

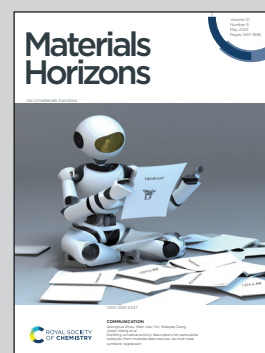


This review was conducted by the research teams of Tomonori Saito and Chelsea Chen at the Chemical Sciences Division in Oak Ridge National Laboratory, Oak Ridge, Tennessee, USA.

Recent development of end-of-life strategies for plastic in industry and academia: bridging their gap for future deployment

This review serves as a guide to understanding the gap between industry and academia in plastic recycling and creates a path for new discoveries in academic research to be integrated into industrial practices.

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## MINIREVIEW

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## Recent development of end-of-life strategies for plastic in industry and academia: bridging their gap for future deployment†

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Plastics have advanced society as a lightweight, inexpensive material of choice, and consequently over 400 million metric tons of plastics are produced each year. The difficulty with their reuse, due to varying chemical structures and properties, is leading to one of the major global challenges of the 21st century—plastic waste management. While mechanical recycling has been proven successful for certain types of plastic waste, most of these technologies can only recycle single types of plastics at a time. Since most recycling collection streams today have a mixture of different plastic types, additional sorting is required before the plastic waste can be processed by recyclers. To combat this problem, academics have devoted their efforts to developing technologies such as selective deconstruction catalysts or compatibilizer for commodity plastics and new types of upcycled plastics. In this review, the strengths and challenges of current commercial recycling processes are discussed, followed by examples of the advancement in academic research. Bridging a gap to integrate new recycling materials and processes into current industrial practices will improve commercial recycling and plastic waste management, as well as create new economies. Furthermore, establishing closed-loop circularity of plastics by the combined efforts of academia and industry will contribute toward establishing a net zero carbon society by significant reduction of carbon and energy footprints. This review serves as a guide to understand the gap and help to create a path for new discovery in academic research to be integrated into industrial practices.

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‡ Equal contribution.



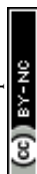
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# 1. Introduction

The volume of yearly plastic production has exceeded 400 million metric tons (Mt)<sup>1</sup> and had an estimated value of \$593 billion in 2021.<sup>2</sup> This demand will continue to grow at a projected rate of 3.7% for the next decade. Plastics are a critical material for the advancement of society, but improper end-of-life management of plastic-based materials is also detrimental to society. Only ~9% of global plastics are recycled today,<sup>3</sup> with the rest being incinerated for energy, ending up in landfills, or even worse, littered into the environment, polluting our lands and oceans. While recycling technologies to process single component plastic waste streams already exist on a commercial scale, plastics currently account for over 20% of the waste in landfills due to the lack of technology to sort and separate mixed plastic streams because of size limitations as well as mixed materials that cannot be physically separated.

Most plastics today are made from monomers that are synthesized from fossil fuels. Small molecules extracted from crude oil are chemically modified *via* multi-step reactions to form monomers, which can then be polymerized into polymers of various structures and chemistries. The processes used to make monomers and then polymers are associated with high energy consumption, an estimated 3.2 quadrillion Btus per year, and 104 MMTCO<sub>2</sub> equivalent greenhouse gas (GHG) emissions in the U.S. alone.<sup>4</sup> The most common commercially available polymers include polypropylene (PP), polyethylene (PE), polyethylene terephthalate (PET), polyurethanes (PU), and polystyrenes (PS), with global production volumes of 68, 166, 33, 27, and 25 Mt, respectively.<sup>5</sup> Among them, PE, PP and PET represent the most recycled plastics, which occupy 95.2% of recycled materials (Fig. 1). Since the chemistry of each polymer is different, the physical and mechanical properties are also different, thereby requiring different processing conditions to recycle each type of plastic.

Mechanical recycling is the most commercially used recycling method, but has its limitations due to its sorting challenges, decreased physical properties, and the need for additives to allow recycled materials to be used again. As such, chemical recycling is being developed as a complementary method to mechanical recycling. One of the most prevalent techniques currently used to chemically recycle polymers is pyrolysis, where plastics are heated to high temperatures to yield chemicals that would otherwise be extracted from fossil fuels. Academically, there is a large effort toward chemical recycling with advanced techniques such as pyrolysis, depolymerization, and biobased plastics, on which large quantities of publications are seen each year.

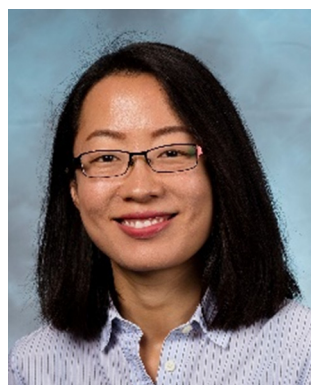
In this short review, the current efforts and advantages in industrial recycling processes will be discussed as well as their unmet needs. Then, recent academic efforts relating to novel



**Xiaomin Tang**

*as well as energy storage and conversion.*

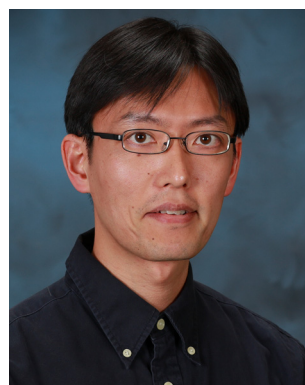
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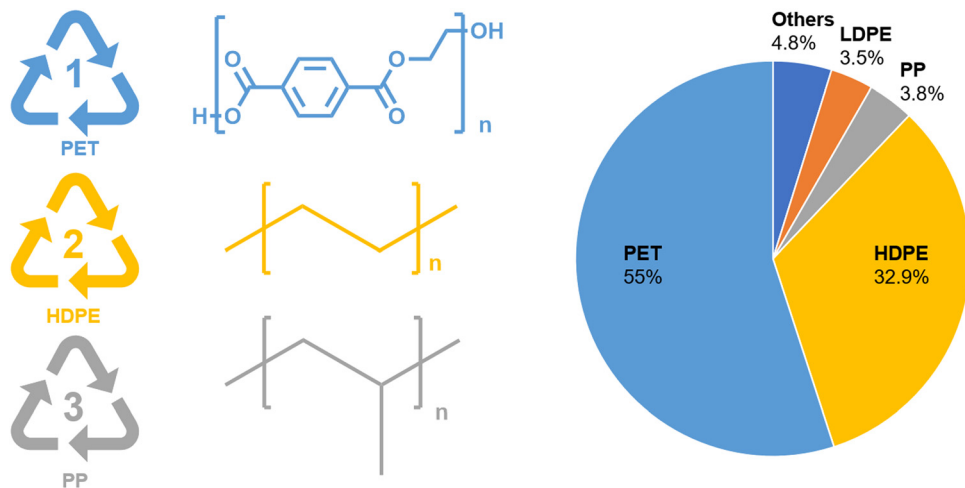


Fig. 1 Polyethylene terephthalate (PET), high density polyethylene (HDPE), polypropylene (PP), and low density polypropylene (LDPE) are some of the most commercially recycled polymers today with PET and HDPE making up the majority of the plastic recycling market.<sup>6</sup> Adapted in part from theroundup.org.

recycling methods as well as the design of recyclable materials will be featured. This review will primarily focus on recycling efforts for PET and polyolefins such as PP and PE, since those commodity polymers are some of the most used and recycled consumer plastics that will allow for comparison between industry and academia. There is a gap between what industry needs and what academia offers. This review will help academics understand the current major challenges that industrial recycling processes have as well as highlight technologies and research that have been developed in academic laboratories that can have strong sustainability and economic impacts.

## 2. Industry efforts

### 2.1 Sorting of waste feedstock

Curbside recycling in the U.S. generally comes in two forms: single-stream and dual-stream (Fig. 2). In single stream recycling, paper-based waste and old corrugated cardboard are combined with glass, metal, and plastic containers. To extract the plastic waste, materials are processed at materials recovery facilities (MRFs). Plastic only makes up a small portion of single-stream feedstocks and must be separated from the single stream feedstock before plastic recycling can occur. On the other hand, dual stream recycling is collected as two different feedstocks, where one has paper-based recyclables and another includes glasses, metals, and plastics. Dual-stream recycling yields a higher percentage of plastics but requires effort from consumers to separate their recyclables as well as the extra collection cost of having specialized types of collection trucks for each type of recyclable.

Regardless of single or dual stream, plastics must be further separated out at MRFs before mechanical recycling can be done. Sorting is conducted manually or *via* a machine/AI driven separation process, where an AI can optically detect the type of plastic by shape of the object or using near infrared (NIR)

detectors that are able to detect the types of chemical bonds in a polymer to determine the type of plastic. In a modernized system, blasts of air are used to move the plastics to positive and negative sorting lines, where the blast of air can push the plastic into another line or allow it to drop down and pass to a different line. There are also optical systems being developed by companies in which robotic arms are incorporated that can quickly remove contamination along the lines.

