycling, or alternatively, mineralising them into CO_2, water, and new biomass.[104–106]

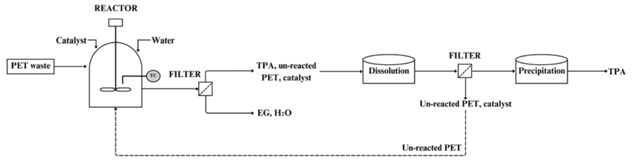

Fig. 22 Proposed schematic of the neutral hydrolysis recycling process (based on provided information in ref. 6 and 93).

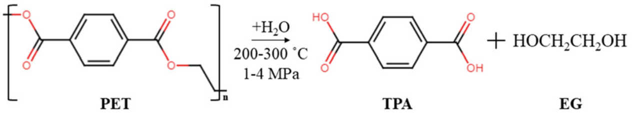

Fig. 23 Schematic representation of the mechanism of PET neutral hydrolysis.[6,93]

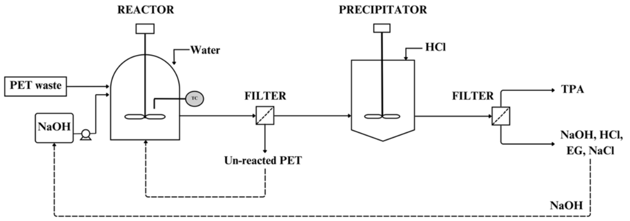

Fig. 24 Schematic of the alkaline hydrolysis recycling process (adapted with permission.[93] Copyright 2013, Springer Nature).

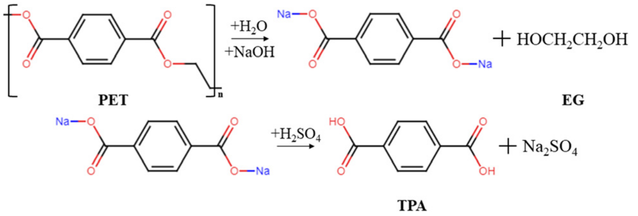

Fig. 25 Schematic representation of the mechanism of PET alkaline hydrolysis.[6,93]

Enzymatic

Enzymatic degradation of PET involves the utilisation of enzymes as biocatalysts to catalyse chemical reactions, with a specific focus on cleavage of ester bonds in PET. The enzymes initially adhere to the polymer's surface, after which the bonds undergo hydroperoxidation/hydrolysis.[104,107,108] This reaction degrades PET into its constituent components, namely TPA and EG,[104,109] enabling their subsequent use for production of recycled PET. Enzymatic hydrolysis is the most extensively researched process for PET degradation. However, other such reactions include oxidative enzymatic reactions, enzymes exhibiting polymerase activity, and enzymes possessing esterification or transesterification capabilities.[4,110] Fig. 26 shows a schematic of the enzymatic recycling process. Extensively investigated enzymes for PET include lipases, cutinases, esterases, and carboxylesterases.[111] Enzymes have a major limitation in that they cannot work properly on bulk PET waste; their function is often limited to small oligomers.[112] Moreover, the activity of PET hydrolysing enzymes can be hindered by by-products from incomplete hydrolysis, specifically mono(2-hydroxyethyl) terephthalate (MHET) and BHET, which function as competitive inhibitors of enzymes. The elimination of MHET and BHET when using enzymes for PET biodegradation, without compromising the enzyme's integrity, presents significant challenges. These factors highlight the

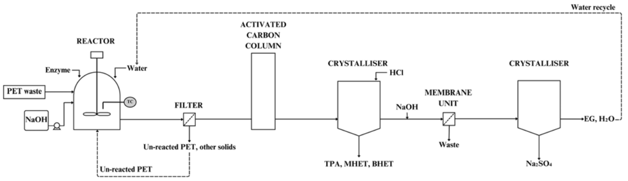

Fig. 26 Schematic of the enzymatic recycling process (adapted with permission.[114] Copyright 2021, Elsevier).

importance of acquiring enzymes that are resistant to inhibition by MHET and BHET, either through direct screening or enzyme modification.[111,113] Fig. 27 provides a schematic depiction of the PET enzymatic degradation mechanism.

Microbial

PET has the potential to undergo degradation through microbial metabolism processes, whereby microorganisms utilise PET as a source of carbon and energy. The schematic illustration of a microbial recycling process is shown in Fig. 28. The microbial metabolism process encompasses a wide variety of mechanisms, including enzymatic hydrolysis, surface erosion, and intracellular depolymerisation, which collectively facilitate PET degradation. Several bacterial and fungal species, including *Ideonella sakaiensis* and *Rhodococcus ruber*, can synthesise enzymes such as PETase and MHETase. These enzymes can catalyse PET hydrolysis, leading to PET depolymerisation into its constituent components. Surface erosion is an additional degradation mechanism that occurs when microorganisms engage in the physical breakdown of the surface of PET. The mechanism of intracellular depolymerisation involves the internalisation of PET constituents below 600 Da by microorganisms, followed by subsequent mineralisation within their cellular compartments into H_2O, CO_2, N_2, and CH_4.[111,115] Furthermore, it is crucial to emphasise that all microbial processes are restricted by scale limitations, necessitating continuous efforts in scaling up to overcome this obstacle.[116] Fig. 29 provides a schematic representation elucidating the intricate mechanism.

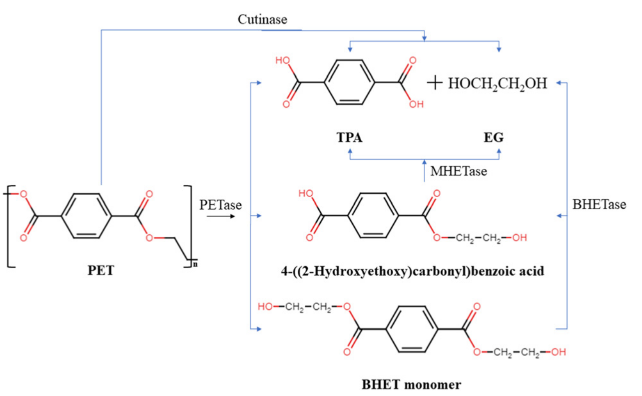

Fig. 27 Schematic representation of the mechanism of PET enzymatic degradation.[111,113]

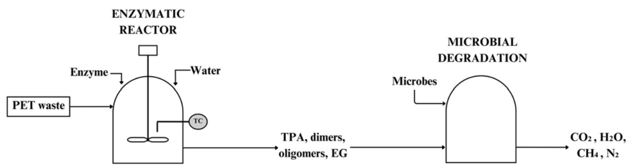

Fig. 28 Proposed schematic of the microbial recycling process (based on provided information in ref. 53).

Photodegradation

Fig. 30 shows the schematic of an ultraviolet (UV) radiation recycling process. Photodegradation is a unique approach for plastics recycling that utilises sunlight or UV radiation, with the assistance of additives or catalysts like titanium dioxide (TiO_2), to induce the disintegration of the molecular composition of plastic *via* free radical generation.[117–120] The resultant smaller constituents can subsequently be employed as a resource for the production of recycled plastics.[118,121] The observed surface effects are believed to be linked to the spontaneous degradation of polymer chains in plastic caused by oxidation. As shown in Fig. 31, this degradation process decreases polymer chain length and molecular weight and increases functional end groups. Additionally, volatile products are released during this degradation process.[118,121,122]

Quaternary recycling

The quaternary recycling route refers to the recovery of energy from plastic waste.[6,53,63,106,125] When alternative recycling routes are neither practical nor affordable, this approach is often the technique of choice.[56] Nevertheless, this route fails to generate a recycled material suitable for repolymerisation, thus lacking the ability to contribute to the circular economy of plastics.[54] This route is typically achieved through incineration.

Incineration is a process that involves the combustion of waste at elevated temperatures, resulting in its conversion into

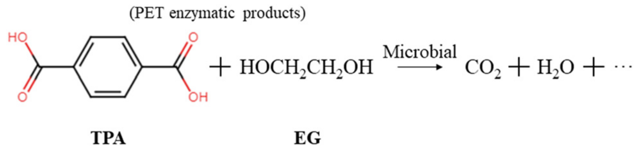

Fig. 29 Schematic representation of PET microbial degradation.[102,104]

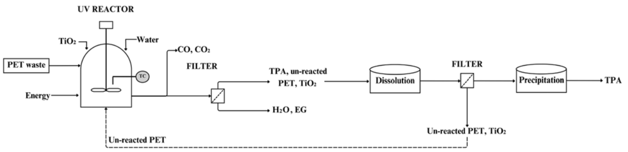

Fig. 30 Proposed schematic of the UV radiation recycling process (based on provided information in ref. 118, 120, 123 and 124).

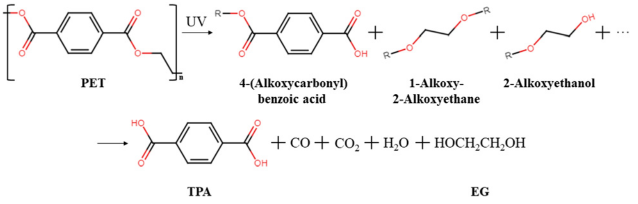

Fig. 31 Schematic representation of the mechanism of PET degradation *via* UV radiation (R: linear straight-chain alkanes).[118]

residue waste (primarily ash[126]), gases, and the generation of heat energy. The generated heat energy can be effectively harnessed to produce electricity, steam, or heat for a wide range of applications.[126] Burning waste with the introduction of air results in significant CO_2 emissions, as well as a variety of undesirable gases such as CO, nitrogen oxide (NO_2), sulphur dioxide (SO_2), and heavy metals that must be managed.[6] A schematic of incineration recycling process is shown in Fig. 32.

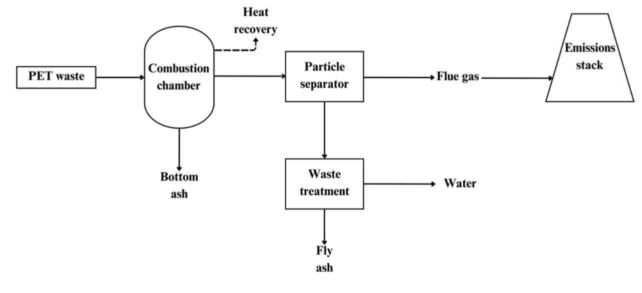

Fig. 32 Schematic of the incineration recycling process (adapted under terms of the CC-BY license.[126] Copyright 2018, Faculty of Engineering of Nnamdi Azikiwe University).

Comparison between different valorisation routes

When selecting a PET recycling method, several factors should be considered, including the quantity and quality of available PET waste, energy consumption, cost-effectiveness, the effectiveness of the method, its environmental impact, and the desired quality of the recycled product. A synergistic approach incorporating a combination of different methods may be utilised to optimise the efficiency of PET recycling.[97] Table 1 provides a summary of significant advantages and disadvantages of each PET recycling method. In addition, the table provides context on the applicability of the recycling techniques for other common plastic types, to provide a broader perspective into the plastic recycling landscape. Moreover, Table 2 has been developed to outline various stages of PET recycling processes, grouped by feedstock pre-treatment, separation of contaminants and unwanted components, purification of monomer and valuable components, and waste treatment opportunities. This table sets the stage for the plethora of opportunities for integration of membrane technology. The integration of membranes at key stages of PET recycling processes not only improves the effectiveness and sustainability of PET recycling but also provides the opportunity to enhance resource efficiency, decrease energy consumption, and mitigate negative environmental impacts. The selective separation and purification capabilities of membrane technology provide an appealing choice for addressing issues related to pre-treating feedstock, contaminant removal, and monomer purification. Moreover, the use of membrane technology presents exciting ways to tackle waste treatment challenges. Furthermore, the strategic use of selective membrane processes enables accurate targeting of specific components, hence enabling the recovery of valuable by-products while reducing the production of additional waste. This not only aligns with sustainability goals but also presents economic opportunities by creating alternative revenue streams.

