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Enabling a greener plastic future through molecular science

Latest techniques to reduce the impact of plastic pollution on public health and the environment

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Headlines

Issues

- Plastic is a versatile, high-performance material that has become the preferred material for a wide range of applications.
- Plastic production is expected to double in 20 years and almost quadruple by 2050, by which time the plastic industry will account for 20% of the world's total oil consumption.
- Plastic pollution is one of the main socio-environmental challenges that we face today. It is a multi-dimensional problem with several causes that involve not only plastics as materials but also the way they are made, commercialised, used, discarded and often mismanaged.
- The way plastics are made is complex, and some of the substances used in their manufacture make them problematic and uneconomical to recycle.

Solutions

- Researchers at Imperial College London are researching how we can reduce the amount of plastic leaked into the environment. However we need:
  - more research into how molecular science can improve recyclability;
  - new policies, better commercial and civic practices, and increased public awareness of resource efficiency. Although there is no "silver bullet" to address the complex problem of plastic pollution, making plastic products more reusable and recycling friendly through novel material combinations, processing methods and overall design can make a significant contribution.
- New technologies are being developed that can recover polymers and monomers (the building blocks that make up plastic) as a way to exclude impurities and improve recyclability in future.
- It is worthwhile for stakeholders to rethink how we can manufacture plastics in future – helping to reduce toxicity and increasing the amount of material that can be recovered via recycling.

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Objectives of this Briefing Paper

The Institute for Molecular Science and Engineering (IMSE) has brought together world-leading experts at Imperial College London in the fields of environmental policy, material sciences, mechanical engineering, and molecular chemistry to assess how we can improve the recyclability of plastic, recover the most material, and enhance value in the future.

In this Briefing Paper we will:

- Provide an overview of the impact of our use of plastic on the environment and some of the risks to our health.
- Assess current plastic recycling and manufacturing methods, highlighting issues, successes and opportunities.
- Suggest how future developments in manufacturing and recycling could reduce the negative impacts of plastic use.

What is plastic?

Plastics, from the Greek *πλαστικός* (*plasticos*), meaning mouldable, are materials made by joining a large number of small molecules together. These small molecules, called monomers, are joined together in a chemical reaction known as polymerisation. The properties of the resulting plastic ("polymer") can be controlled by changing the conditions of the chemical reaction and the combination of monomers. Polymer properties that can be varied in this way include thermal conductivity, crystallinity, electrical insulation, elasticity and permeability.

If a polymer is made from only one type of monomer, the product is a *homopolymer*, referred to simply as a polymer. Where two or more types of monomers are used, the final product is called a *copolymer*. A common example of a copolymer is PET, which is made from the polymerisation of ethylene glycol and terephthalic acid, and is used as a food packaging material and in the textiles industry (Figure 1).