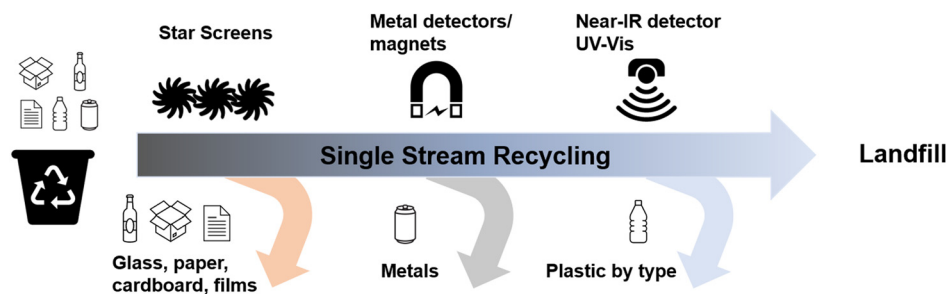
While sorting for mechanical processes continues to be improved, there are still various challenges that make certain plastics extremely hard to recycle. The shape and the size of plastic containers can be a hindrance for the sortation process. For example, the shape of clamshell takeout containers (*i.e.*, containers that might be used for salad or other take out) make it hard for the machine to separate them *via* blasts of air due to the shape of the material and air resistance, causing the material to fly in unexpected trajectories. Additionally, even though the plastic type for clamshells is classified as *e.g.*, PET or similar analogues, they sometimes have slightly different chemical structures giving an altered glass transition temperature ( $T_g$ ) and melting point that shifts their optimal processing temperature. These types of containers frequently have adhesives and labels on their surface, which also causes issues during mechanical recycling.

The color of an object can also cause issues during sortation. Since NIR relies on the reflection of light off the material into detectors, plastic objects with additives like carbon black are hard for NIR detectors to pick up. Additionally, if the color of the object blends in with the conveyor belt it's on, optical systems can have trouble detecting the material too. Another color related challenge is that some MRFs use the color and shape of known objects to detect the type of material. If a certain type of consumer good comes in a specific color and shape, MRFs will use that information in their optical sorting system. However, if the manufacturer were to change the material from PP to PET, machines will continue to sort the





## Single-Stream Collection



## Dual-Stream Collection

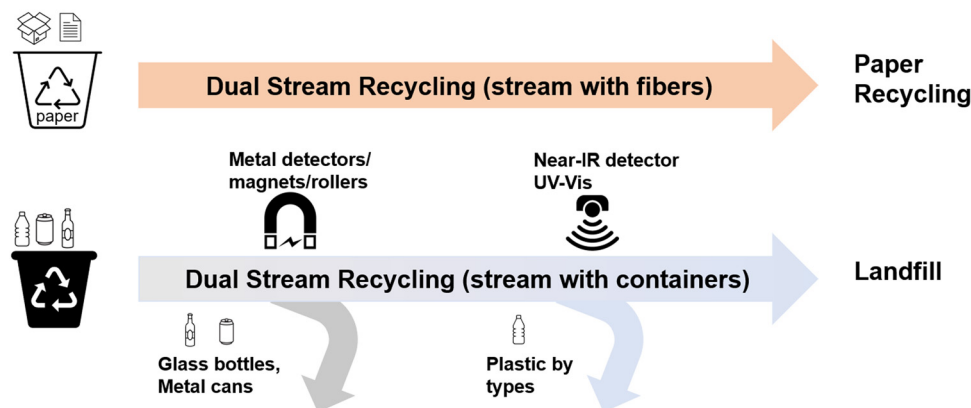


Fig. 2 Single stream vs. dual stream recycling feedstock, where the single stream contains all recyclable materials while the dual stream separates materials such as paper and cardboard from glass, metals, and plastics.

consumer product as if it were PP, causing a contamination of their bale of sorted plastics. Contamination causes the quality and value of the bale to drop, which can even result in the entire bale being rejected by buyers. Following the separation of plastic waste by polymer type, they can be sold to downstream recyclers.

### 2.2 PET recycling

PET is a versatile plastic that has a wide range of applications from food containers to thermoforms for manufacturing and even textiles. However, due to the durability of PET, it poses a significant long-term persistence problem in the environment if not disposed of and recycled properly. Only 14% of consumer PET is recovered for recycling after being used, and just 2% is recycled appropriately and put back into circulation.<sup>7</sup> PET is commercially recycled *via* both mechanical and chemical recycling.

**2.2.1 Mechanical recycling.** The majority of recycled PET is sourced from plastic bottles and containers. Mechanical recyclers generally work with bales of plastics that were sorted out by MRFs from municipal waste. While these PET bales are comprised mostly of bottles, trays, cups, and lids, the bales are sorted once again after arriving at the mechanical recycler by NIR as well as UV-Vis detectors to sort plastics by color. The sorted PET is then chopped up into flakes and put through a

wash process, which can include a caustic treatment with base-containing detergents to further clean the materials. The washed flakes are then dried and put through another round of color sorting and a large magnet to remove any remaining metal waste that might have made it through the process. The clean and color sorted flakes are then heated into the melt state and extruded as pellets which can then be sold to downstream manufacturers.

Prior to 2016, a large amount of plastic waste in the U.S. was sold to China. But when China's National Sword program was announced, U.S. companies had to start recycling more PET domestically. Companies such as DAK Americas, a subsidiary of Alpek, and Indorama Ventures are some of the larger mechanical recycling companies in the U.S. that process PET. According to the 2018 annual report, polyesters account for 74% of Alpek's (DAK's parent company) sales. Alpek's capacity for PET production is 2.46 Mt while recycled PET (rPET) only has a capacity of 70 kt.<sup>8</sup> Indorama Venture Limited (IVL) also recycles PET *via* a mechanical process, however they also have a subsidiary to their business, where PET flakes are melted and spun into recycled fibers and yarn under the brand DEJA. 317 kt of PET was recycled by Indorama in 2021<sup>9</sup> into the form of resins, fibers, and flakes.

**2.2.2 Chemical recycling.** Chemical recycling utilizes the process of breaking down polymers into their component parts



that can be reused in the production of new plastics, as well as fuels and other useful products. Chemical recycling has several advantages over other types of recycling methods. A significant benefit of chemical recycling is that it can be used to process plastic waste that is currently categorized as non-recyclable or difficult to recycle, thereby recovering a massive amount of recyclable PET resin that would otherwise be destined for landfill or incineration. This includes colored bottles, black food service items such as utensils, food packaging such as clamshell containers, carpeting, fabrics, and more.

**2.2.2.1 Pyrolysis.** Pyrolysis is a method of thermally decomposing polymers into small molecules. By applying high temperature and pressure to a polymer in the presence of a catalyst and the absence of oxygen, the polymer structure can be broken down into lower molecular weight components. The pyrolysis of PET often entails the random scission of the polymer into chemicals of varying sizes. However, the high temperature will cause a rearrangement *via* a hydrogen bond transfer and finally a decarboxylation, resulting in the production of CO<sub>2</sub>, which can cause an increase of emissions as well as interference with the process. As such, PET is not a typical polymer target for pyrolysis.<sup>10</sup>

**2.2.2.2 Depolymerization.** Solvolysis, also known as chemical depolymerization, is a technique that involves the use of chemicals in order to break the polymer down into its constituent monomers. In comparison to pyrolysis, PET deconstruction can take place at temperatures lower than pyrolysis, since it involves the breaking of hetero-atom bonds in the polymer backbone. There are various approaches that can be taken in order to solvolyze PET (Fig. 3), and the end products of this deconstruction vary greatly depending on which approach is used.

Glycolysis is the oldest technique used to depolymerize PET. Glycolysis of PET by catalytic transesterification occurs with an overabundance of a glycol at temperatures between 110 and 270 °C.<sup>11</sup> The glycolysis of PET with ethylene glycol (EG) produces bis(2-hydroxyethyl) terephthalate (BHET) (Fig. 3). BHET is a favorable product for PET synthesis as it is an intermediate in the reaction of terephthalic acid (TA) with EG. There are several

Table 1 Glycolysis and methanolysis of PET in industry

Company name	Country	Established	Capacity (kt a <sup>-1</sup> )
Eastman Chemical Co	USA	2022	100
CuRE Technology	Netherlands	2020	25
IFP Energies nouvelles	France	2020	22
Ioniqa Technologies	Netherlands	2019	10

Table 2 Hydrolysis of PET in industry

Company name	Country	Established	Capacity (kt a <sup>-1</sup> )
Carbios	France	2011	50–100
Gr3n	Switzerland	2021	30

companies globally that currently have glycolysis plants to produce BHET or methanolysis plants to produce dimethyl terephthalate (DMT) (Table 1).

PET can also be industrially depolymerized by the process of hydrolysis, which can be done in an acidic, alkaline, or even neutral aqueous environment, where water is used to cleave the ester bond in the backbone of the polymer. The process generally takes place at high temperatures and pressures and yields a final product of TA and EG. Some companies that depolymerize PET *via* hydrolysis are listed in Table 2.

## 2.3 Polyolefins

**2.3.1 Mechanical recycling.** PP and PE, both LDPE and HDPE, are the most commonly collected and recycled polyolefins. In a bale of waste plastic, PP and HDPE can be separated from each other and from other plastics like PET by density. The density of PP is generally between 0.90–0.92 g cm<sup>-3</sup><sup>12</sup> while the density of HDPE is 0.94–0.96 g cm<sup>-3</sup>. LDPE, on the other hand, is slightly harder to separate as the density falls between 0.91–0.93 g cm<sup>-3</sup>. PET has a density of 1.38 g cm<sup>-3</sup> which allows for the separation of polyolefin and PET flakes by submerging the plastics into water and using a grated moving belt to remove the PET flakes that sink in the water (Fig. 4).

Following the sortation of the different types of plastics, the different types of plastics are washed and dried before heating and extruding as recycled pellets. Recycled pellets can then be used by downstream manufacturers to make consumer goods to be resold. While this is generally the case for plastic recycling, certain types of plastic waste have to be processed differently. Plastic bags, generally made from LDPE, can sometimes cause issues due to the nature of bags. As plastic bags get shredded by rotating machinery, it is possible for a bag to wrap around the rotating parts, resulting in the processing lines being turned off to deal with the caught bags.