Navigating the membrane landscape in plastic recycling

To effectively navigate the membrane landscape in plastic recycling, it is essential to have a comprehensive understanding of membrane applications in this field and closely related fields. This understanding enables the identification of approaches for integrating membrane technology into PET recycling. As presented in Table 3, the direct application of membranes in plastic recycling is still very limited, with the primary focus on the removal of MPs, and their application in related processes is similarly restricted. However, each membrane process possesses distinctive characteristics that can be customised to meet the specific recycling requirements. For instance, it highlights processes targeting plastics such as PE, PS, and PVC in wastewater treatment (*e.g.*, a Hydrotech HF2220 disc filter achieves 89.7% removal efficiency[137]), as well as applications such as the separation of phenolic monomers from depolymerised lignin oil (*e.g.*, a separation factor of 1.09 with NF90 membranes[138]) and the purification of pyrolysis reactor products.[139] These applications highlight the potential for membrane technology to enhance performance across diverse processes, stages and polymer types. Although these cases represent only a fraction of membrane technology's potential, they provide promising pathways for enhancing recycling efficiency and sustainability, building on the valorisation routes. The following sections investigate these opportunities by analysing the various ways in which membrane technology can be integrated to meet the changing requirements of sustainable plastic waste management.

Rationale for membrane integration in PET recycling

A comprehensive evaluation of benefits and drawbacks on a route-by-route basis was conducted to identify the recurring limitations that eventually govern yield, product quality, and energy consumption in PET valorisation (Table 1). This comparison revealed five route-agnostic challenges: contaminant and additive removal without quality loss; solvent and monomer recovery with low thermal duty; gas upgrading in thermolysis processes; heat recovery between hot and cold recycle loops; and wastewater treatment that removes microplastics and trace organics at low cost. Membranes are well suited to tackle these limitations due to their size- and solubility-based selectivity, combined with near-isothermal operation, and could simultaneously decrease specific energy and solvent consumption while enhancing product purity and yield.

To ensure that this conceptual framework is applicable to process design, the separations required by each route were divided into four intervention stages: feed pre-treatment, separation of contaminants and unwanted components, purification of monomers and valuable components, and waste treatment (Table 2). Therefore, membrane selection is linked to the specific separation task rather than a general process unit. This framework enables the integration of membrane

Table 1 Significant advantages and disadvantages of each PET recycling route, as well as their applicability for other common plastics

Recycling route			Advantages	Disadvantages	Also applicable for	Ref.
Primary	**Re-extrusion**		Efficient, easy and cost-effective	Material properties deteriorate at each cycle	LDPE, HDPE, PP, PS, PVC, PLA, PU^a, nylon, ABS, PC	32, 55, 61 and 90
			Highest-quality recycled products	Limited to clear PET		
			Lower energy consumption than secondary mechanical recycling as the process doesn't deal with contaminated waste	Difficult to find pure and single-type polymer waste feedstock		
Secondary	**Mechanical**		Does not involve toxic chemicals or produce toxic pollutants	Cannot handle highly contaminated and heterogeneous feedstock	LDPE, HDPE, PP, PS, PVC, PLA, PU^a, nylon, ABS, PC	2, 54, 58, 82 and 127
			The most widely used method of secondary PET recycling	Requires significant amounts of energy and water		
			Well-established, mature and cost-effective technology	This can lead to the degradation of the polymer chains, reducing mechanical properties		
	Dissolution		Can produce high-quality recycled PET products	Less mature technology than mechanical recycling, and can also be difficult to perform at scale	LDPE, HDPE, PP, PS, PVC, PLA, PU, nylon, ABS, PC	3, 65–67 and 128
			Can separate impurities and additives, and can deal with a mixed plastic feedstock due to "selective dissolution"	More expensive than mechanical recycling since it requires more specialised equipment and expertise		
			Relatively low energy requirements compared with mechanical recycling	May require the use of toxic or hazardous solvents		
Tertiary	**Thermolysis**	**Pyrolysis**	Can handle a mixed plastic waste feedstock	High energy requirements	LDPE, HDPE, PP, PS, PVC^b, PLA, PU, nylon, ABS, PC	55, 77, 85 and 90
			Can produce a range of valuable products	PET produces an acidic pyrolysis oil, which can impact the infrastructure, and reduce the fuel efficiency of the product		
			A mature technology, with commercial plants already in operation internationally	Emissions can have an environmental impact if not managed		
		Gasification	A very compatible process with plastic waste feedstocks	Very high energy requirements	LDPE, HDPE, PP, PS, PVC^b, PLA, PU, nylon, ABS, PC	77, 82, 87 and 90
			Can produce a range of valuable products, including electricity, and chemicals	Requires additional expertise/equipment to remove tar from product gas		
			An established technology, already been performed in the petrochemical industry	Emissions can have an environmental impact if not managed		
		Hydrocracking/ hydrogenation	Produces liquid fuels of a higher quality than pyrolysis	H_2 is required for the reaction which is an expensive commodity	LDPE, HDPE, PP, PS, PVC^b, PLA, PU, nylon, ABS, PC	90, 91 and 129
			Handles PET better than pyrolysis due to the H_2 feed	High energy requirements		
			A well-established and well-understood process in the petrochemical industry	Emissions can have an environmental impact if not managed		
	Solvolysis	**Glycolysis**	Produces a high yield of BHET of 85%	Purification of BHET monomers can be complex	PU, PLA, nylon, PC	6, 29, 33, 61 and 94
			No toxic/corrosive chemical components	Difficult to separate the catalyst from the reaction medium as soluble in EG		
			Most industrialised chemical recycling techniques	Relatively high cost		
		Alcoholysis (methanolysis)	DMT is easier to purify than BHET	DMT needs to be converted to TPA to meet PET industrial production trends	PLA, PU, nylon, PC	6, 29, 55 and 94
			High yields of 80 to 85%	Complex and costly separation processes are required to refine the DMT, and remove the phthalate derivative and alcohol by-products		
			Methanol is cheap and widely available	Water must be removed as it can inactivate the catalyst, and the catalyst must be separated from the products to prohibit further reaction and loss of DMT		
		Aminolysis	High yields of around 90%	Amines can be toxic and require careful handling and disposal	PU, PLA, nylon, PC	6, 96 and 97
			Low energy consumption	High cost of amines		
			Products can be used for a variety of applications, such as epoxy resin hardening, or synthesis of PU for adhesives or coatings	Has not been utilised on a commercial scale		
		Ammonolysis	High yields exceeding 90%	Ammonia gas can be toxic and requires careful handling and disposal	PU, PLA, nylon, PC	6 and 96

Table 1 *(Contd.)*

Recycling route				Advantages	Disadvantages	Also applicable for	Ref.
				TPD product can be achieved in high purities of over 99%	High cost of ammonia		
				TPD can be utilised to create PAs with good commercial value	Not well explored		
		Hydrolysis	**Acidic**	High yields of around 90%	Production of inorganic salt by-products in large quantities, which are difficult to dispose of	PLA, PU, nylon, PC	4, 6, 61, 94, 99 and 130
				EG can be oxidised to oxalic acid, which has greater economic value	Generates corrosive acid waste, which can be harmful to the environment if not properly treated		
				Uses water as a reactant, which is abundant and cheap	Costly to recycle large quantities of H_2SO_4, and for the purification of EG from H_2SO_4		
			Neutral	More environmentally friendly than acidic/alkaline hydrolysis	Slow process	PLA, PU, nylon, PC	6, 94 and 99
				High yields of greater than 95% have been reported	Lower purity of TPA, due to persistent contaminants		
				Uses water as a reactant, which is abundant and cheap	A lower yield of EG may result due to catalytic dehydroxylation		
			Alkaline	High yields of over 95% reported	Production of inorganic salt by-products in large quantities, which are difficult to dispose of	PLA, PU, nylon, PC	6, 61, 94 and 99
				Can handle highly contaminated waste PET, whilst still producing TPA of high quality	Difficulty in preventing the formation of unwanted by-products		
				Uses water as a reactant, which is abundant and cheap	Alkaline reagents used in this process can be toxic and corrosive and require careful handling and disposal		
	Enzymolysis	**Enzymatic**		Potential to recycle low-quality and contaminated PET	Enzymes used in the process can be costly to produce	PLA, PU, nylon, PC	4, 53, 114 and 131–135
				Low energy requirements	Scale-up challenges		
				Enzymes can be produced from renewable sources and have good availability	Enzymes can be sensitive to temperature and pH, which can affect their stability and efficiency		
		Microbial metabolism		A more environmentally friendly method of PET degradation	Can be sensitive to environmental conditions	LDPE, HDPE, PP, PS, PVC, PLA, PU, nylon, ABS, PC	53, 102, 134 and 135
				Low energy requirements	Can be a slow process		
				Produces valuable product (methane)	Less mature technology with little knowledge at the industrial scale		
	Photodegradation	**UV radiation**		Can produce high-quality recycled PET products	May require specialised equipment and expertise	LDPE, HDPE, PP, PS, PVC, PLA, PU, nylon, ABS, PC	122 and 136
				A relatively simple and low-cost method of PET recycling	Reaction kinetics may be slow		
				Does not require high temperatures or harsh chemicals, can be more environmentally friendly	Can generate volatiles which can be harmful to the environment		
Quaternary	**Incineration**			Can generate energy in the form of fuel or electricity	Emission of significant greenhouse gases, toxic pollutants and heavy metals	LDPE, HDPE, PP, PS, PVC, PLA, PU, nylon, ABS, PC	6 and 127
				Can provide an option to deal with plastic waste when no other method is available	Highly energy-intensive		
				Reduces the demand on fossil fuels for energy purposes	Not a circular method		

[a] Only thermoplastic PUs can be mechanically recycled, whereas thermoset PUs can undergo physical recycling where they are simply shredded and repurposed. [b] Themolysis of PVC encounters issues with HCl.

Table 2 Feedstock pre-treatment steps, separation of contaminants and unwanted components, purification of monomers and valuable components, and waste treatment opportunities of each PET recycling route

Recycling route				Pre-treatment	Separation		Purification		Waste treatment
					Contaminants	Unwanted components	Monomers	Valuable components	
Primary	**Re-extrusion**			—	—	—	—	—	—
Secondary	**Mechanical**			Cleaning and grinding	—	—	—	—	MP
	Dissolution			Cleaning and grinding	Dissolved impurities, residual colours, additives	—	—	Solvents such as NMP, and non-solvents such H_2O	Wastewater
Tertiary	**Thermolysis**	**Pyrolysis**		Cleaning and grinding	BA and derivatives	C1–C4 gases, aldehydes	TPA, EG	BA, benzene, syngas (H_2 and CO), CH_4	Char, tar
		Gasification		Cleaning and grinding	—	—	—	Syngas, CH_4, N_2	Ash, tar, wastewater
		Hydrocracking		Cleaning and grinding	—	C1–C5 gases	—	C7–C19 oil, syngas, CH_4	Coke
	Solvolysis	**Glycolysis**		Cleaning and grinding	Impurities, residual colours, additives	Dimer of Ethylene Glycol (DEG), and Trimer of Ethylene Glycol (TEG)	BHET, EG	IL catalyst or cobalt/zinc acetate catalyst, un-depolymerised PET, oligomers	Wastewater
		Alcoholysis (methanolysis)		Cleaning and grinding	Impurities, residual colours, additives	Phthalate derivatives and alcohols	DMT, EG	Cobalt/zinc acetate catalyst, methanol, undepolymerised PET	—
		Aminolysis		Cleaning and grinding	Impurities, residual colours, additives	—	EG and corresponding mono- and di-amines of TPA	Corresponding mono- and di-amines of TPA, zinc/lead acetate catalyst, amine, undepolymerised PET	Wastewater
		Ammonolysis		Cleaning and grinding	Impurities, residual colours, additives	—	TPD and EG	TPD, ammonia, zinc acetate catalyst, undepolymerised PET	—
		Hydrolysis	**Acidic**	Cleaning and grinding	Impurities, residual colours, additives	—	TPA and EG	Undepolymerised PET, H_2SO_4	Wastewater
			Neutral	Cleaning and grinding	Impurities, residual colours, additives	—	TPA and EG	Cobalt/Zinc Acetate catalyst, un-depolymerised PET	Wastewater
			Alkaline	Cleaning and grinding	Impurities, residual colours, additives	Inorganic salt; water, HCl	TPA-based salt and EG	Un-depolymerised PET, NaOH	NaCl, wastewater
	Enzymolysis	**Enzymatic**		Cleaning and grinding	Impurities, residual colours, additives	—	TPA and EG	NaOH, HCl, enzymes such as PETases, sodium sulphate, water, MHET and BHET, undepolymerised PET	—
		Microbial		Cleaning and grinding	Impurities, residual colours, additives, oligomers	CO_2	TPA and EG	TPA, Microbes, dimers oligomers, CH_4, and N_2	Wastewater
	Photodegradation	**UV radiation**		Cleaning and grinding	Impurities, residual colours, additives	CO, CO_2	TPA and EG	TPA, TiO_2, un-depolymerised PET	Wastewater
Quaternary	**Incineration**			—	—	Flue gas	—	—	Wastewater, ash