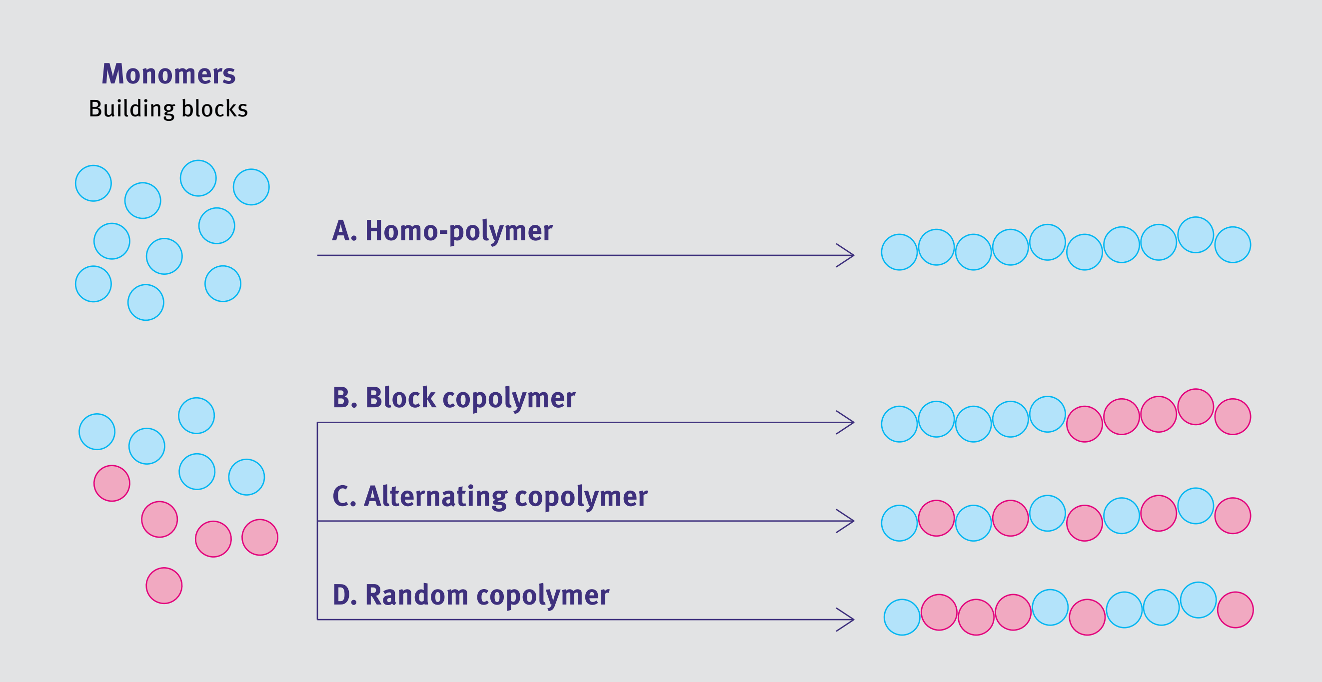


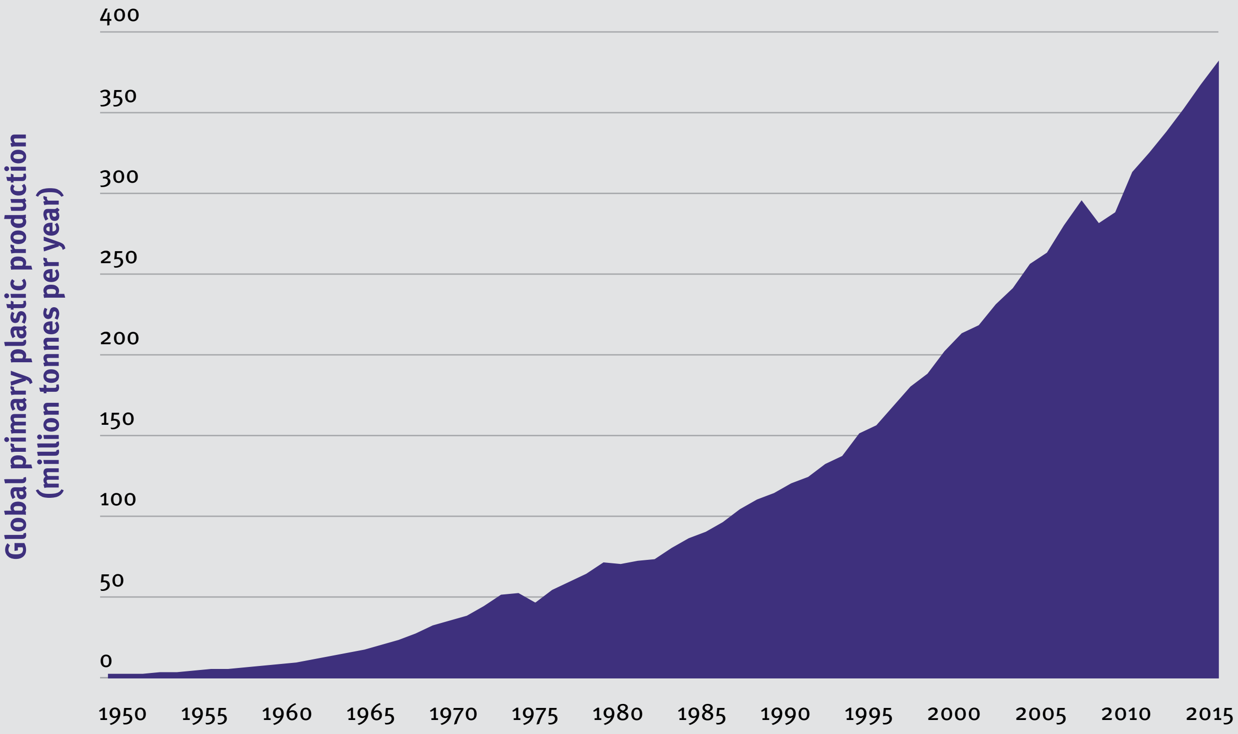
Figure 1: Representative polymerisation routes to various types of polymers.

In broad terms, the more complex the make-up of the polymer, the harder it is to break it down to its original building blocks, so high-quality recycling is difficult to achieve.

The use of plastic in modern society

Since its invention by Leo Bakeland as “Bakelite” in 1907, plastic production has increased exponentially over the past half-century, reaching 381 million tonnes per annum in 2015 (Figure 2).[1] Plastics are now an integral part of the global economy.[2]

In the EU alone, the plastics sector employs 1.5 million people and generated a turnover of €360 billion in 2018.[3] Packaging is the largest application of plastics in Europe, representing approximately 40% by weight of total market demand. Polyolefins (PE, PP) make up the biggest share thereof, followed by PP, HDPE, and PET. Smaller shares are contributed by PVC, PS, polyamides, and other plastics.[4] As packaging materials, plastics are inexpensive, lightweight, and high performing (Figure 3). Their light weight reduces fuel consumption for transportation. Barrier properties increase



Percentage market share for production by plastic type in Europe for 2018

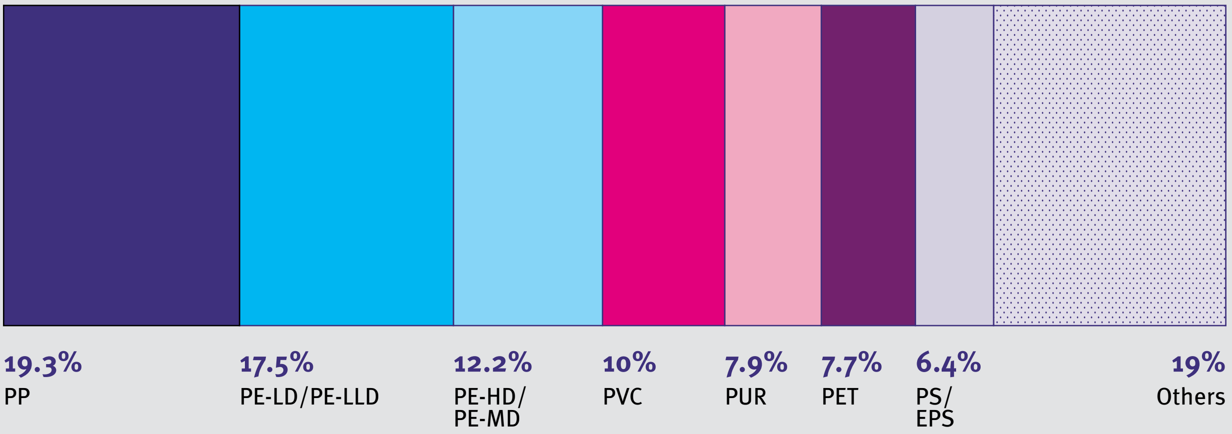


Figure 2: Global growth in plastic production during the years 1950–2015,[2] and market share by plastic type in Europe for 2018.[3]

the shelf life of fresh products, which reduces food waste. As a result of these characteristics, plastics have increasingly replaced other packaging materials.