One of the largest mechanical recyclers of HDPE and PP in the U.S. is KW Plastics which has a processing volume of over 450 kt of plastics per year.<sup>13</sup> Additionally, some of the recycled plastics that KW Plastics produces are FDA approved food grade PP. Natura PCR is one of the largest processors of plastic films and shrink wraps in the world and has a goal to reach an annual processing capacity of 180 kt of plastic film within the

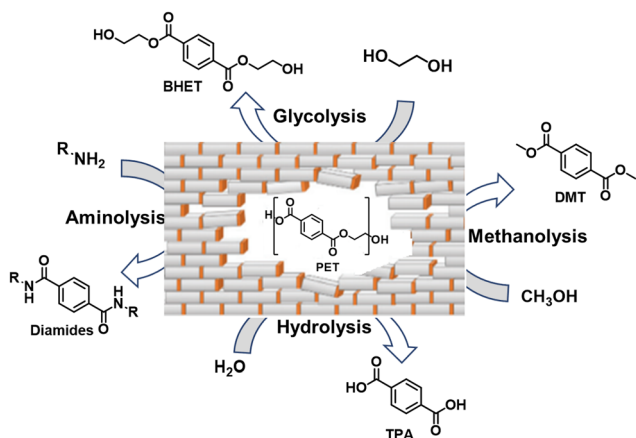


Fig. 3 PET can be deconstructed using various techniques to obtain different monomers with various functional groups as end products.





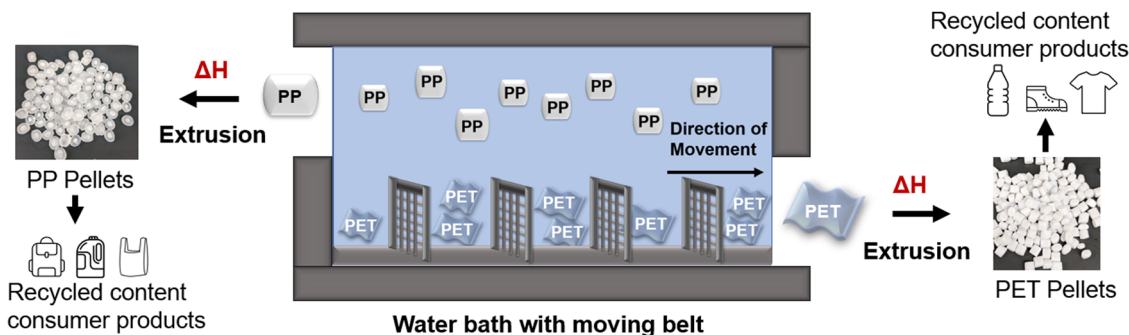


Fig. 4 Difference in density can be used to sort plastic types (PET vs. PP) in a water bath. Once the plastics have been sorted by type, they can be processed separately and then heated and extruded into pellets to be made into recycled content containing consumer products.

next five years.<sup>14</sup> WM Inc. reported a processing volume of 13.6 Mt for combined PP, PE, and PET waste.<sup>15</sup>

**2.3.2 Purification/solvent extraction.** Mechanically recycled plastic resins generally have lower mechanical properties compared to virgin plastics, which either get downcycled into other products that can tolerate the decreased mechanical strength or get blended with virgin plastics to have acceptable mechanical properties. PureCycle is a company that processes isotactic polypropylene (iPP) via a solvent-based purification method that uses high temperature and pressure.<sup>16</sup> The process takes in a contaminated feedstock of waste plastics and is able to produce high purity, colorless, and even low odor resins by selectively dissolving polymers and removing volatile organic compounds in a multistep extraction. The extracted polymers exhibit virgin-grade properties without degradation of mechanical properties. The processing volume of PureCycle's Ironton, Ohio facility is expected to reach over 45.8 kt per year and will be expecting to increase their capability with additional plants in Georgia, USA and South Korea.<sup>17</sup>

**2.3.3 Thermal catalytic process/pyrolysis.** Pyrolysis and gasification are used industrially to transform plastic waste into petrochemicals and refined hydrocarbons. The pyrolysis reaction is conducted in a low oxygen environment to ensure that the polymer pyrolyzes instead of burning. Since the reaction must be performed in the absence of oxygen, polymers that contain oxygen in their structures are generally not favorable for pyrolysis processes. During the process, polymers, generally with hydrocarbon backbones, are heated up to temperatures above 400 °C in order to cleave the various bonds in the backbone to produce syngas,

short hydrocarbons, and other molecules such as benzene, toluene, and xylene. The shorter hydrocarbons can then be used as fuels or used to make alternative polymers. The products of pyrolysis can also be used to produce energy, oil as a replacement for fuels, and syngas for the chemical industry.

Companies involved in pyrolysis are proliferating at a rapid rate, particularly those involved in the production of fuel or syngas. Some innovations that have recently been developed for pyrolysis include more efficient heating of the waste plastic feedstock using supercritical steam by Mura Technology. Since steam can penetrate the bulk material faster than heat transfer occurs through the material, the processing time can be shortened, saving both time and money. The following selection of companies highlights a few of the most recent developments, with a focus on the recycling of plastics into building blocks, monomers, or naphtha and is listed in Table 3.

One challenge that the pyrolysis industry faces is the concern that the process seems very much like burning plastic for energy or fuel. Since a plastic waste feedstock is heated to high temperature and an oil is produced, there is a public misconception that plastic is being burned for fuel in the process. In reality, the products are useful molecules used to make other industrially relevant chemicals. As such, there has been a lot of pushback by environmentalists as well as policymakers, where some states do not recognize pyrolysis as a recycling process and is classified as a manufacturing process instead. Additionally, pyrolysis also tends to generate various byproducts such as waxes, crude oil, syngas, or even fuel, as well as hydrocarbons that get cleaved too short to be useful.

Table 3 Companies that are currently employing pyrolysis processes

Company	Country	Feedstock	Major products
Agilyx	USA	Mixed plastics	Synthetic crude oil
Mura Technology	UK	Flexible, multi-layered materials	Crude oil
BASF (ChemCycling)	Germany	Mixed plastic	Synthetic crude oil
LyondellBasell	Netherlands	PP, PE	Oxyfuels
Brightmark LLC	USA	Mixed plastics	Synthetic crude oil
Nexus Circular LLC	USA	HDPE, LDPE, PP, PS	Synthetic crude oil
Plastic Energy Ltd	UK	Mixed plastics	Synthetic crude oil
New Hope Energy	USA	Mixed plastics	Synthetic crude oil
OMV	Austria	Mixed plastics	Synthetic crude oil
Indaver NV	Belgium	Polyolefins	Waxes



Pyrolysis is a technology that is still being developed; there is a lot of research in both academia and industry to develop catalysts that will enable more controlled length of polymer chain cleavage, and catalysts that will allow pyrolysis to be done at lower temperatures or even shorter reaction times. Overall, pyrolysis has potential and high research activities can be seen in many petrochemical companies such as ExxonMobil and Shell investing in the development of their own pyrolysis plants.

## 2.4 Mixed plastics

**2.4.1 Mixed plastics feedstock.** One of the biggest challenges in recycling today are objects that are made out of several types of plastics. Frequently, various types of plastics are combined in a way that is extremely hard to mechanically separate, resulting in the whole object being thrown away, even though the individual component could have been easily recycled. Waste plastics such as multilayer packaging are a prime example of mixed plastics and can be found as packaging for things such as chips, cookies, and many other consumer items. Generally, each layer of the packaging has a different purpose ranging from acting as a barrier layer for moisture, oxygen, and even sunlight, while other layers provide stronger integrity, adhesion, and durability. A simple bag for chips commonly contains over 4–5 different layers of plastics,<sup>18</sup> each with different physical properties, as well as layers of adhesives to hold everything together. Since each component has a different  $T_g$  and melting point, it is virtually impossible for mechanical recyclers to process these plastic wastes.

In addition to multilayer packaging, blended materials are also a challenge. Consumer waste such as carpets and textile are often blended or woven together. They are generally not collected by recyclers due to the inability to process the materials as a whole. There are some carpet collectors that will collect nylon carpets due to the availability of feasible processes to separate nylon fibers, which can be spun into recycled nylon fibers. However, the same does not apply to PET carpets, and any PET carpets that have been recycled need to be further processed to separate the PET face fibers from the PP backing. Textiles are very rarely recycled, and instead get reused or downcycled into other products. As such, there is virtually no established process for consumer wastes such as textiles and PET carpets in the current recycling industry.

## 3. Academic efforts

Chemical upcycling of polymers has the promise of altering the paradigm for discarded plastic from waste to a valued resource. Significant basic research is needed to advance toward a circularity of plastics, in which the chemical ingredients of plastics are reformed into polymers or recycled to give them another life. The basic research conducted in universities and national laboratories has the potential to quickly transform the process of recycling plastic waste into useful fuels, chemicals, and materials.

### 3.1 Efforts to improve mechanical recycling

One of the main challenges for traditional mechanical recycling is reduced properties upon recycling, *i.e.* downcycling. For example,

typical causes for degradation of mechanical strength upon mechanical recycling are related to the decrease in molecular weight ( $M_w$ ) due to repeated grinding and heating of the polymer. Additionally, some polymers, such as PE and PP, iPP in particular, have a very similar chemical composition and density, making it difficult to separate the two types of plastic into a single stream for mechanical sorting. The two polymers have very different physical properties including  $T_g$ , melting point and phase behaviors. Due to their property differences, it is hard to make a good blend of the two polymers as there will be a tendency to phase separate, causing a drastic decrease in mechanical strength when mixed. Therefore, items made from direct mechanical recycling of mixed plastics typically exhibit undesirable end-use qualities. In order to mitigate the challenges of blending or separating these types of polymers in a mixed feedstock, a number of studies on compatibilizers have been conducted.