Table 3 Summary of applications and advancement strategies of membranes in plastic recycling, including the pre-treatment of plastic waste feedstock, separation of contaminants, purification of valuable components, and waste treatment

	Targeted component characteristics					
Membrane type	Component	Size (μm)/concentration	Shape/state	Nature of treated sample	Removal efficiency (%)	Ref.
Hydrotech HF2220 disc filter	PE, PS and PVC	4.2–407.1	Irregular solid fragment	Water from wastewater treatment plant (WWTP) by Billund Vand and Energi A/S in Grindsted, Denmark	89.7	137
Biomimetic gill-inspired membrane	PS	0.5, 0.7 and 1	Spherical solid particle	Synthetic aqueous feed	97.6	140
PVDF-quaternary ammonium nanofibrous membrane	PS	0.107 to 1.450	Spherical solid particle beads	Synthetic lake water	92.0	141
PVDF (Millipore VVLP)	PVC, PE and PL	0.74	Solid particle	Synthetic wastewater	84.3	142
Polyethylenimine (PEI) nanofibrous membrane	PS	0.1 and 0.05	Spherical solid particle	Synthetic aqueous feed	99.3 and 89.9	143
CS-polycaprolactone (PCL) nanofibrous membrane	PS	0.1, 0.3 and 1	Spherical solid particle	Synthetic aqueous feed	14.1, 99.8 and 100	144
PVDF (Durapore) Gridded PVDF (Durapore) PTFE (WhatmanTM)	PS	5 wt%	Dissolved organic solution	Packaging waste PS in xylene and limonene	95.8 and 88.90 83.8 and 72.8 38.2 and 19.9	145
Membrane disc-filter	—	~70.4% microbeads, 14.6% fibres, 10.8% fragments, 4.1% films/sheets	Solid microbead, fibre, sheet and fragment	WWTP from municipal sector in Daegu, South Korea	79.4	146
PVDF membrane (GVHP04700 Durapore)	PE, PVC	150, 250	Spherical solid particle	WWTP from domestic sector in Karaduvar, Turkey	100	147
CA nanofibrous membrane	PS	0.1, 0.5, 2, and 5	Spherical solid particle	Synthetic aqueous feed	11.8, 76.9, 99.9 and 99.9	148
PS nanofibrous membrane	PS	0.1, 0.5, 1, 2, 3, 7, 8 and 10	Spherical solid particle	Synthetic aqueous feed	89, 88, 94, 84, 92, >99, >99 and >99	149
Electrospun PAN nanofibrous membrane	PS	0.1 and 0.2	Spherical solid bead	Synthetic aqueous feed	~100 and 100 (for 100 mL feed)	150
Nylon-6 nanofibrous membrane (Ube group)	PS	0.5, 1.0, 6.0, and 10.0	Spherical solid particle	Synthetic aqueous feed	84.5, 95.9, 99.9 and 100	151
Poly dimethyl diallyl ammonium chloride (PDDA)/wood membrane	PS	0.05, 0.3, and 3	Spherical solid bead	Synthetic aqueous feed	98.7, 96.9 and 81.2	152
PES membrane (Guochu Technology Ltd)	PS	0.1	Spherical solid beads	Synthetic lake water	97.3	141
PES membrane (100 kDa, Millipore PBHK)	PE, PVC and PL	0.74	Solid particle	Synthetic wastewater	91.0	142
Commercial membrane (10 kDa, RC70PP, Alfa Laval Nakskov A/S)	Enzyme and TPA	—	Dissolved organic solution	Synthetic feed from PBAT film	Majority recovery of TPA with >95% purity	153
Commercial TiO_2 tubular NF module with 0.9 nm pores (molecular weight cut-off (MWCO) ~ 450 Da, 19 channels, Tami Industries)	PS	0.15/2 mg L^{-1}	Spherical solid particle	Synthetic aqueous feed	100	154
Commercial membrane (200 Da, MPF-34, Koch Separation Solutions, Wilmington, MA, USA)	1-,4-Butanediol (BDO) and adipic acid (AA)	—	Dissolved organic solution	Synthetic feed from PBAT film	70% recovery of AA with 69% purity 65% recovery of BDO with 74% purity	153
NF90 and NF270 (Dow/DuPont)	Phenolic monomer	—	Mixture of organic compounds	Depolymerised lignin oil containing monomer, dimer and low molecular weight oligomers (<3 kDa)	Separation factor of monomer: 1.09 and 0.82	138

Table 3 *(Contd.)*

Membrane type	Targeted component characteristics			Nature of treated sample	Removal efficiency (%)	Ref.
	Component	Size (μm)/ concentration	Shape/state			
Commercial membrane in HMS	PP, PE, PA, PET and PVC	0 to 450	Solid pellet fibre, film, and fragment	Water from WWTP Yantai city, Shandong Province, China	98.0 (after MBR processing and then UF pre-treatment)	155
XC-N (Dow/DuPont)	Phenolic monomer	—	Mixture of organic compounds	Depolymerised lignin oil containing monomer, dimer and low molecular weight oligomers (<3 kDa)	Separation factor of monomer: 1.27	138
PPO/GO supported on a fluoroplast F42L MMM	EG	10 to 30 wt%	Dissolved organic solution	EG aqueous feed	99.8–99.6 wt% water in the permeate	156
Polyester P(BiL$^-$)$_{ROP}$ membrane (624 Da)	Styrene dimer	—	Dissolved organic solution	Synthetic feed containing Styrene dimer in common solvents such as methanol.	3.53	157
Commercial thin-film composites (TFC) membrane (Puramem 280 (Evonik Industries), NF030105, NF030705, NF070706 (SolSep) and oNF-2 (Borsig Membrane Technology))	—	—	Mixture of organic compounds	Pyrolysis reactor liquid products from Urbaser, Spain	Evonik Puramem 280 retention: Cyclohexane, 1,3,5-trimethyl: + 14.5 Cyclohexane, 3,3,5-trimethyl: + 10.9 2,4-Dimethyl-1-heptene: + 7.7 1-Propene, 2-methyl-: −9.2 1-Heptene: −9.8 Pentane, 2-methyl-: −13.1	139
Commercial ceramic tubular membrane (C8, Ph, HDPA (VITO))					Vito C8 retention: Cyclohexane, 1,3,5-trimethyl: +14.1 Cyclohexane, 3,3,5-trimethyl: +12.0 2,4-Dimethyl-1-heptene: +7.9 1-Propene, 2-methyl-: +3.3 1-Heptene: −12.4 Pentane, 2-methyl-: −15.1	
DuraMem 200, DuraMem 300, DuraMem 500, PuraMem 280 and PuraMem s600 (Evonik Industries), NFDS, NFDE and NFX (Synder Filtration), ONF-1, ONF-2 and ONF-3 (Borsig Membrane Technology), NP010 and NP030 (Nadir)	Phenolic monomer	—	Mixture of organic compounds	Depolymerised lignin oil containing monomer, dimer and low molecular weight oligomers	Separation factor of monomer from dimer: 0.64, 1.25, 1.85, 1.26, 3.90 1.57, 1.03, 1.42 4.39, 3.65, 3.20 1.15, 1.48	138
NP030 (Nadir)	Components with <350 Da	—	Mixture of organic compounds	Depolymerised Protobind 1000 alkali lignin containing monomer, dimer and low molecular weight oligomers	Separation factor of components with <350 Da from 350–700 Da: 3.66	158
DM500, DM900, PM280, PMs380 (Evonik Industries)	Vanillin derivatives monomers	—	Mixture of organic compounds	Depolymerised Kraft lignin from pine containing derivatives of vanillin monomer, dimer and low molecular weight oligomers	Separation factor of monomers from dimers: 3.75, 4.02, 4.05 and 3.67	159
Palladium (Pd) alloy membrane	H_2	—	Gas mixture	Pyrolysis reactor gaseous products	H_2 recovery enhancement in the permeate stream and aids in the conversion of biochar into gaseous fuels	160
PTFE membrane	DEG	85 wt%	Dissolved organic solution	DEG aqueous feed	84.9 (separation factor = 2500)	161
Stabilised cellulose-based MBR	PET	Solid film	Hydrolysis PET solution	Synthetic feed from enzymatic hydrolysis	6% PET film weight loss in 24 h and 70% increase in efficiency	162

Table 3 *(Contd.)*

Membrane type	Targeted component characteristics: Component	Size (μm)/concentration	Shape/state	Nature of treated sample	Removal efficiency (%)	Ref.
Regenerated cellulose-based MBR	PET	840/1 mg mL^{-1} and 10 mg mL^{-1}	Hydrolysis PET solution	Synthetic feed from enzymatic hydrolysis of extruded post-consumer recycled bottle-grade PET MPs	More than two-fold improvements in overall PET degradation efficiency and TPA yields	163
PVDF-based MBR	PP, PE, PA, PET and PVC	0 to 450	Solid pellet fibre, film and fragment	Water from WWTP Yantai city, Shandong Province, China	93.2	155
Monochannel nanoporous tubular ceramic membrane (Nanjing Aiyuqi Membrane Technology Co., Ltd, China)	Heat	—	High-temperature gas	$H_2O(g)/CO_2$ gas mixture from stripper stream in a monoethanolamine (MEA)-based rich-split carbon capture process	80% heat recovery	164
Commercial PP, lab-made PP, lab-made PVDF MF membrane	Water and sodium chloride (NaCl)	—	Dissolved mineral solution	Wastewater provided by Kuwait institute of scientific research	Wastewater conversion into freshwater while extracting useful salt, with a recovery factor of 37%, yielding 16.4 kg NaCl/produced water m^3	165

technology at key points to enhance resource efficiency, decrease energy consumption, and mitigate environmental impacts *via* selective separation and purification. To complement this, a comprehensive understanding of membrane options and performance variables can be derived by analysing a summary of the applications and advancement strategies of membranes in plastic recycling and closely related areas (Table 3).

The membrane technology landscape was categorised into three families aligned with these stages. Conventional filtration was employed where pore/cut-off governs performance; emerging solution-diffusion processes were implemented where affinity and volatility dominate; and specialised applications were used for thermally driven separations, phase-change mediated product isolation, bioprocess intensification with selective retention, and membrane-enabled heat integration. Then, a comparative mapping of process streams to suitable membrane technologies was established (Table 4), with a focus on size exclusion, separation targets, material compatibility, and operational rationale. This table bridges the route-specific opportunities in Table 2 with mechanism-led choices and highlights how membranes tackle critical challenges *via* staged, hybrid designs.