The low cost, versatility, durability, and high strength-to-weight ratio of plastics provide significant economic benefits in other applications. Approximately 20% of a car by weight and about 50% of a modern passenger aircraft[6] are plastic.

Plastics production is expected to double in the next 20 years and almost quadruple by 2050.[2] The production of plastic relies heavily on oil from non-renewable fossil resources. If the growth in plastic production continues at the current rate, by 2050 the plastic industry will account for 20% of the world's total oil consumption. It is currently 5%.

Therefore, even if global energy production could become independent of fossil fuels, fossil resources may continue to be extracted for plastic manufacturing. In the EU, for example, the full potential of recycling plastic waste remains largely unexploited. Current reuse and recycling of end-of-life plastics is very low, particularly in comparison with other materials such as paper, glass, or metals.

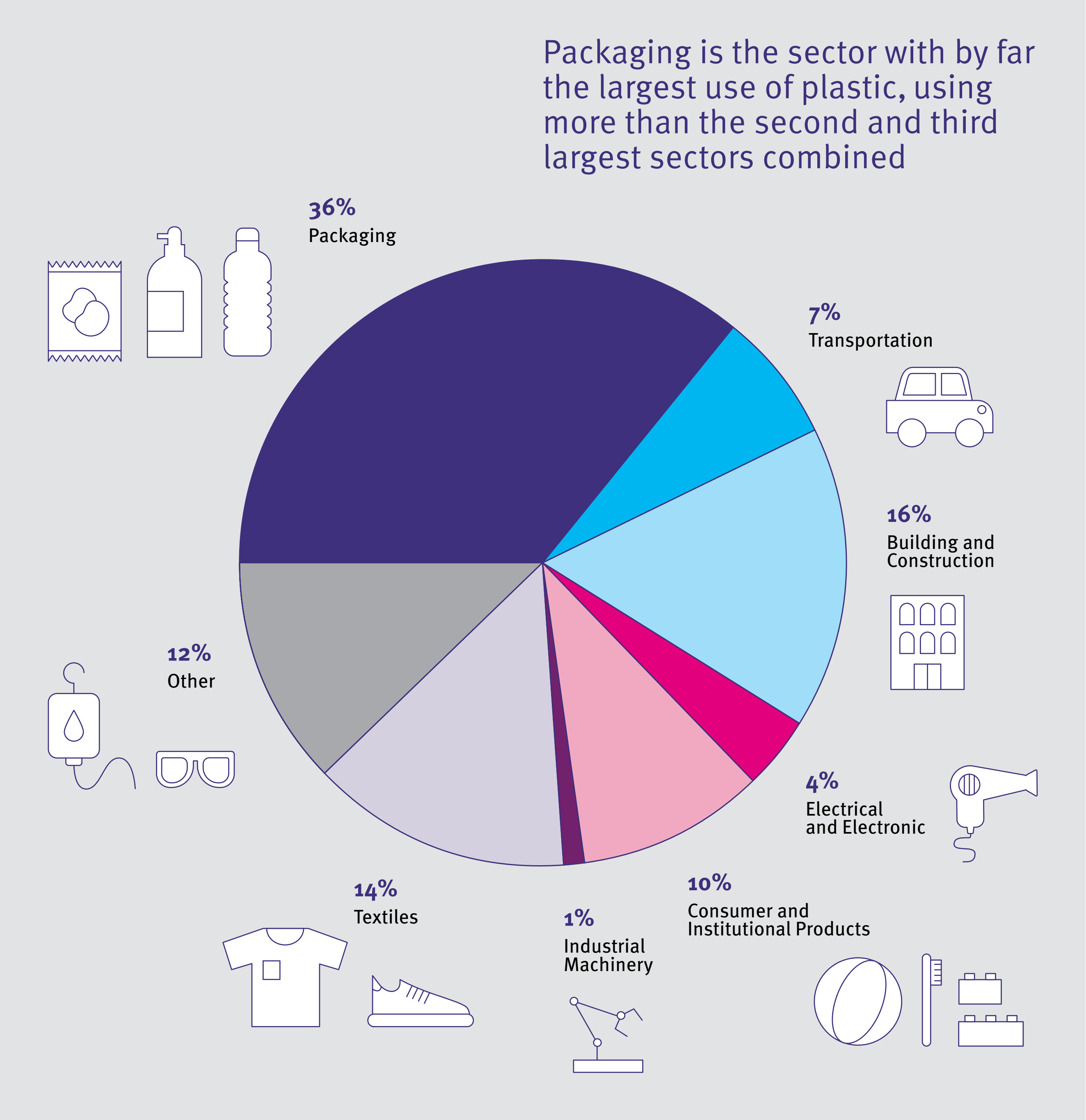

Packaging is the sector with by far the largest use of plastic, using more than the second and third largest sectors combined

Figure 3: Global plastic production by industrial sector in 2015.[2,5]

Additives

Plastics can be combined with certain *additives* in order to enhance their thermomechanical or physiological properties depending on their intended use.[6] Additives can be subdivided into (i) modifiers (ii) stabilisers and (iii) fillers. Modifiers are added to alter the base properties to improve performance, stabilisers are added to maintain the molecular structure during storage, processing and operational life, and fillers are mainly added in large quantities to dilute cost and alter physical properties.[7]

Such additives can reinforce the plastic material or allow the plastics to be processed at high temperatures. Plasticisers are used to render the material pliable and flexible, flame retardants to discourage ignition and burning, and UV stabilizers to prevent degradation by sunlight. Dyes, matting agents, opacifiers and lustre additives might also be used to enhance the appearance of a plastic product. They are also used to compensate for the degradation during mechanical recycling.

Additives are often the most expensive component of a formulation, and the minimum quantity needed to achieve a given level of performance is generally used. The additives are intimately mixed with the polymer or compounded into a formulation that is processed into the shape of the final product. Plastic materials may also include contaminants as a result of improper cleaning, staining or absorption of substances from the environment, as well as 'legacy' materials which have been phased out of plastic production but may still exist in older plastic products.