For instance, compatibilization of PE/iPP blends by PE-PP multiblock copolymers greatly improves interface adhesion as well as prevents droplet coalescence during the melt-extrusion process, thereby changing brittle materials into mechanically robust blends. As depicted in Fig. 5, the addition of 1 to 5% PE-*b*-PP-*b*-PE-*b*-PP tetrablock copolymer drastically reduces the size of the dispersed PP phase in an HDPE matrix. The strengthening of the interface,

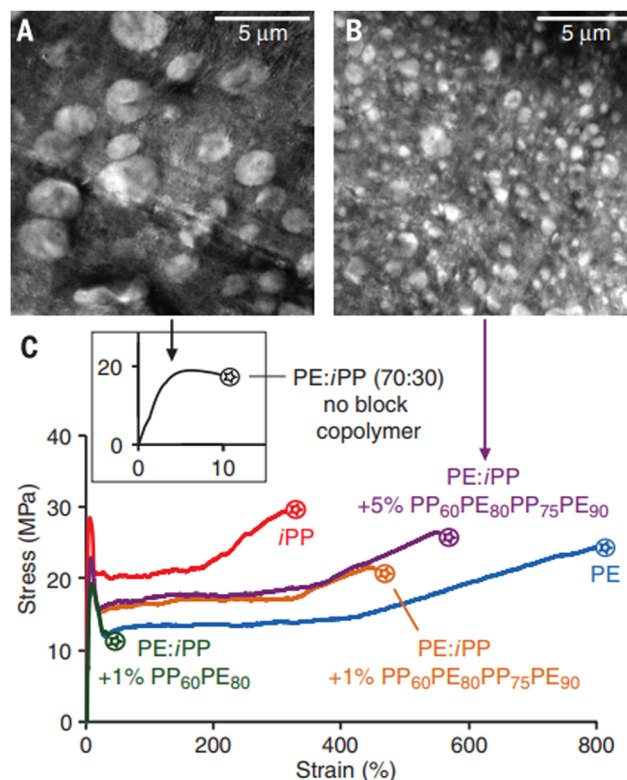


Fig. 5 Electron microscopy images of a 70/30 blend of HDPE and PP (A) without any block copolymer and (B) with 5 wt% of tetrablock copolymer. (C) The blend without any block copolymer is brittle and there is only modest improvement with the inclusion of 1 wt% of a diblock copolymer. The use of the tetrablock copolymer results in a blend with mechanical properties comparable to the pure (HDPE and PP) components.<sup>19</sup> Reproduced with permission from ref. 19. Copyright 2017 Science.





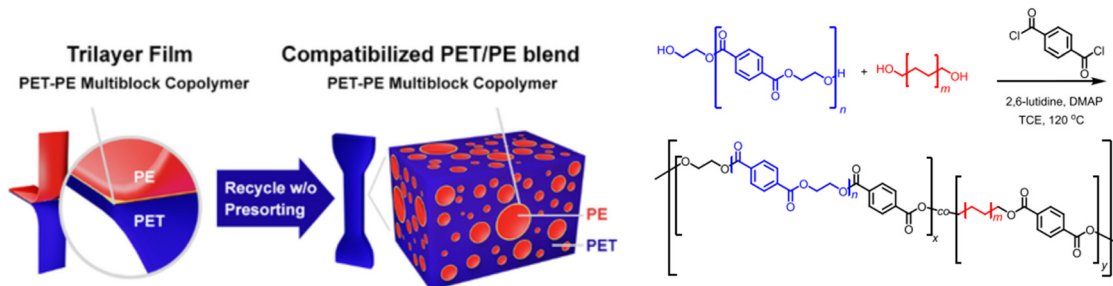


Fig. 6 Compatibilization of PET-PE multiblock copolymer and synthetic scheme for the preparation of PET-PE MBCP. Reproduced with permission from ref. 20. Copyright 2020 ACS Appl. Mater. Interfaces.

together with droplet size reduction, has transformed the HDPE/PP blend from brittle to tough and ductile. This study indicates that these block copolymer compatibilizers could enhance the recycling of HDPE and PP by reducing the need for sorting.<sup>19</sup>

Another example to improve recycling rates of immiscible polymer blends is using PET-*b*-PE multiblock copolymers (PETPE MBCPs) (Fig. 6), which have been shown to effectively improve the adhesion between the otherwise weak interfaces in melt reprocessed PET/PE blends. PET/PE (80/20 wt%) blends with less than 0.5 wt% of PETPE MBCP, which were melt mixed to simulate the recycling of mixed plastic waste, were found to be superior to those of pure PET. By understanding the mechanism in which the compatibilizer functions, highly immiscible PET/PE blends can be blended due to PETPE MBCPs acting as an interface adhesive, enabling the viability of recycling PET/PE mixed waste streams.<sup>20,21</sup>

### 3.2 Catalyst design to deconstruct polyolefin and condensation polymers

Catalytic deconstruction provides selective functionalization or degradation of one kind of polymer possibly within a mixture or

composite to recover particular chemical components. For instance, tandem catalysis demonstrates the high yield conversion of waste polyolefins to liquid alkylaromatics by Pt/g-Al<sub>2</sub>O<sub>3</sub> (up to 80 wt%). In a verification experiment, a low *M<sub>w</sub>* PE (0.118 g, *M<sub>w</sub>* = 3.5 kg mol<sup>-1</sup>, *D* = 1.9) was mixed with Pt/g-Al<sub>2</sub>O<sub>3</sub> in a 10 ml mini-autoclave without solvent or additional H<sub>2</sub>. The liquid/wax products (80% by mass) were recovered after 24 h at 280 °C by dissolving them in hot CHCl<sub>3</sub> (Fig. 7).<sup>22</sup>

Many nitrogen-containing nucleophiles, including amines and anilines, can also be used for PET deconstruction catalysts in addition to metal catalysts. Use of a readily available guanidine catalyst 1,5,7-triazabicyclo[4.4.0]dec-5-ene (TBD), enabled organocatalytic glycolysis of PET. Waste PET beverage bottles were deconstructed in 3.5 h at 190 °C with an excess of ethylene glycol (EG), yielding BHET as the predominant product (Fig. 8). Computational analyses showed that TBD and EG facilitated the depolymerization of PET by forming and activating hydrogen bonds with the polymer.<sup>11</sup> A protic ionic salt, combining TBD and methane sulfonic acid (MSA) at a 1 : 1 molar ratio was further developed for PET depolymerization, while exhibiting

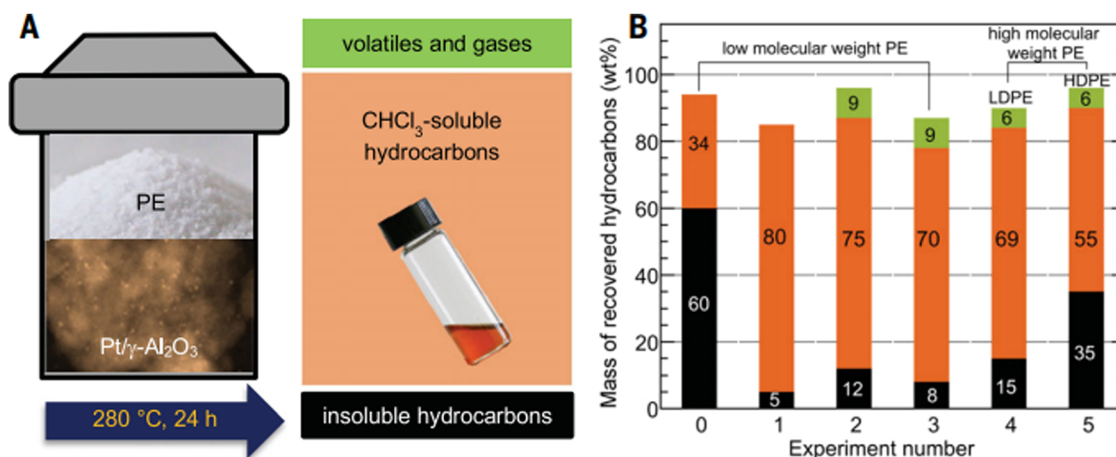


Fig. 7 Solvent-free conversion of various types of PE. (A) Schematic of reactor and product fractions, with photographs of the powdered polymer and liquid products, as well as a transmission electron micrograph of the catalyst. (B) Hydrocarbon distributions after 24 hours at 280 °C. Reactions of a low molecular-weight PE (*M<sub>w</sub>* = 3.52 kg mol<sup>-1</sup>, *D* = 1.90) in an unstirred mini-autoclave reactor: (0) catalyzed by g-Al<sub>2</sub>O<sub>3</sub> (no gas recovery); (1) catalyzed by Pt/g-Al<sub>2</sub>O<sub>3</sub> (no gas recovery); (2) catalyzed by Pt/g-Al<sub>2</sub>O<sub>3</sub> (with gas recovery). Reactions catalyzed by Pt/g-Al<sub>2</sub>O<sub>3</sub> in a stirred autoclave reactor with gas recovery: (3) low-molecular-weight PE; (4) LDPE bag (*M<sub>w</sub>* = 94.5 kg mol<sup>-1</sup>, *D* = 7.37); (5) HDPE bottle cap (*M<sub>w</sub>* = 53.5 kg mol<sup>-1</sup>, *D* = 3.61). Orange represents CHCl<sub>3</sub> soluble hydrocarbons, black represents CHCl<sub>3</sub> insoluble residues, and green represents recovered gases including H<sub>2</sub> and light hydrocarbons (C<sub>1</sub>–C<sub>8</sub>). Reproduced with permission from ref. 22. Copyright 2020 Science.