Perspectives on integrations of membrane technology in PET recycling

Membrane technology could potentially address a wide range of applications in PET recycling, including the pre-treatment of plastic waste feedstock, separation of contaminants, purification of valuable components, and waste treatment. PET recycling could involve the application of a number of different membrane categories, including conventional membrane processes, emerging membrane processes, and specialised membrane applications.

Conventional membrane processes

Membrane filtration processes, such as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO), utilise membranes to separate particles or solutes based on their respective sizes and/or affinity for the membrane. Except for NF and RO membranes, these membranes are categorised according to pore size and their capacity to retain particles or solutes of specific dimensions. As shown in Fig. 33, filtration techniques find crucial applications in PET recycling. These applications include separating contaminants like dirt, dust, labels, dyes, pigments, and adhesive residues, and purifying valuable components such as monomers, solvents, catalysts, and various chemicals. Moreover, filtration could be used to eliminate MP and nanoplastic (NP) pollutants from waste streams. The efficiency of this approach depends on the dynamic interaction of various factors. These factors include the characteristics of the membrane material (*e.g.*, porosity and surface area), plastic waste variable attributes (including particle size, shape, and concentration), and operating conditions.[137,166–170]

MF membranes feature a pore size ranging from 0.1 to 10 micrometres (μm) and are commonly made from a variety of polymeric materials such as polyethylene (PE), polyvinylidene fluoride (PVDF), polyacrylonitrile (PAN), and PA, as well as ceramic materials like alumina, zirconia, titania, kaolin, and mullite.[171] MF membranes can show strong potential in PET recycling, particularly mechanical recycling processes such as dissolution by efficiently separating contaminants, including suspended solid impurities such as undissolved materials, PET additives, and undesired by-products from the recycling stream. Furthermore, MF membranes can be used in tertiary recycling processes, such as alcoholysis or methanolysis, to remove MPs, solid impurities, and particulate additives from the reaction slurry. This application helps reduce waste stream pollution while improving the purification of valuable components such as DMT or glycol derivatives. MF membranes primarily operate on size exclusion, where they effectively capture particles in this range while allowing other components to pass through for collection. This technique enhances downstream processing efficiency and reduces environmental impact by decreasing solid waste generation. Furthermore, MF has shown efficacy in removing MPs from waste streams. Pramanik *et al.* documented an 84.3% removal efficiency using a commercial PVDF MF membrane with a pore size of 0.1 μm to remove a combination of PVC, PE, and polyester (PL) particles, averaging 0.74 μm in size, from water.[142] The removal of PET additives and by-products from tertiary recycling streams using MF membranes becomes increasingly significant as the complexity of waste feedstock evolves in modern recycling systems. However, MF membranes are prone to fouling and clogging due to the accumulation of PET additives and by-products like plasticisers, which can reduce their long-term performance. For example, a key challenge in purifying acid-precipitated TPA crystals from the alkaline hydrolysis process is fouling of the MF membranes. To address these issues, the development of anti-fouling membrane materials with hydrophilic coatings or other surface modifications could improve operational stability, while innovative MF systems with self-cleaning mechanisms could effectively mitigate clogging. Furthermore, employing materials with higher chemical and thermal resistance could increase the durability of these membranes under extreme recycling conditions. Due to the wide size range of hydrolysis and glycolysis liquors – from very small molecules like EG (~62 Da; ~0.35 nm) and methanol (~32 Da; ~0.26 nm), to monomers such as MHET (~194 Da; ~0.65 nm) and BHET (~254 Da; ~0.76 nm), and extending to oligomers (~500–1000 Da; ~1–1.5 nm) – a staged design incorporating tight-UF/loose-NF (~1–3 kDa), NF (~200–500 Da), and RO (<200 Da) cut-offs can enhance the separation performance while mitigating fouling.

UF membranes have pore sizes between 0.008–0.2 μm and are commonly fabricated from PAN, PVDF, polysulfone (PSF), polyethersulfone (PES), and cellulose acetate (CA).[172] These polymers are chosen for their chemical resistance, mechanical strength, and ability to form tight pore structures. These characteristics make UF membranes effective in processes such as glycolysis and hydrolysis, which frequently generate compounds that contain EG, oligomers, and catalysts. In the PET recycling process, particularly in tertiary recycling processes such as acidic hydrolysis, UF can selectively recover valuable products such as BHET and remove smaller dissolved by-products or dispersed organic and inorganic contaminants such as residual additives and residual oligomers. This is achieved by separating these components based on the size exclusion principle, thereby improving the quality and purity of the recycled PET material. Numerous studies have underscored the potential of UF membranes in enhancing separation efficiency by their effectiveness in removing a wide range of contaminants.[173–176] Using UF membranes also offers a promising way to mitigate the environmental impact of plastic waste treatment by enhancing purification and reducing waste generation.[177] For instance, Alterkaoui *et al.* used a loose 150 kDa PES UF membrane to recover NaOH from industrial PET-washing streams. The membrane directly treated the highly alkaline fluid and recovered 98.6% of the NaOH while operating at only 5 bar, which means offering a low-energy route to recycle cleaning agents.[178] Additionally, UF membranes can be implemented in enzymatic hydrolysis to concentrate enzymes for re-use, thereby enhancing enzymatic efficiency and reducing process costs. For example, the recovery of the enzyme PETases has been a subject of research in the past decade. This line of inquiry aims to translate laboratory successes in PET microbial degradation into a tangible real-world impact using membranes.[179] Ismail *et al.* used a commercial UF membrane to separate enzymes and TPA from a dissolved organic solution obtained from depolymerisation of polybutylene adipate-*co*-terephthalate (PBAT) film.[153] In these enzyme loops, UF cut-offs within the 10–100 kDa range effectively retain PETases/cutinases (~30–50 kDa; ~4–6 nm) while allowing the passage of small depolymerisation products such as MHET (~194 Da) or BHET (~254 Da) to the permeate. This facilitates enzyme recycling without product accumulation. Despite these advancements, UF membranes exhibit limitations in removing ultra-fine dissolved solutes or nanoparticles and sometimes need high operating pressures, resulting in increased energy consumption. Therefore, ongoing research should focus on refining pore structure precision to increase separation efficiency while preserving high flux, as well as on the development of selective membranes capable of enhancing the retention and recovery of valuable components, such as PETase enzyme, from depolymerisation processes. Furthermore, integrating UF with pre-treatment processes like MF, using membranes with optimized pore sizes, could enable more precise filtration, reduce fouling, increase efficiency, and improve the quality of the recovered material.

NF membranes with pore size varying from 0.0008 to 0.01 μm, commonly fabricated from materials such as PA, PS, PET, and CA,[180] can effectively reject dissolved components from the PET recycling process such as alkaline or acidic hydrolysis. Components include residual colours, monomers, dimers, trimers and low molecular weight oligomers as well as NPs from waste streams. For example, Severino *et al.* recently demonstrated that a commercial TiO_2 tubular NF module with 0.9 nm pores (MWCO ~450 Da, 19 channels) could capture PS NPs with 100% rejection. They concentrated the NPs from 2 mg L^{-1} to 100 mg L^{-1} and reduced the wastewater volume by a factor of 44.25.[154] In addition, alkaline hydrolysis produces compounds such as NaCl, EG, and TPA, which can be effectively separated and recovered using NF membranes. Aside from the purification of PET components, NF can also play a vital role in recycling valuable components like catalysts owing to its pore sizes that are suitable for these components, such as the transition metal-containing ILs ([Bmim]$ZnCl_3$ and $[Bmim]_2CoCl_4$) utilised for the PET glycolysis process, which are known for their high efficiency and mild catalytic conditions.[181] Employing membrane technology to recover these IL catalysts is promising, as glycolysis occurs under relatively mild conditions, allowing for flexibility in selecting appropriate membrane materials. Studies utilising commercial NF membranes like FilmTec (NF270),[182] and Desal DK[183] have demonstrated 70% IL recovery. Particularly for glycolysis liquors, NF with an effective MWCO of ~200–1000 Da can be adjusted to retain the BHET monomer (~254 Da; ~0.76 nm) and strongly its dimers/trimers (~ 500–1000 Da; ~1–1.5 nm) while allowing ions to pass. This makes tight-UF/loose-NF a practical boundary for oligomer separation. Other PET-related solutes are captured by similar NF membranes across processes, including DMT from methanolysis (~194 Da; ~0.61 nm), amide derivatives from aminolysis (~250–300 Da; ~0.7–0.9 nm), and aromatic acids formed in thermolysis/pyrolysis, such as benzoic/phthalic species (~122–166 Da; 0.6–0.7 nm). In this range, critical additives are also addressable. For instance, the phthalate plasticiser DEHP (~390.6 Da; ~0.9 nm) and the azo dye Solvent Red 24 (~380–500 Da; ~1.0–1.2 nm) are efficiently rejected by NF with ~300–1000 Da and ~300–500 Da cut-offs, respectively. Smaller stabilisers, such as benzotriazole (~119.1 Da; ~0.5–0.6 nm), are located near the NF/RO boundary (~200–300 Da), which influences the selection of the membrane for separation. However, NF membranes often face issues including fouling and scaling from organic and inorganic contaminants and complex additives. Additionally, their scalability is hindered by the high capital expenses associated with high-performance membranes. Hence, the development of improved fouling-resistant materials and coatings could substantially extend their operating lifetime. At the same time, improving NF membranes for enhanced selectivity and recovery rates of specific PET components, such as catalysts, could improve the economic viability of recycling processes. Cost-effective manufacturing techniques for the large-scale production of NF membranes, while maintaining performance, will be essential for their broader implementation.

RO membranes with pore sizes between 0.0001–0.004 µm, commonly produced from PA or CA materials, are a promising technology for treating plastic recycling wastewater produced during PET recycling processes.[184] It can be particularly beneficial for wastewater streams generated from tertiary recycling processes, such as aminolysis, ammonolysis, alcoholysis, or hydrolysis, as it significantly reduces impurities and dissolved solids. For instance, while aminolysis is effective at PET recycling, it raises environmental concerns due to amine waste. T. Fujioka *et al.* addressed this by proposing a hybrid ultraviolet RO process tailored to remove *N*-nitrosodimethylamine (NDMA) from aqueous solutions.[185] In the RO process, PET waste streams pass through membranes at elevated pressure, selectively allowing water to permeate while retaining smaller impurities, dissolved solids, salts, monomers, and trace organic contaminants, which results in purified water of exceptional quality and purity, suitable for diverse applications. Moreover, RO could prove valuable for treating and purifying plastic recycling wastewater generated during incineration, aiding in meeting discharge regulations and mitigating environmental impact while concentrating valuable monomers like TPA and EG. For instance, alkaline hydrolysis produces a mixture of salts and water that RO membranes can purify, thereby promoting environmental sustainability by decreasing wastewater discharge. The development of recycling technology significantly contributes to more environmentally conscious waste management and efficient resource utilisation practices. Apart from their advantages, RO systems are energy-intensive due to high-pressure requirements. Moreover, the brine produced during RO processes presents a disposal challenge, and the membranes are prone to clogging and degradation when exposed to amine waste, organic materials or viscous by-products. Consequently, future research can focus on developing low-energy RO membranes that require reduced operating pressures, hybrid systems with pre-treatment techniques like UV oxidation to reduce fouling, and innovative strategies for brine management *via* valorisation or advanced disposal techniques. Operationally, RO's < 200 Da selectivity is well suited to the recovery of solvents and small molecules across PET routes. This allows for the refining and recycling of methanol (~32 Da) from methanolysis, EG (~62 Da) from hydrolysis/glycolysis, and even DMSO (~78 Da) used in solvent-extraction stages.