Additives or precursors such as bisphenol A (BPA) and certain phthalates, which are used as plasticisers to increase flexibility and durability of PVC and PC materials, have already raised concerns because of the risk of adverse effects on human health and the environment.[8–12] However, **they can also cause major issues in polymer recycling. For example, if contaminants, legacy materials or unknown substances are incorporated into recycled plastics, this may prohibit their re-use in food packaging or consumer goods.**

What is the impact of plastics?

The large scale of production of plastic is extremely demanding in terms of resources, energy and economic cost, because the fossil hydrocarbons and energy used in their production are generally not recovered.

It has been estimated that, as of 2015, approximately 6,300 million tonnes of plastic waste had been generated, of which approximately 9% had been recycled, 12% incinerated, and 79% accumulated in landfills or the natural environment. Under a business-as-usual trajectory of production and management, approximately 12 billion tonnes of plastic waste will accumulate in landfills or the natural environment by 2050.[2]

The reason why only a relatively small percentage of plastics are recycled is due to the economical challenge particularly for packaging applications where the material is normally designed to be used only once.

Globally, over 40% of the 80 million tonnes of plastic packaging used every year is discarded. The vast majority (approximately 32%) escapes collection or is dumped illegaly.[1,13]

Effective containment of waste depends on capital-intensive infrastructure and services only available in high-income countries. Lower-income countries often lack waste management infrastructure and business models for reuse and recycling. Consumers resort to open-burning of waste, or disposal in unmanaged dumps, from which wastes may be dispersed by wind and rain through land and marine environments.[14] Figure 4 illustrates the proportion of plastic packaging that leaks into the environment globally.

In Europe, more than 40 years after the launch of recycling programmes accompanied by the well-known recycling symbol, only 32.5% of plastic packaging is recycled, while 24.9% ends up in landfill. An additional 42.6% is incinerated with partial energy recovery processes, mostly in mixed solid waste incinerators.[3]

Packaging plastics, particularly in food-contact applications, potentially represent a contribution to public health hazards.[31] The increasing amount of plastic leaked into the environment has given rise to both health and environmental concerns.

Dr Arturo Castillo Castillo (Centre for Environmental Policy, Imperial College London) works on analysis of the causes of waste generation, and on the development of technical and societal interventions to address plastic pollution, including 'reduce' and 'reuse' approaches.

Plastics in the Environment

The impacts of plastic pollution on the environment over the last fifty years have become a global issue that is largely irreversible.[15] The poor recyclability of certain plastic products is an important aspect that can be linked to their leakage into the environment.

Due to the sheer scale of global plastic pollution, it is increasingly difficult to fully quantify its effects on the environment.[14]

Plastic pollution can range from relatively large pieces, termed *macroplastic*, to small pieces in the micro (between 100 nm and 5 mm) and nano (less than 100 nm) scales. Plastic waste has profound implications for marine and land-based wildlife. Plastic creates physical hazards (e.g. discarded fishing nets entangling marine animals) and ingestion can lead to starvation and death caused by blockage or damage to the gut.[16,33,17]

Further prominent effects of plastic pollution include blockage or entanglement of propellers, gears or air intakes in ships. This impacts the shipping, fishing and leisure sectors.[5 16 18]

An impact which is under-researched, but potentially very large, is the effect of plastic particles in reducing the flow of oxygen within soils and sediments affecting functions and health of plants and soil microorganisms.[19]

Health

The health impacts of plastics are not fully understood. Human exposure pathways to microplastics and their chemicals vary, and occur in parallel with exposure to other chemical substances. However, exposure through consumption of invertebrates and crustaceans is well documented. [36,20,21,27] Other sources of exposure include