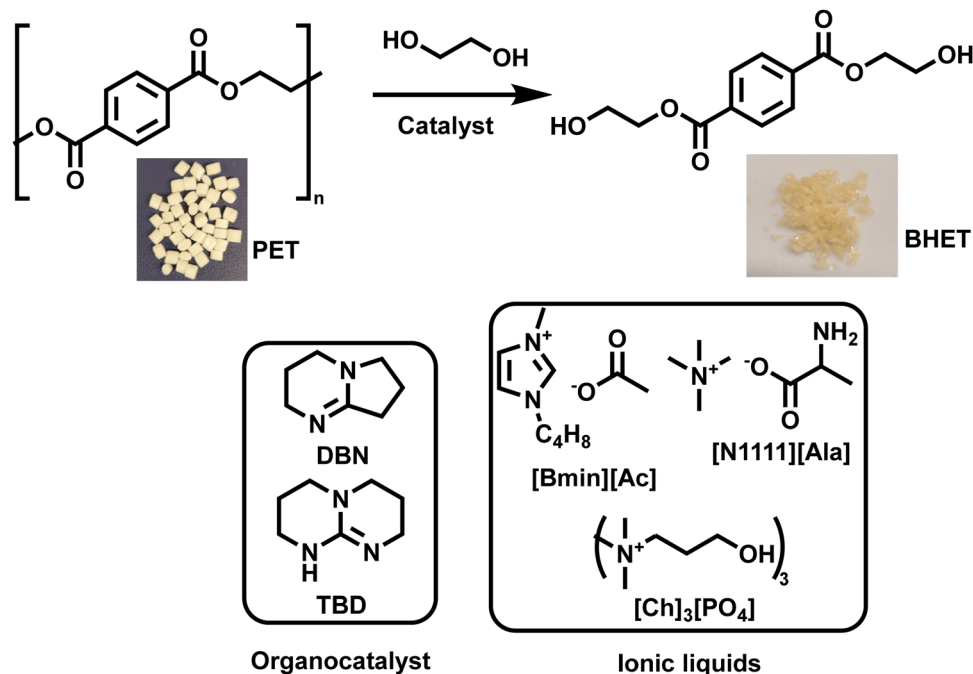


Fig. 8 Glycolysis of PET using different organocatalysts and ionic liquid.

high catalyst stability at temperatures up to 400 °C without degradation. In the presence of EG, PET can be successfully depolymerized by TBD:MSA in less than 2 h at 180 °C under an inert atmosphere. Reusing the TBD:MSA catalyst more than five times does not significantly degrade its ability to mediate the self-condensation of BHET and produce fresh PET with characteristics similar to those of virgin PET.<sup>11,23,24</sup> Additionally, the same technology can be used to deconstruct polycarbonates to obtain monomers such as bisphenol A.

Metal based catalysts have also been reported to show significant advancement in the deconstruction of various plastics. Using a combined tandem catalytic and biological process, mixed plastics are transformed into single products (Fig. 9). The first catalytic step employs metal-promoted autoxidation to depolymerize mixed or single polymers of HDPE, PS, and PET

into oxygenated intermediates (dicarboxylic acid, benzoic acid, and terephthalic acid) that are advantageous substrates for further bioconversion. In the biological stage, a robust designed bacterial strain such as *Pseudomonas putida* converts the mixed oxygenates into  $\beta$ -keto adipate monomer, which can be polymerized to polyhydroxyalkanoates, a biologically synthesized polyester that has industrial applications.<sup>25</sup>

### 3.3 Materials designed for recycling

New designs are necessary for new generation of recyclable or upcycled polymers that have properties equal or better than those of today's materials. To establish close-loop circularity of plastics *via* mechanical recycling and chemical upcycling, new plastics must be purposefully designed to have a specific set of

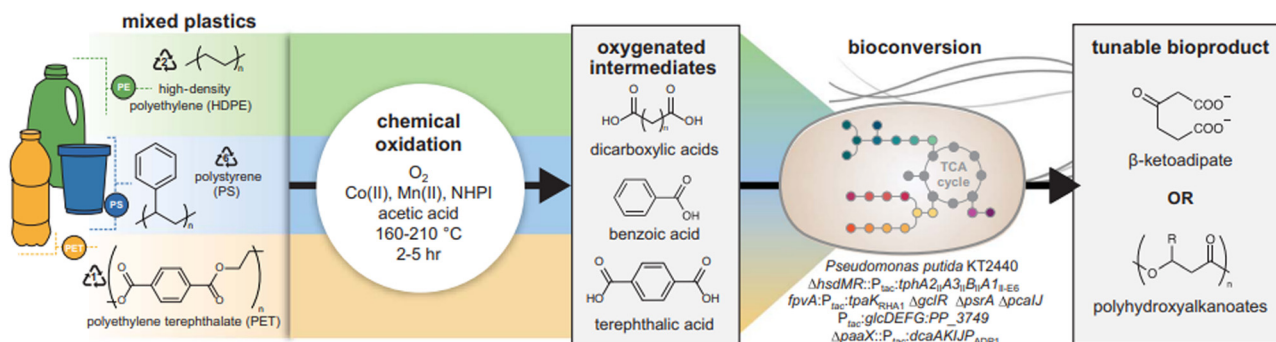


Fig. 9 Upcycling of mixed plastic waste through tandem chemical oxidation and bioconversion. Metal-promoted autoxidation simultaneously deconstructs multiple polymers, generating a mixture of oxygenated intermediates that are advantageous substrates for bioconversion. An engineered *P. putida* strain funnels the heterogeneous mixture of oxygenates into a single target product. Reproduced with permission from ref. 25. Copyright 2022 Science.





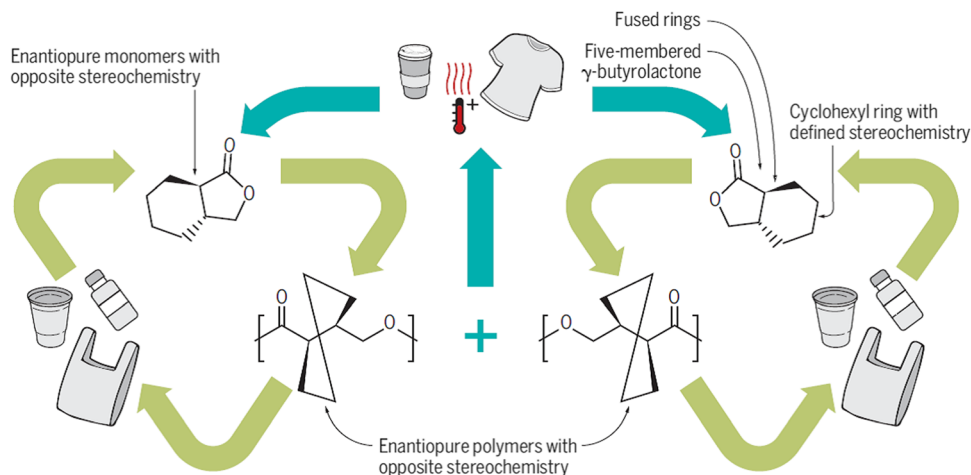


Fig. 10 Polymers synthesized from  $\gamma$ -butyrolactone exist as two enantiomers and when blended, this allows for materials with improved thermal stability which can be recycled repeatedly without losing mechanical properties. Reproduced with permission from ref. 28. Copyright 2018 Science.

physical and mechanical properties as well as a mechanism to efficiently breakdown by atomic- and energy-efficient processes at the end of life. In this section, we will highlight some promising areas of materials development, beyond traditional polymer, that offer potential solutions in the future plastics economy.

One method to develop materials that are better suited for circularity is to design monomers that can be polymerized into a polymer and can be depolymerized to form the same monomer. For example, a polymer system based on  $\gamma$ -butyrolactone (GBL) has been reported to be polymerizable at room temperature to yield high molecular weight poly( $\gamma$ -butyrolactone). The structure of the synthesized polymer exists as either a linear or a cyclic structure and can be readily depolymerized *via* thermolysis at high temperatures or chemolysis in the presence of a catalyst (Fig. 10).<sup>26</sup> Similarly, a bridged bicyclic thiolactone (BTL) monomer can be polymerized *via* ring opening polymerization to obtain a ductile tough plastic, where poly(BTL) can be deoligomerized and ring closed to reform the original BTL monomer.<sup>27</sup>

Since these polymers can be depolymerized to reform the exact same monomer as used in the synthesis, recovered monomers can be easily integrated back into the manufacturing process of the polymer.

A closed-loop recyclability of monomer to polymer, then to monomer, has also been reported for cyclic acetals.<sup>29</sup> The use of reversible-deactivation cationic ring opening polymerization enables control of the polymerization of cyclic acetals (Fig. 11A) to produce polyacetals with controlled molecular weights of 38–182 kg mol<sup>-1</sup>. The synthesized poly(1,3-dioxolane) exhibits desirable chemical and thermal stability, suitable for consumer products (Fig. 11B). Moreover, they can be depolymerized at relatively low temperatures to reform cyclic acetal monomers with the presence of a Lewis acid catalyst. Monomers can be separated after the depolymerization by simple distillation even in the presence of various other polymers and be used to repolymerize polymers with properties identical to virgin polymers.