Emerging membrane processes

Separation processes, including pervaporation (PV), organic solvent nanofiltration (OSN), and gas separation (GS) are membrane-based processes that have been developed with the primary objective of effective separation of contaminants, purification of valuable components from mixtures, such as liquid–liquid or gas–liquid systems, and catalyst retention. These separation processes rely on advanced materials and tailored membrane structures to improve separation efficiencies. The incorporation of novel materials, such as intrinsically microporous polymers, metal–organic frameworks (MOFs), and hybrid nanocomposites, can enhance selectivity and chemical resistance.

PV membranes are commonly made from polymeric materials including CA, poly(vinyl alcohol) (PVA), polyether-*block*-amide (PEBA), polydimethylsiloxane (PDMS), and chitosan (CS) as well as inorganic materials like zeolite, silica and silicate, and metal oxides (*e.g.* γ-alumina, titania, zirconia).[186]

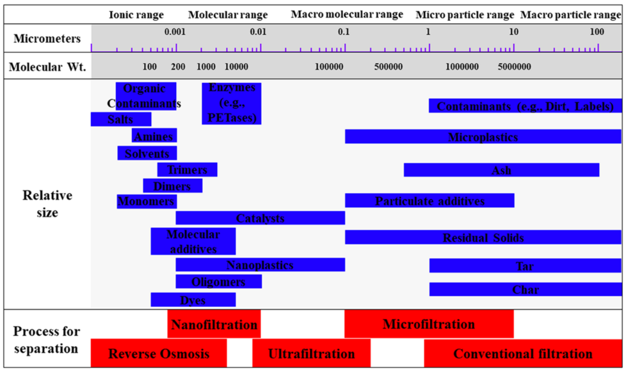

Fig. 33 Perspectives on the integration of conventional membrane processes in PET recycling: illustration of potential component separations from the outputs of various PET recycling processes.

These membranes operate based on the solution-diffusion mechanism, where components dissolve into the membrane, diffuse through it at varying rates, and then evaporate as permeation. This mechanism enables the selective separation of solvents from contaminants according to their volatility and solubility. Meanwhile, PV membranes operate under moderate conditions and bypass the thermodynamic vapour–liquid equilibrium limitations of traditional distillation processes, which leads to lower energy consumption. These can reduce reliance on new solvents and decrease total expenses.[187,188] Therefore, PV membranes may have significant importance in PET recycling, as they allow for precise and efficient separation of contaminants while simultaneously purifying valuable components from impurities and by-products with low energy consumption. On the other hand, the use of PV membranes can also contribute to environmental sustainability by reducing overall waste volume. For example, as shown in Fig. 34a and b, PV membranes can excel at recovering valuable solvents like EG from mixtures with water and other by-products in PET glycolysis process, outperforming traditional distillation methods due to their enhanced selectivity. They can increase the efficiency of solvent separation, resulting in a substantial boost in the yield of pure solvent during recycling processes. PV membranes can also recover high-purity solvents for reuse in other recycling processes like aminolysis, ammonolysis, and alcoholysis. Additionally, PV membranes may prove advantageous in regenerating solvents commonly used in dissolving PET for dissolution recycling process, such as NMP and benzyl alcohol, owing to their high efficiency and low energy consumption. Furthermore, PV membranes can be integrated into PET pyrolysis. Catalytic pyrolysis converts a large amount of the material into valuable products such as benzene.[81] PV membranes could then be employed to purify this benzene by effectively removing impurities like aldehydes. Despite these advantages and potential applications, PV membranes face specific challenges such as membrane fouling and maintaining performance under industrial conditions. These challenges could potentially be solved by developing new materials, such as crosslinked block copolystyrene membranes with flexible side chains, to ensure consistent performance throughout long operational times.

OSN, a special form of NF, is typically made from materials like PSF, PES, PAN, PA, polyimide (PI), and polymers of intrinsic microporosity (PIMs).[189] OSN is a membrane-based process where selective separation of the species present in the solution is obtained based on their size and relative affinity for the membrane. Molecules smaller than the so-called MWCO of the membrane will pass through the membrane, while species larger than the MWCO are selectively retained. Typical MWCO values for commercial OSN membranes range from 150 to 1000 Da. This potentially makes them ideal for the selective recovery of homogeneous catalysts having a large molecular weight, separation of monomers from oligomers, and organic solvent recovery. In PET recycling, OSN membranes exhibit strong potential in the recovery of valuable liquid products such as in hydrocracking and pyrolysis processes, while simultaneously allowing for the effective reuse of solvents in solvolysis processes. For instance, OSN membranes can retain high-molecular-weight catalysts such as cobalt-based IL or selectively recover important liquid products like C_7–C_{19} oil fractions or solvents like methanol in methanolysis (see Fig. 34c, and d). Additionally, OSN has the distinct advantage of low energy requirements since phase transitions, such as evaporation, are avoided in this separation technique. The absence of thermal phase transitions also removes the issue of thermal stress for the recovered catalyst, which is present in other recovery techniques. This highlights the significant potential of OSN for catalyst recovery while maintaining its integrity. Recent studies, such as the work by Wen *et al.*, have demonstrated effective catalyst recovery using commercial OSN membranes.[190] For aminolysis, ammonolysis and alcoholysis processes, OSN could aid in the recovery and recycling of these catalysts, reducing waste and minimising environmental impact. Although this study showcased the potential of OSN in catalyst recovery, it also highlighted challenges such as catalytic deactivation and membrane fouling. This challenge could intensify with viscous solutions like depolymerised PET oligomers. Future research should focus on developing more robust membrane materials and advanced surface chemistries to minimize fouling and ensure consistent performance in chemically aggressive, high-viscosity environments often seen in PET recycling. Also, catalyst recovery work in PET-relevant catalytic systems is still lacking.

GS membranes are fabricated from a diverse range of materials, including polymeric membranes such as PIs, PIMs, and perfluoropolymers; inorganic membranes like zeolites, silica, MOFs, and carbon molecular sieves (CMSMs); metallic membranes made from metal alloys; and emerging materials such as graphene and other two-dimensional (2D) materials.[191] These membranes are highly efficient at selectively separating gas mixtures based on differences in molecular size and membrane affinity.[192] GS membranes allow specific gas components to preferentially pass through the membrane, driven by differences in partial pressures. This leads to advantages such as low energy consumption, environmental sustainability, and operational simplicity. These characteristics make GS membranes particularly promising for PET recycling like pyrolysis or gasification.[193] In PET pyrolysis and gasification, the aim is to transform polymer waste into valuable gas products like syngas and by-products like C_1–C_4 gases. To meet the required composition to be used in various technologies such as Fischer–Tropsch synthesis, it is important for the syngas to generally have a high concentration of H_2 and low quantities of acid gases.[194,195] As illustrated in Fig. 34e and f, GS membranes can be used to enhance the quality of syngas and decrease downstream processing costs by adjusting the H_2/CO ratio and removing impurities. For instance, in pyrolysis off-gas, where the primary target species are C_1–C_4 hydrocarbons (~<100 Da; ~0.5 nm), sub-5 Å GS membranes can selectively sieve CH_4 and C^{2+} from the more rapidly diffusing H_2/CO, thereby enhancing the syngas in the permeate and concentrating a valuable light-fuel fraction in the retentate. In addition, GS membranes have the potential to remove CO_2 that is produced during biological PET degradation processes by enzymes or microorganisms.[196,197] For example, the inclusion of a palladium (Pd) alloy membrane in the pyrolysis reactor, due to its excellent H_2 selectivity, enhances H_2 recovery in the permeate stream and aids in the conversion of biochar into gaseous fuels.[160] Additionally, selective membranes can also be designed to remove N_2 and CO_2 from H_2 during the gasification process and enrich H_2 purity. The presence of CH_4 as a valuable energy resource in syngas presents another opportunity for its selective purification and recovery. However, GS membranes experience difficulties in harsh conditions despite these benefits. High-pressure and high-temperature gas streams can compromise membrane performance. Therefore, the development of new materials with improved chemical and thermal stability remains a critical area for future research.

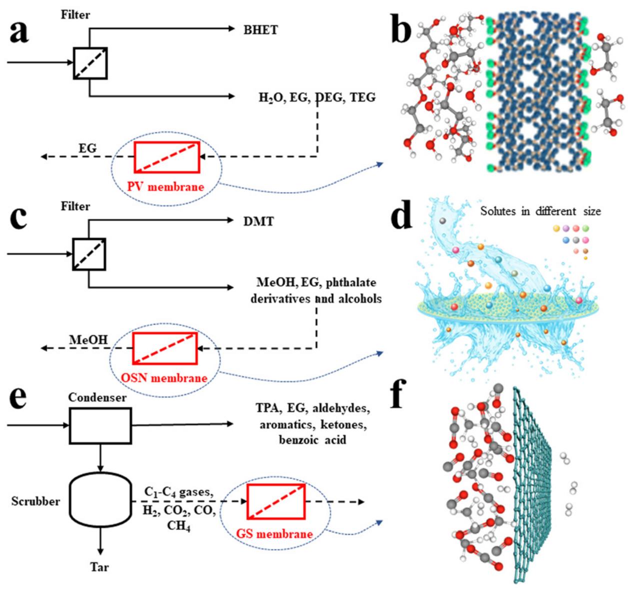

Fig. 34 Perspectives on the integration of emerging membrane processes in PET recycling, showing process and membrane schematics for: (a and b) potential application of PV membranes in PET glycolysis for EG purification; (c and d) application of OSN membranes in PET methanolysis for methanol recovery; (e and f) use of GS membranes in PET pyrolysis for selective H_2 separation (image for b): adapted with permission.[198] Copyright 2023, RSC; image for (d): adapted with permission.[199] Copyright 2024, ACS; image for (f): adapted with permission.[200] Copyright 2013, Elsevier.

Specialised membrane applications

Specialised membrane applications refer to membrane applications that are designed to fulfil distinct and specific purposes. One example is membrane distillation (MD), which is commonly employed for water treatment and resource concentration processes.[161,201] Another application, the membrane bioreactor (MBR), combines membrane separation with biological treatment for efficient plastic recycling wastewater treatment.[155] Furthermore, membrane heat exchangers (MHEs) excel in heat transfer processes,[164] while membrane crystallisation (MCr) is specifically employed for crystallisation of solutes from solutions.[165]

Membrane-based vapour separation processes such as MD are emerging as an effective thermal separation technology. MD membranes are typically fabricated from polymeric membranes such as PP, PE, PAN, PVDF, PTFE, and PES/PVDF blends or ceramic membranes like alumina, titania, and kaolin.[202] As shown in Fig. 35a, MD uses hydrophobic membranes to segregate liquid mixtures based on differences in vapour pressure.[203] In this process, a hot feed solution contacts one side of the membrane, while the other side remains cold or under vacuum, or swept with a gas. This creates a vapour pressure gradient that drives the selective transport of volatile components through the membrane, while non-volatile compounds remain in the feed. This mechanism allows the recovery and recycling of solvents used or produced in processes such as dissolution, glycolysis, and hydrolysis, substantially reducing the need for fresh solvent and enhancing the economic and environmental sustainability of recycling processes. Moreover, MD has the ability to selectively remove impurities such as water from recycling stream mixtures, thereby concentrating valuable components like EG, and monomers (*e.g.*, TPA). For instance, for example, MD can recover EG used in the glycolysis process from water or EG produced during alkaline hydrolysis process from inorganic salts to facilitate its utilisation in subsequent reactions. It also shows great promise for concentrating other valuable compounds like catalysts.[204] Additionally, MD can offer effectiveness in concentrating by-products such as sodium sulphate from acidic or neutral hydrolysis waste streams, thereby reducing waste volume and enhancing economic sustainability. It can also be used to recover valuable chemicals from off-gas streams generated during incineration,[156,161] including targeted recovery of specific chemicals from pyrolysis-produced condensate oils. This supports a more circular approach to waste management. Recent advancements in MD have focused on enhancing membrane materials and system design to address issues such as fouling, pore wetting, and energy efficiency.[205] For example, incorporating modified additives into membrane structures has been shown to improve hydrophobicity and reduce fouling by lowering surface energy.[206] Similarly, dual-layer and multi-layer electrospun membranes with customised pore sizes and surface roughness have demonstrated enhanced flux and stability.[207] These enhancements make MD membranes appropriate for demanding plastic recycling applications. However, some challenges remain, including the development of anti-swelling membranes to maintain stable performance in aqueous systems, and the design of pore structures that optimise both selectivity and flux.[208,209] Future studies should focus on the development of such membranes and continue to explore materials that enhance MD performance in plastic recycling.