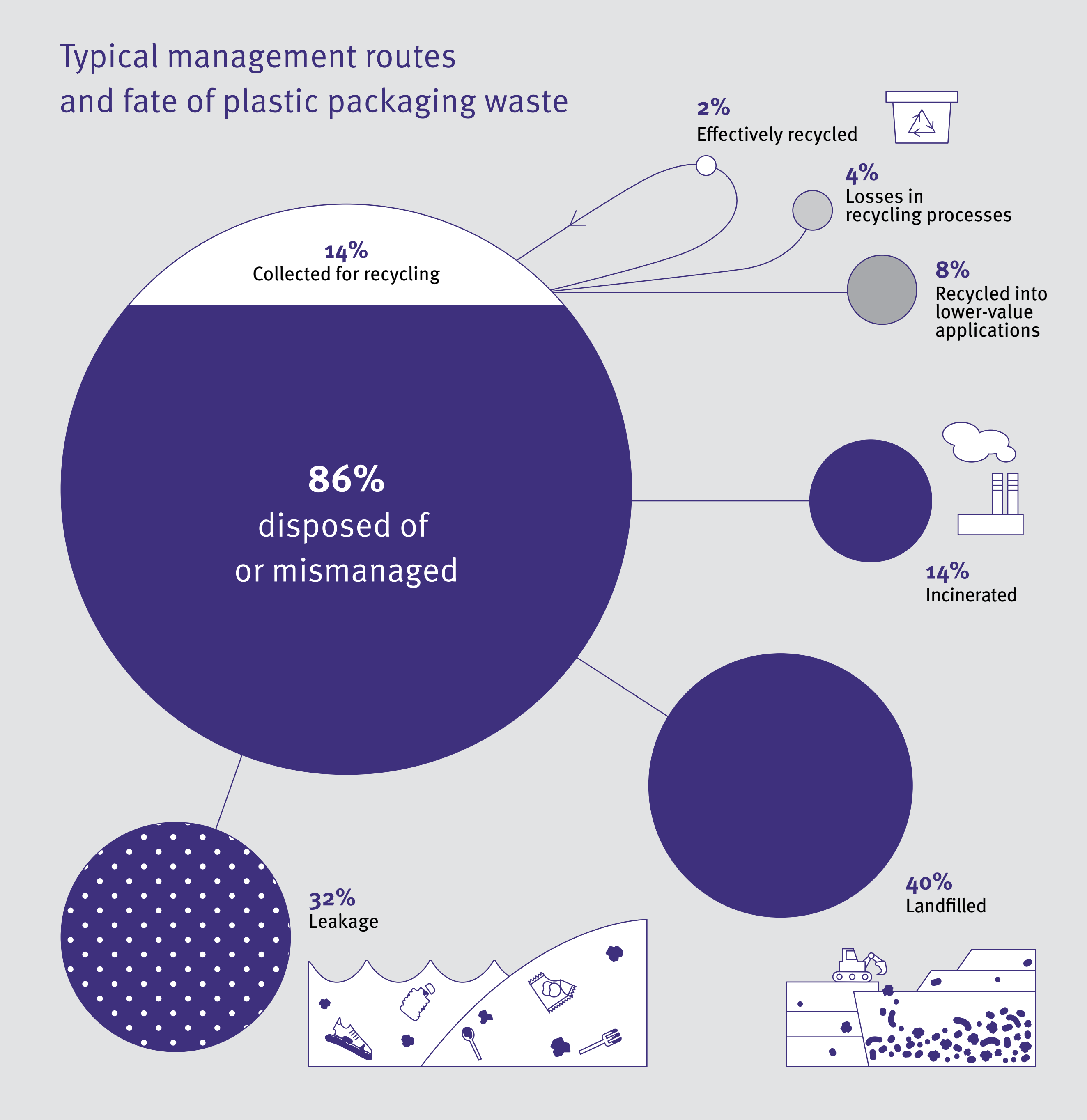


Figure 4: Waste packaging recycled, disposed of or lost to the environment globally.[1]

Box 1: Microplastics

Microplastics, defined as plastics less than 5 mm in size, are found in a wide array of applications from cosmetics to toothpaste. Although they are starting to be phased out in some countries, microplastics are still widely added to some products during manufacture. They can also form from the break-up of larger plastic debris in the environment. Detection and removal of microplastics in the environment is significantly more challenging than for macroplastics. Microplastics are easily ingested by wildlife due to their small size. This can cause damage to wildlife as both intentionally and unintentionally added chemicals are adsorbed onto the plastic.

Microplastics can leak out of wastewater management systems into aquatic bodies, and from there can enter the food chain.[20–22] Micro- and nano-plastics can either pass through or accumulate in the gut of animals. Migration from the gut into gills and liver can occur, indicating that transmission of these particles from the digestive into the circulatory system is possible.[23] Both micro- and nano-plastics can be exchanged across cell membranes due to their small sizes.[24–26]

Recently there has been an increased awareness of the effects of plastic accumulation in the environment and its adverse effects on the planet. Rinse-off primary microplastics used, for example, in skincare and lifestyle products are being phased out in multiple countries. However, the quantity of secondary microplastics in the environment will continue to grow as macroplastic waste breaks up.

Box 2: Effects of feedstocks and additives on health

One aspect of plastic pollution that has not yet received full consideration is the effects of the chemicals used in production at the polymer and product stages. The risk that such chemicals pose depends on concentration and exposure levels. Some hazardous chemicals, some of which are potentially carcinogenic, have been phased out of use as additives during processing or conversion. However, the effect of some additives in polymer production and their potential migration as contaminants or residues is not yet fully understood.

For example, a growing number of studies indicate that use of monomers and additives such as bisphenol A (BPA) and polychlorinated biphenyls (PCBs) correlates with health effects such as obesity, diabetes, reduced brain development, inflammation, and neurological disorders such as early-onset senility in adults and reduced brain development in children.[14, 29] Exposure routes include canned food, dental sealants and cashier receipts.[30]

It is typically accepted that small traces of the chemicals used in production of the bulk polymer remain in the product. These non-intentionally added substances (NIAS) are usually impurities from starting materials, additives or reaction by-products, or result from migration across materials in close contact (e.g. in packaging applications). Their concentrations and exposure levels are low in comparison to quantities that would result in toxicologic effects. However, this must be considered in light of recent evidence that plastic exposure, concentration and accumulation in animals can increase over time, resulting in adverse health effects. Furthermore, food packaging is estimated to be the most relevant source of human exposure in this regard.[31]

bottled and tap water; the original source water could be contaminated by wastewater effluents and terrestrial run-offs containing microplastics.[24]

While the human digestive system has been reported to remove more than 90% of ingested microplastics by excretion,[27] the possible migration and deposition of microplastics into the body, and their effects over time, are yet to be fully understood.

Microplastics have been detected in the atmosphere. When inhaled, these can induce or exacerbate immune responses. Additives in microplastics (see below) can also accumulate in the body, in particular in the lungs and the circulatory system, and can cause chemical toxicity.[28]

Furthermore, the incineration of mixed plastics, particularly when including PVC, releases harmful pollutants such as sulphur dioxide, oxides of nitrogen as well as dioxins and furans, which can have negative health effects if adequate pollution controls are not in place.[13] The resulting by-products captured in pollution control systems need to be disposed of appropriately.

It is therefore urgent not only to gain a wider understanding of the health risks from the use of plastic but also to address the health consequences of making and disposing of plastics. Creating plastic products that are more reuse- and recycling-friendly through novel material combinations, processing methods or overall design can make a significant contribution to overcoming this challenge.

Professor Alexandra Porter and her team from the Department of Materials are using their expertise in electron microscopy techniques to study how nanomaterials and microplastics interact with cells.

How we currently recycle plastic

Mechanical recycling

Mechanical recycling is the process whereby plastic is shredded into smaller pieces, typically 1–3 mm, followed by melting the polymers, which are then extruded into recovered pellets, which are sold. This is currently the main route for those plastics that are widely recycled.