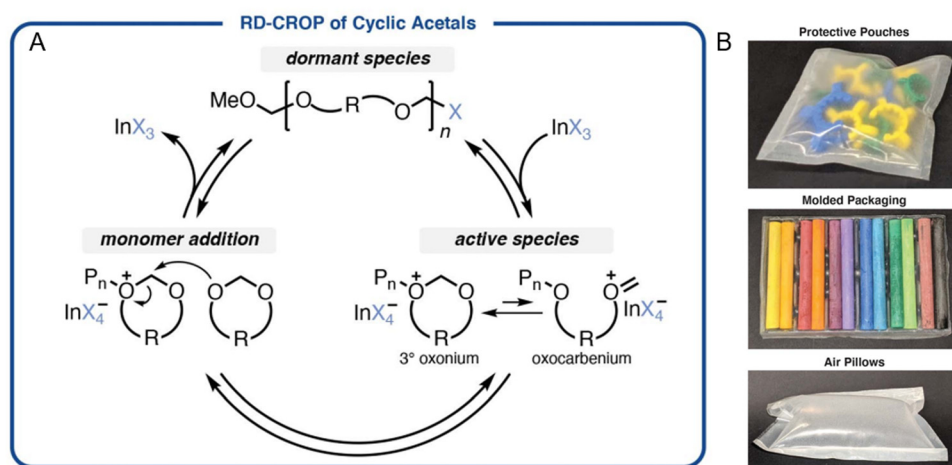


Fig. 11 (A) Mechanism for reversible-deactivation cationic ring opening polymerization of cyclic acetal monomers to produce polyacetals. (B) Polyacetal can be used to make consumer products such as protective pouches, packaging, and air pillows. Reproduced in part with permission from ref. 29. Copyright 2021 Science.



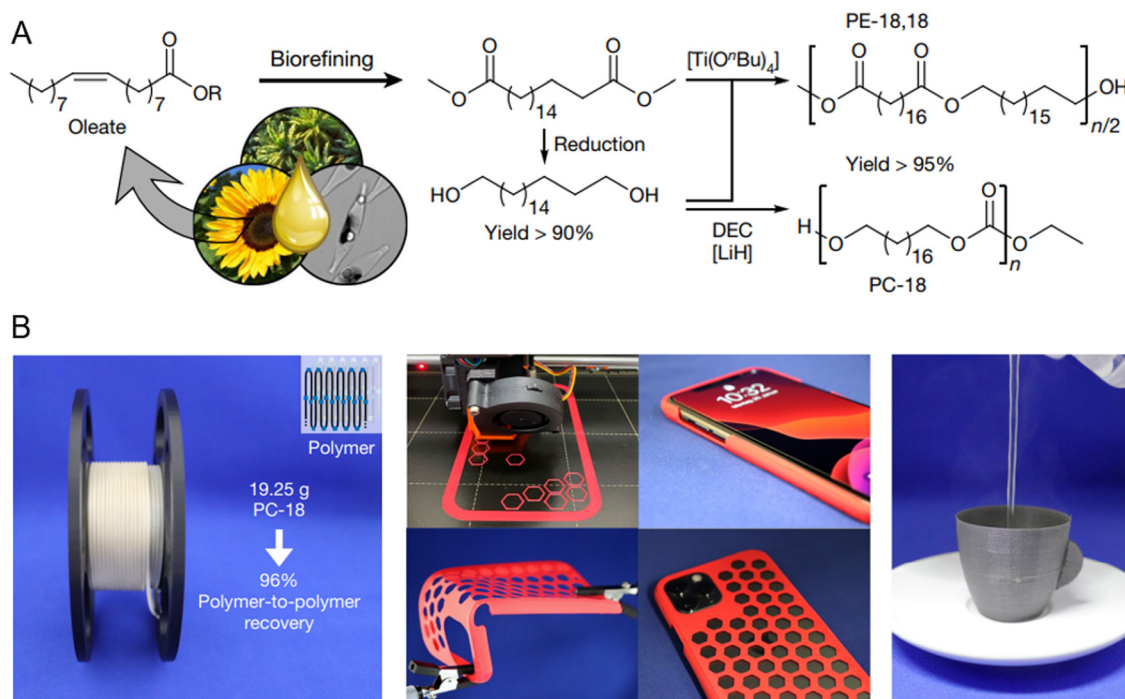


Fig. 12 (A) Biobased materials can be used to make building blocks to make high molecular weight polyethylene-like materials. (B) Polymers can be made into filaments used to 3D print products such as smartphone cases and cups that can hold boiling water while maintaining shape and dimensions. Reproduced in part with permission from ref. 30. Copyright 2021 *Nature*.

Polyethylene-like materials containing readily depolymerizable functional groups have also been synthesized from bio-based building blocks (Fig. 12A). The polymers made from this material are able to have high molecular weights ( $M_w = 300 \text{ kg mol}^{-1}$ ,  $M_n 90 \text{ kg mol}^{-1}$ ) with mechanical properties similar to HDPE, while the polymer can be broken into repolymerizable fragments by reacting at  $120^\circ\text{C}$  in ethanol.<sup>30</sup> The reported polymers can also be used as a filament for 3D printing, and the printed products include snap on smartphone cases and even a 3D printed cup which can hold boiling water while still maintaining its shape and dimensions (Fig. 12B).

Thermoset materials possess crosslinked networks which provide excellent mechanical properties, but they are conventionally not amenable to mechanical recycling or solution processing due to the presence of permanent covalent networks. Covalent adaptive networks (CANs) allow acrosslinked network to be processable and recyclable, while exhibiting thermoset-like properties, due to the presence of dynamic dissociative or associative exchange groups. The dynamic bonds act as movable or reversible anchor points in the network, enabling the bulk material to be reshaped or replaced, sometimes without stimulus (self-healing).<sup>31–34</sup> Epoxy networks with alcohol and ester functionalized components were first reported to exhibit dynamic exchange *via* transesterification processes, and CANs were named as vitrimers.<sup>35</sup> The epoxy-based material exhibits robust mechanical properties at room temperature and when placed in solvents at high temperatures, only swelled and did not dissolve. The material was proven to be reprocessed by injection molding. To enhance the recyclability of epoxy-based vitrimers, polyurea/epoxy vitrimers

with exchangeable disulfide bonds were recently designed. The tailored design allowed lower reprocessing temperatures compared to conventional disulfide-based epoxy-based vitrimers. The material can be reprocessed *via* a heat press in as little time as 1 minute and multi-cycle recycling maintains its tensile strength of  $\sim 60 \text{ MPa}$  even after being reprocessed 6 times, a stark contrast to the conventional vitrimer, losing 63% loss of tensile strength. The vitrimer can be used in conjunction with carbon fibers, while providing the reprocessability and reparability of the carbon fiber composites.<sup>36</sup>

CANs of diketoenamine chemistries have also been used in closed-loop recycling of polymer network systems (Fig. 13A). Diketoenamine bonds from triketones and amines, both aromatic and aliphatic, react spontaneously and can form network structures. Under strong acidic aqueous conditions, the diketoenamine bond hydrolyzes and reforms the triketone and an ammonium salt.<sup>37</sup> Due to the depolymerization conditions, it is possible to extract polydiketoenamine based polymers from a mixed plastic waste feedstock by mixing with a strong acid (Fig. 13B). The acidic solution containing the deconstructed polydiketoenamine, now triketone monomers, can be precipitated and pure monomer is recovered. The monomers can then be repolymerized and used to make more polydiketoenamine based products, exhibiting the same mechanical properties as before and enabling circularity due to the reformation of the diketoenamine bonds. In addition to amines and ketones, this type of dynamic bond can also be seen between imines and hydrogens to exhibit self-healing behavior.<sup>38,39</sup>