MBR is an advanced treatment system that combines biological degradation with MF or UF membrane filtration.[210] It is typically made from organic materials including such as PVDF, PES, PP, and PTFE or inorganic materials like ceramics, zirconium dioxide, silica, and silicates.[211] MBR uses a semi-permeable porous membrane to separate treated water from biomass and suspended solids (Fig. 35b). Membrane modules can be integrated directly into bioreactors to improve contact between microorganisms or enzymes and target components. This creates efficient reaction systems with enhanced degradation efficiency.[212] MBR systems offer additional benefits such as higher effluent quality, better pathogen removal, and compact design.[213] These developments have resulted in the widespread use of MBRs in a broad range of applications and they have become an essential part of modern water treatment processes.[214] Such systems can be effectively integrated into plastic recycling processes like PET enzymatic hydrolysis to separate produced monomers (*e.g.*, TPA, EG) and concentrate enzymes such as PETases for reuse. MBRs hold promise for reducing downstream purification needs, lowering capital costs, and accelerating biological depolymerisation. For example, Barth *et al.* integrated a UF membrane into a PET recycling reactor. The continuous removal of inhibitory hydrolysis products by the UF membrane increased the efficiency of biocatalytic hydrolysis of PET by 70% over 24 h compared with hydrolysis performed in batch mode.[162] Ayafor *et al.* have recently used an *in situ*-product-removal dialysis reactor. This reactor retains LCC-ICCG cutinase while continuously diffusing TPA, MHET, and protons out of the reaction zone through a 7 kDa UF cellulose membrane. This system resulted in a more than two-fold increase in both the overall PET depolymerisation and TPA yield (at 10 mg PET per mL) compared with a similar batch system that was run for 120 h.[163] Moreover, optimising antifouling properties, membrane surface area, and enzyme recycling strategies can further improve MBR performance, especially in breaking down PET additives and by-products. Despite the considerable benefits of MBR technology, it faces obstacles like membrane biofouling, high maintenance expenses, and the need for regular cleaning, which limit its long-term economic viability. Therefore, future research should focus on the development of advanced antifouling membrane materials, including coatings or surface modifications that prevent biofilm growth and reduce cleaning frequency. Moreover, scalable and cost-effective enzyme recycling methods could lower operating expenses and environmental effects, enhancing the feasibility of MBRs for extensive use in plastic recycling.

MHEs are innovative heat transfer devices that utilise semi-permeable membranes to enable efficient heat exchange between two fluid streams while preventing mixing. MHEs are often made from PVDF, PES, silica, alumina, carbon, and titanium.[164,215–217] As illustrated in Fig. 35c, these membranes act as a selective barrier that allows only heat transfer, which makes MHEs particularly suitable for handling fluids with significant temperature differences or those prone to fouling or corrosion. Therefore, MHEs can offer energy-efficient and economically viable solutions for the energy optimisation and thermal integration of plastic recycling processes such as incineration, pyrolysis, and gasification processes. Indeed, by recovering heat from high-temperature streams (*e.g.*, flue gases, syngas, char, and tar), MHEs reduce total energy consumption and enhance process sustainability.[164,218] Recovered heat can be reused for generating steam or preheating combustion air which contributes to greater system efficiency and lower operational costs.[215] For instance, MHEs can optimise syngas production by capturing waste heat. This helps maintain optimal reactor temperatures during the gasification process, thereby reducing overall energy consumption. The

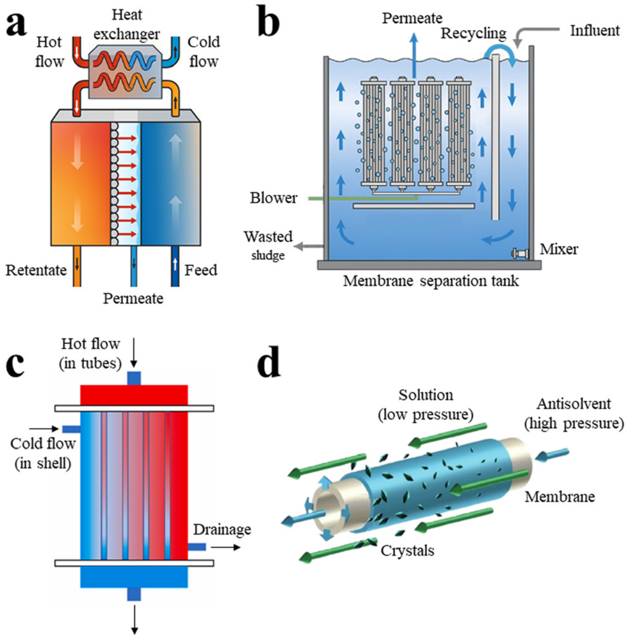

Fig. 35 Perspectives on the integration of specialised membrane applications in PET recycling, illustrating membrane schematics for: (a) MD, (b) MBR, (c) MHE, and (d) MCr (image for a): adapted with permission.[225] Copyright 2021, Elsevier; image for (b): adapted with permission.[226] Copyright 2015, MDPI; image for (c): adapted with permission.[216] Copyright 2023, Elsevier; image for (c): adapted with permission.[220] Copyright 2023, RSC.

Table 4 Comparative mapping of PET recycling process streams to membrane technologies

Process stream (phase)	Key species to recover/remove (examples)	Size guide (Da/~nm or µm)	Suggested membrane sequence integration (top → bottom; typical cut-offs)	Why this works	Material notes (compatibility/temperature/pH)
Wash water/plant wastewater (aq)	Remove: labels, dirt, fines, MPs/NPs; surfactants, trace organics; salts	MPs 1–500 µm; colloids ~0.05–1 µm; ions <1 nm	MF 0.1–1 µm → UF 50–150 kDa (if colloids are high) → NF/RO treatment	Size exclusion protects downstream; RO closes water loop	PVDF/PP/ceramic for MF/UF; PA for RO; cleanable-in-place
Dissolution liquor (org)	Recover: PET; Remove: undissolved, pigments/additives; recycle NMP/benzyl alcohol	Particles >0.1 µm; dyes 300–1000 Da; solvents 32–200 Da	MF 0.2–0.45 µm → OSN 300–600 Da → PV/MD for solvent/non-solvent recycle	Clarify, then cut dyes/additives; close solvent loop at low thermal duty	Solvent-resistant OSN (PI/PES/PIM); PA not suitable for NMP; ceramics/PTFE for MD; $T \leq 80$ °C
Glycolysis liquor (aq/EG-rich)	Recover: BHET (254 Da), EG (62 Da); Remove: oligomers 500–1000 Da; retain IL/metal catalysts	62–1000 Da (~0.26–1.5 nm)	Tight-UF/loose-NF 1–3 kDa (reject oligomers) → NF 200–500 Da (concentrate BHET/retain IL) → RO/PV/MD to treatment and recover EG/water	Staged size/affinity cuts; solvent recovery with low thermal duty; catalyst loop	PVDF/PES suitable in EG; ceramics robust choice
Methanolysis liquor (MeOH)	Recover: DMT (194 Da), EG (62 Da); Remove: phthalate by-products; retain homogeneous catalysts	62–400 Da	OSN 200–300 Da (catalyst retention/monomer split) → Crystallise DMT → RO/MD for MeOH recycle	MW-based fractionation + solvent loop closure; reduces distillation duty	PI/PES/PIM for OSN; PA swells in MeOH; ceramics/PTFE ideal for MD
Aminolysis/ammonolysis (aq/org)	Recover: BHETA/TPD (~250–300 Da); Remove: amines, salts	60–400 Da; ions <1 nm	NF 300–600 Da for organics → RO for brine/amine treatment	Keep monomeric amides; remove amines/salts *via* size/charge	PA/NF suitable if mild; ceramics best if harsh; pH 2–12
Acid hydrolysis (aq, acid)	TPA (166 Da), EG (62 Da); Remove: acids, colorants	TPA crystals >1 µm	MCr to grow TPA crystals → MF 0.45–1 µm capture → RO for EG/water → optional PV/MD for EG dehydration	Crystal-then-filter; water/EG loop with low energy	PA not suitable for strong acid; PVDF/ceramic preferred; $T <$ 90 °C, pH < 3 tolerant
Alkaline hydrolysis (aq, base)	Disodium terephthalate (Na_2TPA) → TPA (after acidify); EG; remove: NaCl brine	Ions-200 Da; crystals >1 µm	RO for EG/water; MCr for Na_2SO_4 (from neutralisation); MF for TPA crystals post-acidify	Brine minimisation + controlled crystallisation	Ceramics/PVDF for high pH; CA degrades; $T <$ 100 °C; pH > 10
Enzymatic depolymerisation (aq)	Recover: TPA/MHET/BHET; retain enzymes 30–50 kDa	Enzyme ~30–50 kDa (~4–6 nm); products 166–254 Da	UF 10–100 kDa enzyme recycle → NF/RO product/water treatment; MBR option	Retains enzyme; removes MHET/BHET inhibitors; boosts yield	UF PES/PVDF common; antifouling coatings; mild T (30–50 °C), pH 6–8
Pyrolysis oil (org)	Fractionate aromatics (78–300 Da); remove acids/aldehydes	100–300 Da	OSN 300–600 Da (light/heavy split; amine-functional MMMs for acid removal) ± PV for benzene separation	Size/solubility selectivity; lowers hydrotreating duty	Solvent-resistant OSN (PIM/PI); T limit <150 °C
Pyrolysis/gasification off-gas (gas)	Upgrade H_2/CO; remove C_1–C_4, CO_2, H_2S	Kinetic diam. 0.29–0.40 nm	GS (PIM/CMS/zeolite) for H_2/CO tuning; Pd-alloy for H_2; integrate MHE for heat recovery	Raises H_2/CO; strips acid gases; energy integration	High-T materials (PI/CMS/ceramics); Pd needs sulphur guard; tar-tolerant designs
TPA crystallisation mother liquor (aq)	EG, residual TPA/oligomers, colours/salts	<500 Da solutes; ions	NF/RO to recover EG/water → optional MCr for Na_2SO_4 → carbon treatment for colour	Closes loops; reduces purge; value recovery	PA/NF suitable; RO brine management needed
Catalyst loop (glycolysis/methanolysis; aq/org)	Retain ILs/metal acetates/cobalt; pass BHET/DMT/solvents	IL/catalyst often 300–1000$^+$ Da	OSN 500–1000 Da for catalyst retention → PV/MD for solvent recovery	Keeps expensive catalyst active; avoids thermal stress *versus* distillation	Solvent-resistant PI/PES/PIM; ceramics for harsh chemistries

direct study of MHE for PET or plastic incineration is lacking. However, recent studies have explored the efficiency of applying MHEs for heat recovery from flue gas. For example, Yan *et al.* reported a commercial ceramic MHE showing a superior heat recovery rate of 80%, surpassing the conventional stainless steel heat exchangers,[164] which shows the significant potential of this membrane process in energy recovery. A recent study focused on operational parameter optimisation for ceramic membrane heat exchangers and highlighted the effect of liquid boundary layers and heat–mass coupling on

MHE performance.[219] To fully realise the potential of MHEs in plastic recycling, future studies should focus on enhancing membrane materials, improving system design (*e.g.*, ceramic–polymer composite membranes), increasing high-temperature thermal stability, and maximising heat recovery efficiency. These developments can significantly improve the performance and adaptability of MHEs for practical plastic recycling applications.