How it works

As shown in Figure 5, the steps involved in recycling include:

- The plastic is initially sorted by size at Material Recovery Facilities (MRFs) using large rotating drums with perforations called trommel screens.
- Wind sifting is used to separate lighter items using a jet stream of air.
- Plastics with different density are separated by passing them through a water tank where those with low density float and those with high density sink ("sink-float separation").
- At advanced recycling facilities, sensor-based sorting techniques are employed to further separate the waste materials.
- Near infrared (NIR) sorting separates items made from different polymers based on the reflection of an infrared beam. This does not work for black plastics due to the absorption of the infrared signal.
- The material response is received by a calibrated sensor for classification; sorting is achieved using compressed air jets across the conveyor belts to blast the identified materials into type-specific streams for different polymers.

Issues

- Although commonly recycled items such as bottles and trays are recovered in this manner, NIR sorting cannot be applied to sort Multi-Layered Plastic (MLP) (see page 10) as only the polymeric layer facing the sensor can be identified, which often leads to erroneous classification and contamination of sorted streams.
- Many mechanical recycling processes lead to some deterioration of the material, for example discolouration or loss of strength, contamination with food residue, or cross-contamination with other polymer types.[13] Currently, post-consumer recycled plastics can rarely be used to make the same original product. It is more common for recycled plastics to be used in lower-grade applications such as garden furniture and traffic cones.[32,33] The terms *downcycling* and *cascade recycling* are used to describe a large proportion of mechanical recycling output.
- In an intentionally or unintentionally mixed plastic waste stream, the phase separation of immiscible polymer blends (polymers that do not mix) may negatively affect the properties of the recycled material. The interfaces between immiscible polymers are sharp and mechanically weak, resulting in a low-grade product.[34–36]

Current challenges to plastic recycling

Waste management systems

Sorting single-use plastic packaging collected from post-consumer and some business-to-business streams is costly, time- and transport-intensive, and the material variation resulting from disparities in formulation at the production and conversion stages (or from consumer separation errors) reduces the quality of recycled plastics.[37,39]

In the current material flow system, it is the norm for significant waste to arise as a basic feature of production and consumption. This means that:

- Affluent societies rely heavily on waste management systems to capture plastics in waste streams.[38]
- Collection, treatment (e.g. incinerators with partial energy recovery), disposal (e.g. landfills that enclose mixed materials) and recycling infrastructure are necessary to either safely dispose of waste or to recover some valuable materials.

Therefore, the viability of recycling and reuse of plastic, particularly packaging waste, faces several challenges:

- The logistical costs of collecting plastic waste in small quantities from households for consolidation and reprocessing affects the viability of recycling compared to centralised virgin resin production.[39]
- In addition to contamination with food or other impurities, the variety of polymers and compositions across products leads to incompatibility and low quality recovered materials.[40]

The recycling of plastic products with good recoverable value and comparatively predictable composition, such as PET drinks bottles, takes place via schemes with some level of market coordination.[41] However, the same coordination has not yet been feasible for MLP due to the potential for cross-contamination between incompatible polymer layers, its low density, the difficulty in sorting MLP products, and its low recovery value.[42]

A significant barrier to preserving material value in circular flows through the economy is the shortage of policy and business models that consider material recovery or product reuse from the outset.