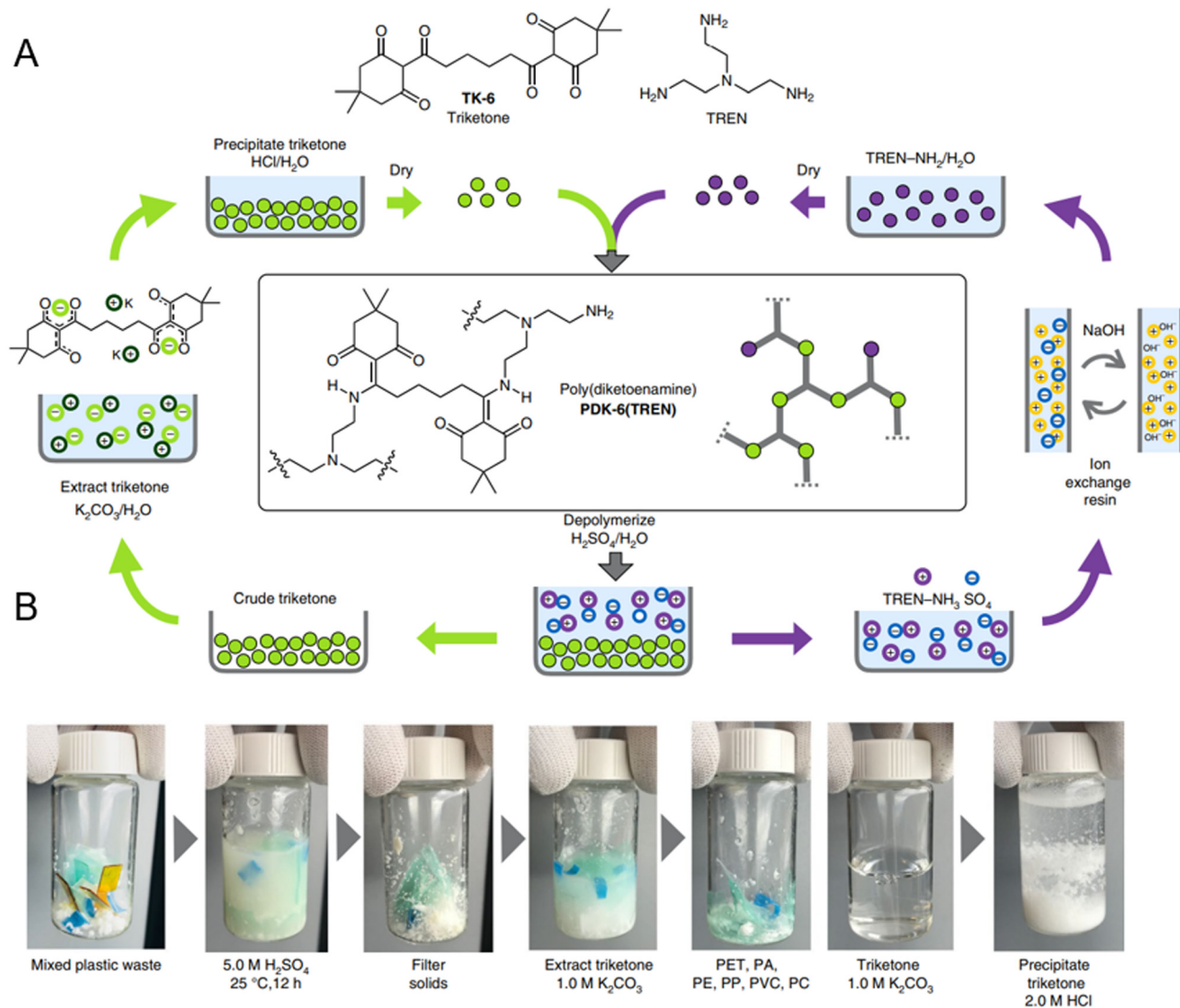


Fig. 13 (A) Mechanism for polydiketoenamine synthesis and depolymerization. (B) Polydiketoenamine based polymers can be extracted from a mixed plastic waste source to obtain pure triketone. Reproduced in part with permission from ref. 37. Copyright 2019 Nature Chem.

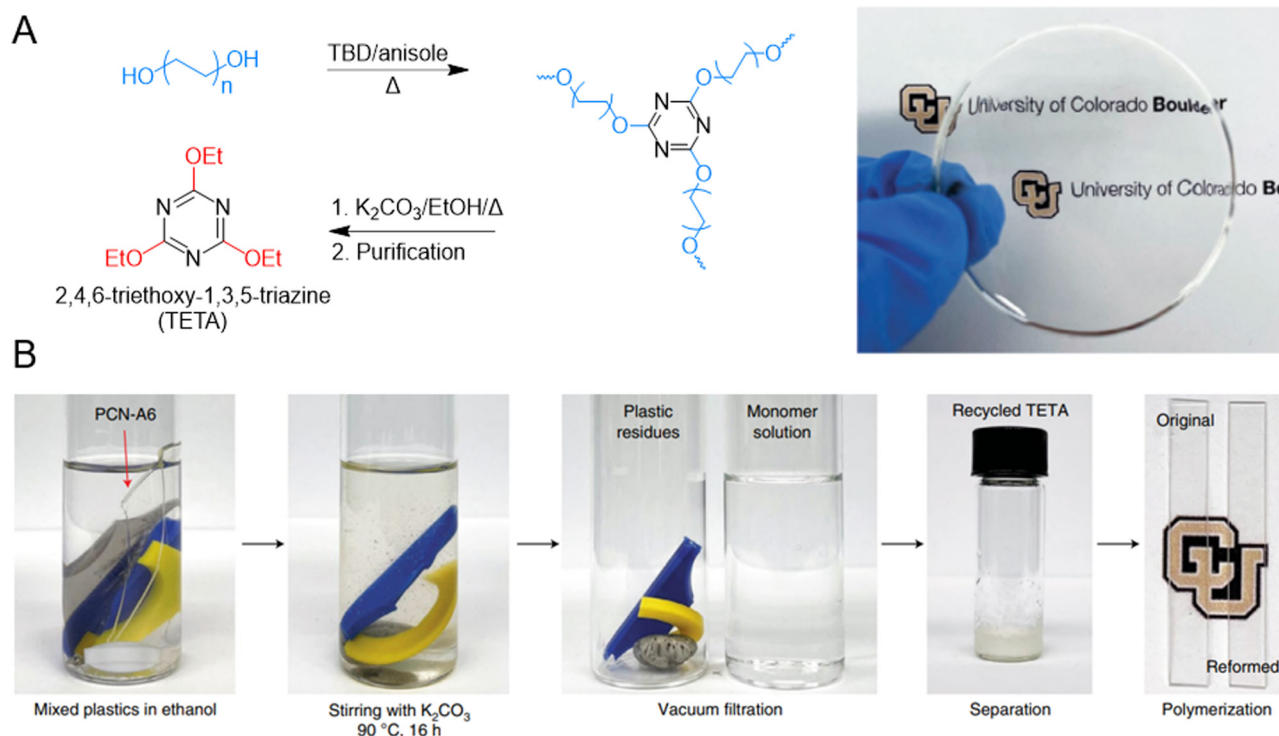
Recyclable and malleable polycyanurate based network polymers have also been reported. Triazine containing monomers, such as 2,4,6-triethoxy-1,3,5-triazine (TETA) monomer synthesized from isocyanates, are polymerized to obtain polycyanurates (Fig. 14A), which exhibit strong mechanical properties with varying ductility based on the structure of monomer used.<sup>40</sup> The polycyanurates can also be depolymerized to reform the triazine monomer by refluxing in ethanol, and the depolymerization can be accelerated with the addition of  $K_2CO_3$ . The recovered monomers can be used to repolymerize materials of similar properties as the original virgin polymer (Fig. 14B).

Polyolefin-based commodity plastic has been upcycled by CANs, providing a processable and recyclable crosslinked network with enhanced properties. Commercial thermoplastic HDPE has been transformed to high-performance vitrimers by grafting dioxaborolane-functionalized maleimides onto HDPE, followed by cross-linking with a bis-dioxaborolane metathesis onto pendant dioxaborolane units in the polymer (Fig. 15A). The

upcycled HDPE vitrimers exhibit improved melt strength, dimension stability and show resistance to cracking in the presence of solvents.<sup>41</sup> Additionally, they exhibited enhanced polymer-polymer adhesion due to the presence of dynamic dioxaborolane (Fig. 15B).

Similarly, dynamic covalent bonds of boronic ester can be used to upcycle triblock styrene-ethylene-butylene-styrene (SEBS) thermoplastic elastomers. The boronic ester groups on SEBS can reversibly bind with hydroxyls on the surface of different fillers and substrates, leading to exceptional adhesion, surpassing the adhesive strength of various commercial adhesives (Fig. 16A). The dynamic interaction also improved their tensile strength, toughness, and temperature service window while maintaining recyclability.<sup>42</sup> Upcycling commodity plastics *via* CANs can also be applied to additive manufacturing with a closed loop. An engineering plastic, acrylonitrile butadiene styrene (ABS) and its waste are first upcycled to ABS-vitrimer. The ABS-vitrimer exhibits superior mechanical performance

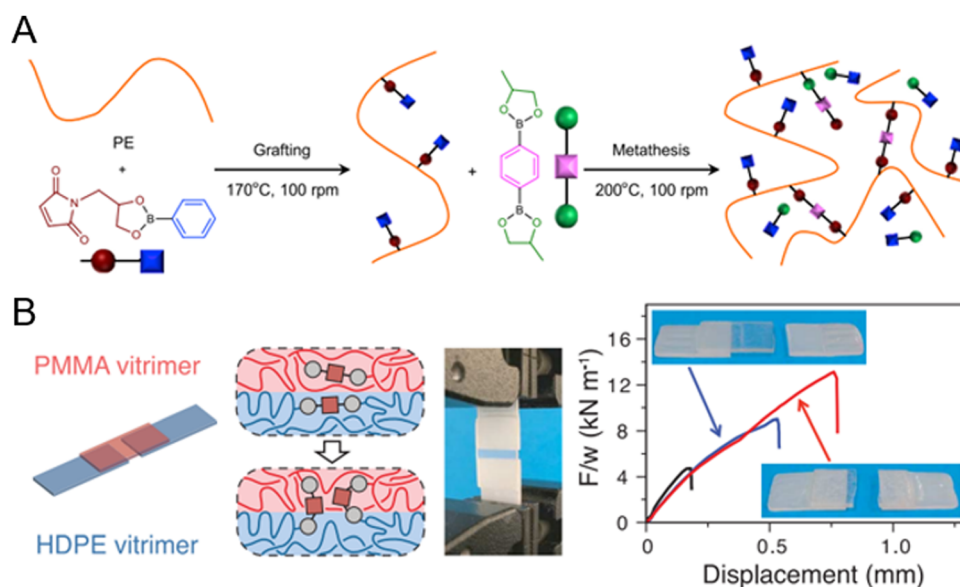




**Fig. 14** (A) Depolymerization and synthesis of polycyanurate network polymers. (B) Polycyanurates can be depolymerized in the presence of ethanol and  $\text{K}_2\text{CO}_3$  added as a catalyst. Reproduced in part with permission from ref. 40. Copyright 2022 *Nature Chem.*

(e.g.,  $2\times$  toughness,  $1.8\times$  tensile strength) and chemical resistance compared with untreated ABS. Importantly, ABS-vitrimer or their mixture with ABS can be readily reprinted multiple times by fused filament fabrication (FFF) (Fig. 16B). This easy adoption to FFF enables the manufacturing of intricate 3D structures unachievable through conventional molding or casting. Furthermore,

the waste of ABS-vitrimer can be easily separated from mixed plastic waste by dissolution for multi-cycle reuse, and the dissolved ABS can be again upcycled to ABS-vitrimer. The multiple paths of chemical upcycling and separation, coupled with facile additive manufacturing offer an effective and realistic carbon-efficient closed-loop circular plastic manufacturing.<sup>43</sup>



**Fig. 15** (A) Conversion of commercial HDPE to vitrimers via metathesis with dioxaborolane. (B) Adhesion between the two vitrimers is also enhanced due to the presence of dynamic dioxaborolane bonds. Reproduced in part with permission from ref. 41. Copyright 2017 *Science*.