MCr is a novel separation technique that employs semi-permeable membranes to selectively precipitate solute crystals from a solution.[220] These membranes are commonly fabricated from CA, PVDF, fluoropolymers (such as Teflon AF 2400 and PDMS), MOFs, and 2D materials like graphene.[220,221] In MCr, a solution flows through the membrane, and when the solute concentration surpasses its saturation limit, crystals begin to form on the membrane surface. The membrane acts as a selective barrier that controls mass transfer, allowing solvent molecules to permeate while retaining solute molecules near the surface to initiate controlled nucleation and growth (Fig. 35d). This provides an ideal environment for the formation of high-purity crystals. MCr offers several advantages compared with conventional crystallisation methods including improved product purity, lower energy consumption, and reduced environmental impact. It may find practical applications in PET recycling, such as concentrating sodium sulphate, a by-product formed during pH adjustment of reaction slurries, where it significantly reduces processing costs.[222] Another potentially valuable application is in the crystallisation of TPA from acid precipitation of post-PET hydrolysis solution. Growing TPA crystals to sizes above 1 μm ensures high product purity and facilitates subsequent separation by MF membrane.[223] Through the facilitation of high-purity recovery during the crystallisation stage, MCr can enhance the overall economic feasibility of PET recycling as well as process efficiency. However, advances in membrane materials are still needed to allow finely tuned nucleation and growth kinetics, essential for achieving crystals with a narrow size distribution and elevated purity.[221,224] Developing membranes that enable the exit of large crystals from the membrane surface while retaining the nucleation seeds is a promising strategy to overcome the bottleneck in MCr systems.[224] Continued exploration of these attributes is vital for expanding the applications and efficiency of membrane crystallisation, especially in PET recycling.

Possible advancing membrane performance strategies in PET recycling

Through ongoing research and development, the advancement of membrane technology is pushing the limits of what is possible. Researchers in both scientific and engineering communities are examining novel materials, surface modifications, and manufacturing procedures to improve performance as well as membrane durability through various strategies. These efforts include material enhancement through the use of MMMs and process integration *via* the implementation of HMSs in the pursuit of advancing PET recycling.[43] On the other hand, with the global shift towards a more circular and sustainable trajectory, there is also a growing need to advance membrane performance strategies in PET recycling.[227] These strategies are not only applicable to single-type plastic waste streams but are also highly adaptable to complex mixed plastic waste which often contains polymers with diverse chemical and physical properties.[228,229] By addressing this variability, advanced membranes can greatly improve process flexibility and separation performance.

However, the utilisation of MMMs and implementation of HMSs also present several challenges. The design, optimisation, and integration of these systems require careful engineering and extensive testing to achieve desired performance improvements.[230] Furthermore, the fabrication of MMMs and HMSs can often involve higher costs compared with conventional membranes. In the long term, the benefits of MMMs and HMSs such as increased selectivity, extended operational lifespan, and improved resistance to fouling may outweigh the initial investment concerns and technical barriers. Therefore, continued innovation in membrane materials, fabrication methods, and system design is crucial to overcoming current limitations and making advanced membranes a practical and competitive option for PET and broader plastic recycling applications.

Mixed matrix membranes (MMMs)

As part of the ongoing efforts to enhance membrane functionality in PET recycling processes, MMMs can be considered a promising solution to the limitations of conventional polymeric/ceramic membranes.[156] MMMs combine conventional membranes (Fig. 36a) with a diverse range of nanofillers such as nanoparticles, porous frameworks, and carbon-based materials, into a unified membrane structure (Fig. 36b). Common polymer matrices include PVDF,[231] polyether sulfone (PES),[232] PSF,[233] CA,[234] PAN,[235] and PI,[236] while ceramic matrices often consist of alumina,[237] zirconia,[238] titania,[239] and silicon carbide.[240] Common nanofillers include zeolites,[241] carbon molecular sieves,[242] activated carbons,[243] mesoporous materials,[244] graphene oxide (GO),[245] carbon nanotubes (CNTs),[246] and metal–organic frameworks (MOFs).[247] The integration of these advanced materials enhances membrane performance and properties by leveraging both the individual strengths and synergistic interactions of the components.[248,249] In the recycling of PET, MMMs improve separation by combining free-volume tuning and diffusion-path tortuosity which are determined by the size, shape, and loading of the filler, with specific sorption on the filler and reduced solvent swelling of the polymer matrix. The size-sieving distribution is tightened and the matrix is stiffened by rigid porous domains, such as ZIF-8, UiO-66-NH_2, silica, and GO/CNTs. This preserves the effective cut-off in solvents like MeOH, EG, and NMP, as well as at high temperatures. Aromatic and hydrogen-bonding surfaces facilitate solution-diffusion selectivity for EG, DMT, BHET, MHET, phthalates, and azo dyes, while –COOH/–NH2 groups enable Donnan exclusion of ionic species generated during hydrolysis.

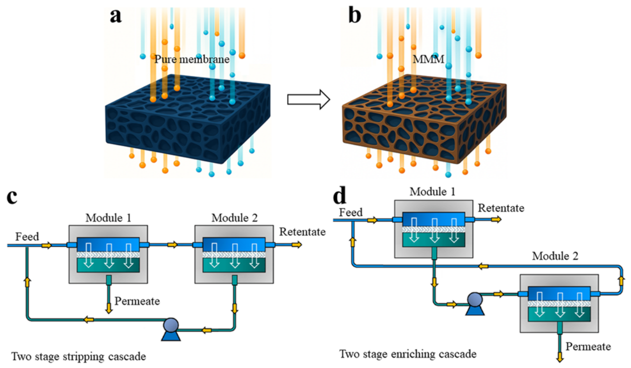

Fig. 36 Strategies to enhance membrane performance in PET recycling through utilisation of MMMs and implementation of HMSs: (a) pure membrane, (b) MMM, (c) two-stage stripping cascade, and (d) two-stage enriching cascade. (Images for a and b): adapted with permission.[273] Copyright 2023, Elsevier; (images for c and d): adapted with permission.[274] Copyright 2019, IWA Publishing.

Robust polymer–filler adhesion by silanes or ligand exchange prevents non-selective voids, so transforming the interphase into a high-resistance, selective pathway.

There are numerous potential advantages associated with using MMMs in the PET recycling process. Primarily, these membranes have the potential to greatly enhance separation efficiency and selectivity. The integration of advanced materials possessing targeted permeation properties facilitates enhanced separation of contaminants, purification of valuable components and waste treatment.[250] Furthermore, the collaborative impact of the polymeric or ceramic matrix and the nanofillers amplifies the overall separation process, resulting in high-quality products associated with PET recycling. Moreover, MMMs effectively tackle various obstacles associated with fouling, chemical degradation, and mechanical stresses. Through surface modification methods or the introduction of antifouling agents such as silver nanoparticles or hydrophilic coatings, MMMs can significantly reduce fouling and extend operational lifetime. The incorporation of robust nanofillers alongside conventional polymer or ceramic membranes also contributes to enhanced mechanical strength and resilience, thereby rendering the membranes more resistant to deterioration and degradation throughout the recycling processes.[251] These enhancements are especially beneficial for the separation and purification of oligomers, where conventional membranes often face performance and stability challenges. For instance, integration of ZIF-8 and GO into MMMs has been shown to increase flux while maintaining high selectivity and improving both mechanical and chemical stability.[252] One additional benefit of MMMs relates to their inherent versatility and adaptability. Researchers are afforded the opportunity to conduct experiments involving diverse combinations of materials, pore sizes, and surface modifications, thereby customising membranes to accommodate the particular composition of feeds and meet the expected result specifications.[253] For example, Dmitrenko *et al.* synthesised a MMM using poly (2,6-dimethyl-1,4-phenylene oxide) (PPO) polymer and GO nanoparticles supported on fluoroplast F42L as a fluoropolymer substrate for PV dehydration of EG (10–30 wt% water). Introducing GO into the PPO matrix resulted in a high permeation flux (180–230 g m^{-2} h^{-1}) along with high selectivity (99.8–99.6 wt% water in the permeate).[156] This example highlights how MMMs can be precisely engineered to meet the needs of specific plastic recycling scenarios such as EG dehydration in PET glycolysis. These chemistries translate directly to performance in key PET streams. In glycolysis liquors, PI/PES OSN MMMs incorporating ZIF-8 or UiO-66-NH_2 exhibit resistance to EG-induced swelling, effectively reject BHET dimers/trimers through a combination of size exclusion and affinity, while retaining bulky IL/cobalt catalysts and allowing BHET monomer and/or EG permeation to reduce thermal duty compared with distillation. In dissolution decolorisation, GO-MMMs with solvent-resistant PI/PES/PIM backbones prevent the passage of aromatic dyes and plasticizers, thereby minimising organic fouling. In EG dehydration and solvent recovery, lamellar GO or zeolitic fillers within hydrophilic matrices enhance water absorption and diffusivity while inhibiting EG, aligning with the PPO/GO pervaporation results. In pyrolysis oil treatment, amine-functional silica/MOF MMMs embedded in organophilic matrices complex with acidic species and stabilise the cut-off in aromatic-rich feeds. Hydrophilic or zwitterionic surfaces decrease the adsorption of TPA crystals, oligomers, and dyes; fluorinated low-energy surfaces inhibit the adhesion of viscous organics; and a negative zeta potential repels terephthalate at neutral to alkaline pH, hence maintaining flow and rejection in hydrolysis loops.

However, achieving these advantages depends heavily on the compatibility and dispersion of the nanofillers within the matrix. Weak interfacial adhesion or poor dispersion can cause a significant decrease in MMMs' performance.[248,254,255] This highlights the careful selection of MMMs' components based on compatibility and dispersion along with target species' properties, operating conditions, suitable separation performance, and long-term stability requirements.[256] Furthermore, long-term performance evaluation under real operating conditions is crucial to assess fouling resistance and maintenance requirements.[257] These factors continue to present substantial challenges for MMM application in PET recycling. Accordingly, implementing strategies that improve the adhesion between polymers and fillers (*e.g.*, ligand-exchange anchoring or silane coupling) is crucial to eliminate interfacial voids and transform the interphase into a selective, high-resistance path that can withstand high temperatures in EG, MeOH, and NMP exposure. To address these challenges, future research should focus on deepening the understanding of structure–property relationships and systematically optimising material combinations to ensure stable, high-performance operation tailored to the specific demands of PET recycling processes.

Beyond single-type plastic recycling, MMMs hold significant promise for processing mixed plastic waste streams due to their tunable chemical functionality and pore structure which allow selective interactions with specific polymer components. This customised selectivity enhances separation performance in complex waste mixtures, addressing the limitations of traditional membranes in achieving high purity levels.[258,259] For example, in a plastic waste mixture containing both hydrophobic and hydrophilic polymers, MMMs can be engineered to selectively interact with one component over the other and enable more precise fractionation.[255] These selective interactions not only enhance the purity of recovered materials but also improve the overall efficiency of the recycling process. Therefore, MMMs are possible solutions in the management of mixed plastic waste with the potential to enhance sustainability outcomes and offer performance flexibility.