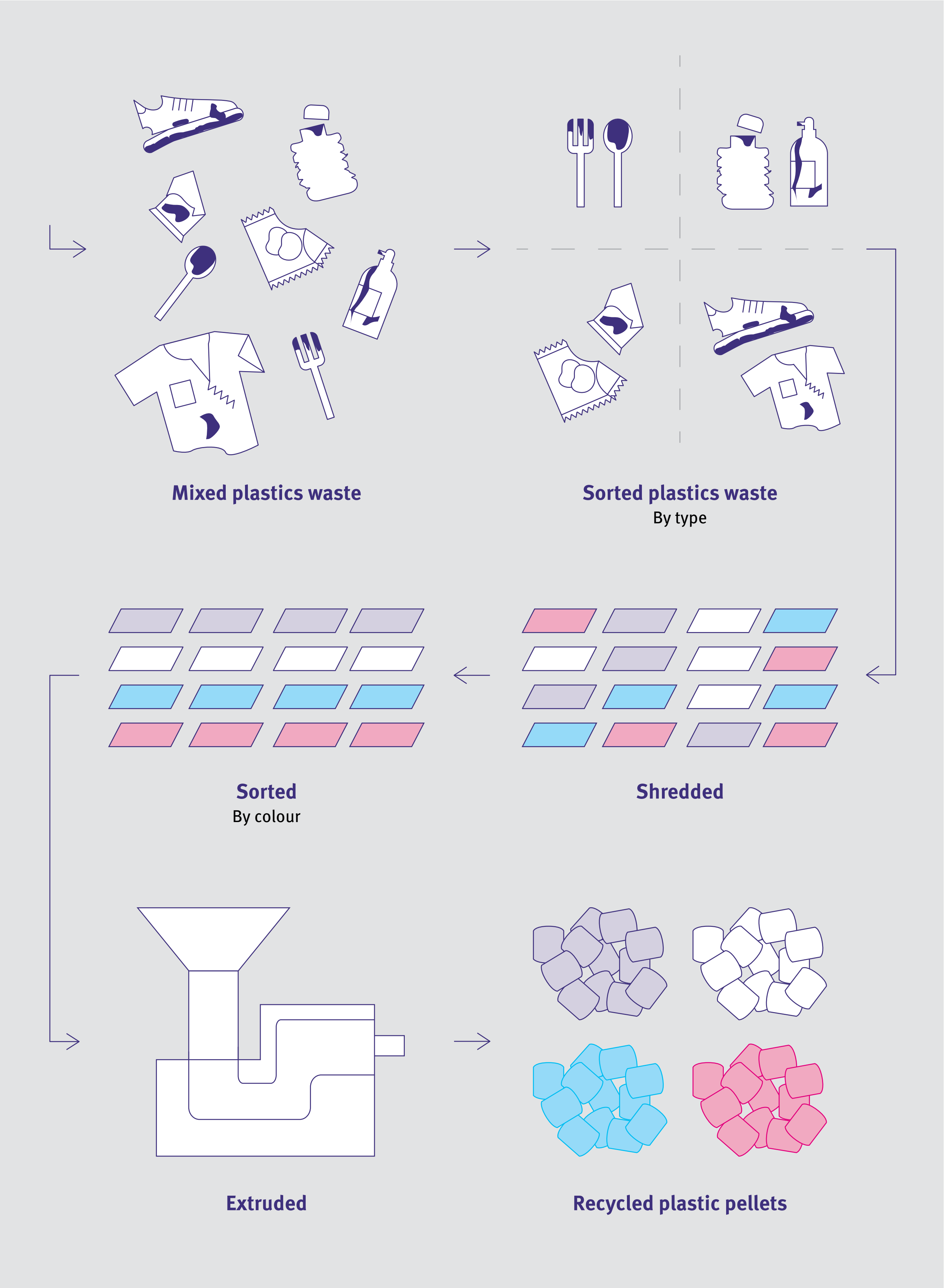


Figure 5: Steps in the mechanical recycling process.

Multi-Layer Packaging

Multi-Layered Packaging (MLP) is essentially layers of different materials, mostly plastics, arranged in a laminate form to offer a multitude of functional properties in a single packaging solution. Common examples of MLP include crisp packets and confectionary wrappers.

MLP provides different functions depending on the product and the intended market. Typically, the different layers provide mechanical support; essential barrier protection from oxygen, water vapour and sunlight; printability for branding and sealing to form a closed environment around the product. The physical properties of each layer are typically not suitable to perform the function of other layers, creating the need for Multi-Layered Packaging. Figure 6 shows a typical construction of MLP. The adhesion between the layers is critical to MLP functionality and various interlayer adhesion strategies such as dedicated adhesives or blends of adjacent polymers are employed.

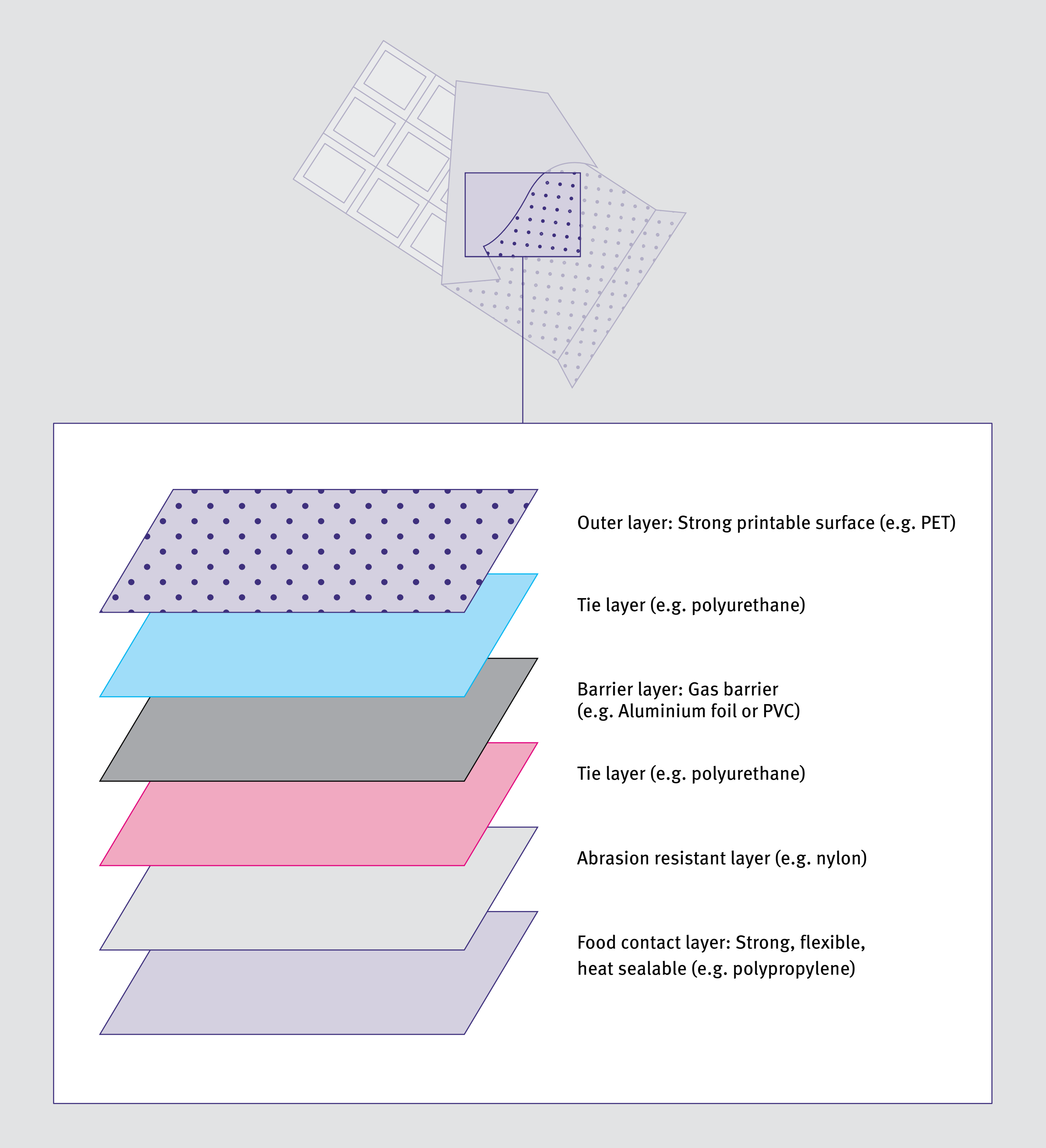


Figure 6: Typical construction of Multi-Layered Packaging.