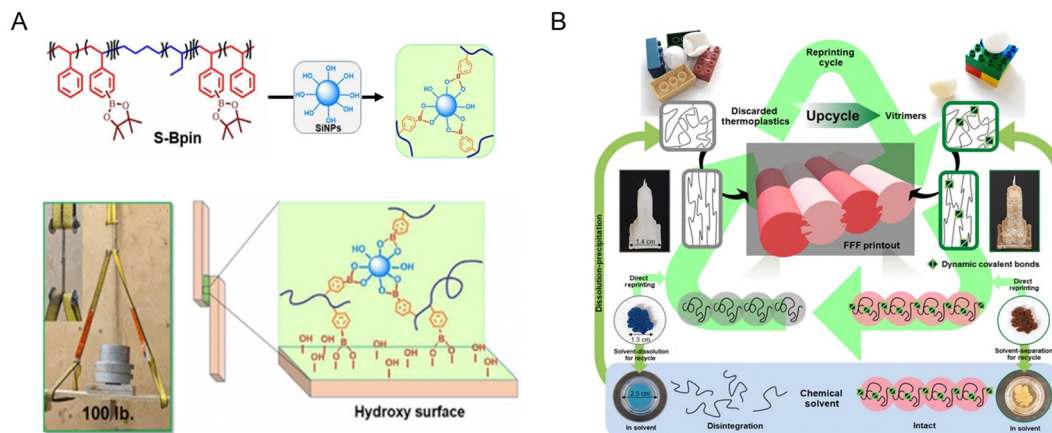


Fig. 16 (A) Tough adhesive from upcycled commodity polymer SEBS. (B) Closed-loop additive manufacturing via upcycled ABS vitrimers. The photographs show structures printed from the untreated ABS (Neat-ABS) (left) and the ABS-vitrimer (right) of a building (top) and an oak leaf before (middle) and after (bottom) immersion in tetrahydrofuran (THF) for 48 hours. Reproduced with permission from ref. 42 and 43. Copyright 2021 *Sci. Adv.* and 2022 *Sci. Adv.*

## 4. Summary and outlook

### 4.1 Commercial recycling is mainly mechanical recycling

The majority of commercial plastic recycling today is mechanical recycling. While the process has been well developed and can be fast and inexpensive for common plastics, such as plastic bottles, it is typically limited to feedstocks of a single type of plastic. Sorting is required in order to obtain a single type of desired plastic feedstock at high percentage, which can involve several detectors such as NIR and UV-Vis to differentiate between plastic type and color. There are some limitations to these detectors to distinguish polymers that might be chemically similar but have significantly different physical and mechanical properties. The difference in physical and mechanical properties can result in a macro-phase separated blend and a large shift or incompatibility in the process temperature and conditions, causing issues with the melting and extrusion of plastics in various industrial machinery.

Another challenge for mechanical recycling is that recycled plastics generally exhibit lower mechanical strength due to the repeated heating and extrusion process as well as degradation during use. Therefore, in order to ensure achieving sufficient properties of recycled plastic resins, the recycled resins are typically blended with virgin polymers or with additives that can increase the strength of the material. Complex types of plastic waste are also a challenge for mechanical recyclers. Blended or mixed plastic waste that has several types of polymers are combined in a way that cannot be easily mechanically separated. Such plastic waste is essentially of no value to mechanical recyclers since the material is generally not processable. Waste sources such as multilayer packaging and textiles fall into this category.

### 4.2 Academia is mostly focusing on chemical recycling

While a lot of promising technology is developed in academia, potential commercialization paths are not frequently considered when designing new technology. Chemical recycling has

significant potential to address feedstock concerns, such as mixed plastic waste that cannot be separated *via* mechanical sorting. However, the process generally costs significantly more compared to mechanical recycling and the monomers produced from chemical recycling have a higher associated cost than producing the conventional petroleum-based monomer. While some of the chemical recycling process indicates significantly lowering associated GHG emissions and energy consumption, few companies are willing to pay a higher price for the green premium offered.

There is often a large uncertainty about what the scaling up process of academic research looks like and how feasible commercial streams of waste plastics are in the scaled-up operation. Unlike the simulated feedstock used in laboratory experiments, real waste has significantly higher levels and more types of contaminations. For example, while a PET deconstruction catalyst can deconstruct a PET water bottle with fewer sorting steps and can tolerate high levels of contamination, certain contaminants in real wastes may completely hinder the deconstruction reaction. Even if the catalyst was tested on a water bottle, even with the presence of labels and some amount of water to simulate water being left in the bottle, it does not account for the unexpected contaminations that a bottle may encounter during its lifetime (*e.g.* cigarette butts or chewing gum). Though these contaminations could be trivial, there could be unintended effects at a scaled up commercial scale.

Another strength of academic research is developing new classes of polymer materials. Polymers can be designed to have more sustainable functionality such as the incorporation of dynamic bonds, which can reform, extend the lifespan of materials, and reduce the amount of waste disposed. There are also polymers being developed that can be depolymerized under certain conditions to obtain the same monomer used to make the virgin polymer, enabling simpler recycling processes. To add even more appeal to this type of polymer, some polymers can even be made using previously deconstructed waste plastic to further improve lifecycle assessments.





However, with the development of new materials, it is important to consider the potential impact of materials used on their adoptability and scalability toward industry adoption as well as the mitigation of unintended adverse environment impacts.

### 4.3 Bridging the gap

Recycling rate and effort are significantly different across the world—even within the U.S., it is done differently based on region. Since most of the U.S. recycling infrastructure was developed in the 1950s and 60s, some systems and technologies are outdated particularly for the collection and sorting of plastic waste. While there are new technologies that can be retrofitted into existing MRFs to improve sorting processes, waste containing mixed or blended plastics or new types of polymers still proves to be a challenge for mechanical recycling. At the same time, academia across the world has innovated and developed a variety of recycling and upcycling technologies that can work with these challenging plastic waste feedstocks. However, there is some disconnect between the technologies academic research offers and the needs of industries, and this gap needs to be closed.

There are several ways that academic research can improve plastic recycling including designing materials that can be more easily recycled, developing new types of polymers that have better end-of-life processing, and developing very efficient methods to break plastics down to valuable chemicals or monomers. By designing materials such as packaging to have fewer types of polymers or that are readily delaminated, mechanical recycling could be more effective at creating valuable recycled materials from waste. Designing polymers that have dynamic bonds, which can be broken and reformed, can also improve the mechanical recycling as well as extend the longevity due to its healing capability, particularly if the materials can be reformed in simple ways such as applying heat.

Another approach is developing methods to deconstruct plastic waste back into monomers or valuable chemicals. Efficient chemical recycling processes for recovering monomers from plastic waste can reduce the amount of fossil fuel needed to make polymers that can be a favorable process if the monomer is used in the manufacturing of the polymer easily integrated in the polymer manufacturing supply chain.

There are many important aspects to consider when considering commercializing academic research. Not only is life cycle assessment important to consider and report in publications, but it should also be reported in a standardized way so that many types of research can be more comparable. Additionally, it is critical that the developed technology considers the potential environmental side effects of the technology developed, such as excessive use of resources in energy or water, or the use of extremely toxic chemicals. Transparency of data is also important when thinking about commercialization. While technologies might work in the lab environment, more often than not, the same conditions are nearly impossible to recreate in the real world. Feedstocks that are collected and sorted by MRFs and reclaimers will likely not contain as high a percentage of the desired type of plastic waste as hoped for, due to the associated cost or feasibility of sorting processes.

Chemical recycling processes are also still very expensive—the investment required to build a chemical recycling facility is significantly higher than mechanical recycling. For example, in 2021, Eastman invested \$250 M to construct their Kingsport, Tennessee, USA methanolysis facility,<sup>44</sup> which has an annual processing capacity of 100 kt of plastic waste. Eastman also invested up to \$1 B to build a similar plant in Normandy, France, which would have a processing capacity of 160 kt y<sup>-1</sup>.<sup>45</sup> Similarly, Encina announced in 2022 a \$1.1 B investment plan for building a state-of-the-art manufacturing facility in Point Town, Pennsylvania, USA with a processing volume of 449 kt of plastic waste.<sup>46</sup> Additionally, due to chemical recycling processes being more complex and newer than mechanical recycling, the estimated minimum selling price for products of processes such as glycolysis and methanolysis is \$0.96–1.04 kg<sup>-1</sup>, almost double the cost of mechanically recycled plastic, \$0.54 kg<sup>-1</sup>.<sup>47</sup> Thus, academic efforts to improve efficiency or processing speed/volume of chemical recycling will reduce the cost, and make chemical recycling much more impactful in the economy.

Advancing recycling efforts is not a task that can be done by academic research or the recycling industry independently. By communicating and bridging the gap of what academic research offers and what the recycling industry needs, we can work together to mitigate the amount of plastic waste being discarded into landfills and the environment. Understanding how the technology works in the lab and how to integrate it into current industrial processes is critical, and the considerations of supply chains and cost minimization will navigate the deployment. Furthermore, establishment of new technology with closed-loop plastic circularity will be able to lower carbon and energy footprint toward industrial decarbonization. Bridging the gap in plastic recycling will accelerate innovation and help us to solve the global challenge of plastic waste management.

## Conflicts of interest

The authors declare no conflicts of interest.

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