Hybrid membrane systems (HMSs)

Another promising strategy for enhancing membrane functionality in PET recycling processes is the implementation of HMSs. The primary objective of HMSs is to leverage the distinct advantages resulting from two or more different membrane processes with distinct separation mechanisms into a single, integrated system.[260] The implementation of this combination creates a multifunctional system that effectively tackles complex separation or purification applications as well as creates more adaptability to varying stream compositions to achieve higher overall efficiency (Fig. 36c and d).[260–262] The HMS's performance is significantly affected by the physicochemical properties of the chosen membranes. The choice of materials usually corresponds to these physicochemical properties and specific processes being integrated and their associated separation mechanisms.[263] Similar to MMMs, the selection process must account for target species properties, operational conditions, mechanical stability, and desired separation efficiency.[256] HMSs in PET recycling operate by sequencing complementary transport mechanisms, ensuring that each step conditions the next one and enhances selectivity under real feed conditions. During dissolution and glycolysis, MF or UF initially removes labels, fines, TPA crystals, and oligomers to reduce organic fouling and stabilise the OSN cut-off in NMP or EG. The subsequent OSN process then conducts affinity-aided molecular-weight fractionation by rejecting BHET dimers and trimers as well as dyes and plasticisers while allowing the passage of BHET monomers and/or EG or solvent, which extends cycle length and facilitates closed solvent loops. In enzymolysis, a UF enzyme loop within the 10–100 kDa range keeps PETase/cutinase at 30–50 kDa while constantly removing MHET, BHET, and TPA to mitigate product inhibition and enhance hydrolysis, with downstream NF/RO to meet water and EG reuse or discharge standards. In methanolysis and glycolysis, the combination of OSN for homogeneous-catalyst retention with MD or PV for methanol or EG recovery maintains catalyst activity and eliminates reboiler duty in contrast to distillation. In thermolysis and gasification, H_2-selective gas separation using CMS/PIM/zeolite or Pd-alloy membranes enhances syngas, while a MHE recovers sensible heat from off-gas or char to heat feeds or power MD/PV, therefore diminishing site energy intensity.

The combination of multiple membrane processes within a hybrid system can present numerous benefits in the context of PET recycling. Firstly, it enables the simultaneous benefit of complementary separation mechanisms. For example, a particular membrane process may demonstrate superior size-based separation, whereas another process may exhibit greater adsorptive separation.[264] By combining these processes, the hybrid system may exhibit enhanced separation efficiency and selectivity relative to the utilisation of a solitary membrane. This improvement is particularly beneficial for PET recycling applications, such as oligomer separation, leading to enhanced fractionation and higher product purity. In addition, HMSs offer increased adaptability in customising varying compositions. Indeed, hybrid membranes facilitate the customisation of systems to address the specific challenges presented by various feed sources.[153,265] Hence, as the streams of PET recycling may exhibit variability with regard to the presence of contaminants, distribution of molecular weights, and various other characteristics, the ability to adapt enables the achievement of consistent performance across a wide range of compositions, thereby enhancing the overall reliability of the process. Moreover, HMSs have the capability to effectively address fouling and membrane long-term stability concerns as well. Fouling is a prevalent issue encountered in membrane-based processes. This phenomenon has the potential to diminish the overall efficiency and effectiveness of such processes as time progresses. Hybrid systems can mitigate fouling rates or mitigate the severity of fouling by leveraging the synergistic effects of various separation mechanisms.[155,266] This leads to an extended lifespan of the membrane and a reduction in energy and maintenance demands of downstream operations such as waste treatment.[267] Across these systems, practical design rules establish a connection between chemistry and metrics. These rules include matching the cut-off to the EG–BHET and MeOH–DMT gaps and utilising π–π, H-bonding, and acid–base interactions to increase marginal molecular-weight differences; selecting solvent-resistant matrices, such as ceramics or PI/PES/PIM, or crosslink PA to reduce the swelling of MeOH/EG/NMP; utilizing the charge to achieve Donnan exclusion of terephthalate and IL/catalyst anions; applying zwitterionic or fluorinated antifouling coatings and removing foulants up front with MF/UF; and closing loops with OSN/MD/PV for solvent, UF for enzyme, and MCr plus MF for TPA crystals.

For instance, Cassano *et al.* addressed the issue of fouling by proposing a HMS that combines UF and NF processes. The main objective of this new approach was to reduce fouling by simultaneously tackling the accumulation of organic and inorganic scale within the system. Additionally, this approach has the opportunity to eliminate contaminants or enhance the purification of valuable components with greater potential. This approach exhibits the potential to improve the effectiveness and durability of the HMS. This is supported by the observed decrease in fouling, which was attributed to the removal of suspended solids and fat substances by the UF process. Furthermore, the NF process successfully recovered and concentrated chromium, resulting in a final concentration of approximately 10 g L^{-1}.[268] Moreover, Cai *et al.* performed a study to assess the effectiveness of a HMS in filtering MPs in a full-scale wastewater treatment plant. The HMS consisted of an MBR, UF as a pre-treatment for RO, and the RO process itself. MP concentrations decreased significantly, with a reduction of 93.2% after MBR treatment and a decrease to 98.0% after RO treatment. The results highlight the advantages of incorporating HMSs into coastal wastewater treatment plants, as they successfully mitigate MP pollution in marine ecosystems while simultaneously recycling wastewater into reusable water resources. This dual capability helps reduce ocean pollution and tackle water shortage concerns.[155] As highlighted in these studies, HMSs not only improve the overall performance of membrane systems but also contribute to the environmental sustainability of various processes such as PET recycling. However, achieving these advantages in the context of plastic recycling faces some challenges. The integration of multiple membrane processes in HMSs introduces engineering complexities that can result in high initial investment and operational costs. Furthermore, the performance that has been demonstrated in laboratory-scale studies may not be directly applicable to pilot or industrial-scale applications, particularly when applied to the diverse and variable feedstocks that are standard in PET recycling. Therefore, future research should focus on systematic approaches for optimising HMS integration specifically within PET recycling systems to address these limitations. This will ensure that the HMSs provide scalable, efficient, and consistent performance in PET recycling environments.

In addition to PET recycling, HMSs offer a strategic solution for managing the complexities of mixed plastic waste. HMSs can leverage a variety of membrane processes, each optimised to separate specific contaminants or polymer types. These membranes can be customised and selected based on the required separation performance, and the characteristics of the mixed polymer waste.[263,269] For example, MF can remove large contaminants, while NF can selectively separate components based on differences in molecular size, charge, or solubility.[270,271] This hybrid configuration enhances both the efficiency and effectiveness of the recycling process.[272] Therefore, by optimising the configuration and combination of these membranes, HMSs can significantly improve the recovery rates and purity of recycled materials. As such, HMSs represent a powerful tool for advancing sustainable and scalable management of mixed plastic waste.

Challenges and limitations

The emergence of membrane technology in PET recycling has shown promise for enhancing the process's efficiency and sustainability. However, that promise is constrained by a set of difficulties that must be overcome before membranes can be implemented at scale to deliver consistent performance. A central difficulty results from three interconnected technical limitations: feed heterogeneity, fouling/wetting, and materials stability. Post-consumer streams exhibit significant variability in grade, colourants, labels, adhesives, fillers, and legacy additives. As a result, systems designed for homogeneous streams tend to underperform with real mixtures, resulting in fluctuating selectivity and inconsistent quality of recycled products. This variability also intensifies fouling caused by oligomers, plasticisers, dyes, fines, salts, biofilms, and TPA crystals, which leads to flux decline and pore wetting that compromises separation. Simultaneously, aggressive chemicals (*e.g.*, methanol, EG, NMP, amines, acids/bases) and high temperatures can swell or hydrolyse membranes and seals, thus shifting the effective cut-off and shortening membrane lifetime. These considerations create the primary performance trade-off: tighter cut-offs are required to separate monomers from oligomers or maintain homogeneous catalysts. However, tighter membranes reduce permeation and are more sensitive to swelling and temperature variations. Practical options to overcome these limitations include robust MF/UF pre-treatment and in-line analytics for feed stabilisation, solvent-resistant materials, fouling-resistant surfaces, hybrid systems that stage conditioning and recovery (*e.g.*, MF/UF → OSN or NF/RO), and targeted solvent/monomer recovery *via* PV or MD. Finally, their effectiveness must be verified on real depolymerisates rather than synthetic laboratory feeds.

In addition, implementation is further limited by scale-up, residual management, and the quality of decision evidence for investment. Modules that operate efficiently in laboratory cells may not perform as well in plants due to maldistribution, non-ideal hydrodynamics, and maintenance logistics. Moreover, without simple monitoring procedures and soft sensors, fouling can escalate into unplanned downtime. Membrane processes also concentrate contaminants and hence produce brines, neutralisation salts (*e.g.*, Na_2SO_4), and catalyst-rich residues. These cause costly streams with regulatory implications unless paired with recovery methods such as MCr for salts and closed-loop solvent/catalyst recycling. Finally, techno-economic analyses (TEA) and life-cycle assessments (LCA) often lack comparability due to omitting membrane replacement, cleaning agents, off-spec penalties, or using inconsistent system boundaries, while the field still lacks standardised protocols for feeds, modules, and key performance indicators. A viable path to scale couples modular skids and energy integration (*e.g.*, MHX) with harmonised TEA/LCA frameworks, routine reporting of flux, rejection, recovery, cleaning energy, and module lifetime, alongside open pilot datasets on real materials recovery facility feeds. Standardised challenge cocktails, defined test geometries, and periodic marker–solute calibration enable direct comparisons and accelerate the development of deployable membrane systems for PET recycling.

Outlook and future opportunities

The outlook for membrane technology in PET recycling is promising, offering substantial opportunities to significantly improve the plastics recycling industry's sustainability. As membrane technology advances, it will become more efficient and cost-effective, making it a more financially feasible option for PET recycling and potentially a game-changer for the recycling industry as a whole. Membrane technology is already in minor use at a number of PET recycling plants around the world, and its widespread implementation is anticipated as more and more businesses see the value in this technology. Although membrane technology has been successfully applied to some limited sections of PET recycling processes, there are still challenges and limitations to overcome and areas for developmental improvement to unlock the full potential of membrane technology. For instance, researchers are investigating the possibility of creating novel membrane materials and coatings that are more resistant to fouling. Moreover, efforts are being made to develop more durable membrane materials that can withstand conditions such as harsh chemicals, and high temperatures and pressures. Researchers are also working to develop membrane processes that use less energy and are exploring the incorporation of solar power and other renewable energy sources. Furthermore, researchers are exploring a range of strategies to advance membrane performance in PET recycling, focusing on materials and engineering aspects. One potentially effective approach is the use of MMMs to improve materials characteristics. Another promising avenue is the implementation of HMSs to improve the overall efficiency of membrane processes. Finally, predictive models are currently being developed by researchers to aid in scaling up. Continuous investment and research in membrane technology for plastic recycling in the future can significantly contribute to the reduction of plastic pollution and the promotion of a circular economy. This has the potential to break down diverse plastic mixtures, separate contaminants, purify valuable components and waste treatment, and eventually revolutionise the field of plastic recycling towards a more environmentally friendly future.

Conclusions

Membrane technology has the potential to transform the plastic recycling industry by directly addressing the challenges of recycling complex plastic streams like PET, which have not been fully explored in the existing literature. PET is a commonly used plastic material, known for its outstanding durability and versatility. Therefore, PET recycling is an integral component of sustainable plastic management, with the primary objective of mitigating environmental consequences *via* reuse of PET materials. The integration of membrane technology into PET recycling processes has opened new avenues for the recycling industry to improve its effectiveness and sustainability. There is optimism that membrane technologies can enhance purification and separation in the PET recycling process, resulting in higher-quality value-adding products suitable for a variety of applications. Moreover, membrane technology has the potential to reduce the amount of energy and resources needed for the recycling process, making it an economically and environmentally feasible option. Of course, membrane technology could also be exploited in a similar capacity to enhance recycling of other plastics. Anticipated advances in membrane technology for plastic recycling represent a significant step toward a more circular economy as the need for environmentally friendly goods grows.

Conflicts of interest

There are no conflicts to declare.

Data availability

This article does not report any primary research data. Consequently, no datasets, software, or code are associated with the work.

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