Increasing consumer preferences towards additive-free, fresh foods with increased shelf-lives have contributed to an increased use of MLP by manufacturers in recent years. Whilst a trend towards avoiding hard-to-recycle material combinations is observed, it is unlikely that single polymer packaging by means of a new 'super-polymer' will address all packaging requirements. Thus there is an urgent need for alternative solutions to re-use or recycle MLP.[43,44] To address this issue, newly re-designed manufacturing routes to existing plastics, using benign materials, less toxic inputs, and inherently recyclable components must be developed.

Among packaging plastics, flexible plastics and MLP are currently not economically recyclable. The end-of-life options for flexible packaging, MLP and mixed plastics are mainly incineration or landfill. Since the main application of MLP is to provide essential barrier and mechanical properties in the food and pharmaceutical industries, direct replacement with more recycling-friendly materials whilst maintaining performance is often challenging.

Design guidelines for recycling-friendly MLP can be formulated by directly addressing the factors that limit recycling. These include: the difficulty in collecting low-density material, handling challenges whilst sorting, and low-value final recovery. The incorporation of digital watermarks and chemical tracers has been investigated to help overcome sorting challenges.[45,46] Non-recyclable, hard-to-recycle, and off-specification novel polymers must be avoided as they often contaminate recycling streams at MRFs. Minimising the variability of polymers used in flexible packaging could also improve the recycling prospects of MLP, as this aids reliable separation. Although un-dyed, mono-material streams represent maximum value for recyclers, the strong adhesion between MLP layers leaves co-processing of constituent polymers and layers as the only practical solution to date.

Box 3: Recycling brand identity

Brand identity is expressed through strong colours to attract consumer attention. Processing different pigments typically results in a dark greyish-green recyclate (product from recycling), for which there is limited demand in recycling or re-use. Although it is technically feasible to remove some inks from the packaging during recycling,[47–49] the cost typically outweighs the financial benefits of producing transparent recycled materials.

In future, water-based primers could improve resource efficiency for printing. De-inking of water-based primers can be performed in surfactant solutions; multiple options are reported to this effect.[47,49] Alternative concepts such as separate sleeves, stuck-on labels or easily separable layers should be prioritised over direct printing on packaging.

The compatibility of different polymers for combined processing has been well studied. Most design-for-recycling guides refer to compatibility charts that describe the level of compatibility among polymers during recycling. MLP configurations must be developed in accordance with such compatibility guidelines if the layers cannot be separated prior to recycling.

When layers can be separated, design-for-recycling guidelines to aid sink-float separation are applicable for MLP. Laminate polymers should have a sufficiently different density to either sink or float (*e.g.* the HDPE and PET used for the caps and body of water bottles, respectively). Essentially these guidelines enable packaging designs to reduce production costs and to maximise value of recyclates.

Box 4: Attitudes and behaviour

A significant challenge to the reuse of plastic products and components is the perception that they have been used by others. This phenomenon is known as *contaminated interaction*, whereby people are reluctant to use something that has been worn, held or personally used by unknown individuals, and is perceived as not clean, impure or of lower value.[37]

Rethinking plastic recycling

Better policy and guidance

Multiple strategies need to be developed to address the consequences of the take-make-dispose pattern. An important challenge is the low cost of plastic products. Integrating recyclability and sustainability into the product design phase is a necessity in order to reduce waste and increase resource efficiency. Several industries are introducing directives aimed to enable maximum material recovery and recycling through product design. The European Union's ELV (End of Life Vehicles) and WEEE (Waste Electrical and Electronic Equipment) directives require the products to be designed to enable ease of separation and recovery to meet re-use and recycling targets.[50,51] In some cases well-established design for recycling guidelines exist, such as for rigid plastic packaging and electronics equipment.[52–56] For example, the Recyclability by Design guidelines by RECOUP, a national charity developing plastics recycling in the UK,[52] proposes guidance on some packaging designs.

The high recycling rates achieved for PET drink bottles and HDPE milk bottles can be attributed to such recycling guidelines. These ensure that material choices, colours, additives, closures, barriers and labels are optimised to maximise recycling and to preserve the value of recyclate.

Development of similar guidelines across the entire range of packaging types is essential to address the global challenge of plastic pollution.

These measures are intended to divert materials from managed or unmanaged disposal and enable prolonged resource circulation either through re-use or recycling. Consequently, new product delivery systems combined with improved packaging designs are emerging to facilitate circular material flows. This changes the relationship between products, the benefits they provide and the way they are delivered. These systems are known as product-service-systems (PSS).

Nevertheless, comprehensive applicability of such products and distribution systems to fully address plastic pollution is not yet practicable. Therefore, improving product recyclability from a material, or molecular, perspective within already existing production, distribution and collection systems is essential.

The research of Dr Marco Aurisicchio and his colleagues at the Dyson School of Design Engineering is focussed on the design and analysis of systems, platforms, products and services to improve reuse and recycling of products.

Improving mechanical recycling

Compatibilisation

As mentioned previously, mechanical recycling results in the deterioration of the plastic, and only rarely is recycled plastic used to make the same kind of product as the original material. To mitigate this, appropriate block copolymers[a] called *compatibilisers* have been developed. These act as “molecular stitches”; they are combined with otherwise immiscible plastics to create a blended product. Compatibilisers work by transferring mechanical stresses across the interface between different polymers, to improve the mechanical properties of the blends, thereby making them more processable as a single material.[35]

The compatibilisation of polymer blends can be achieved by two means:

- Adding compatibilising chemical components (typically either a block or a graft copolymer[b]) to increase miscibility at the interface in an additive, *non-reactive way*; and
- Through *reactively synthesising bonds* in-situ across the contact interface during blending.

The teams of Professor George Britovsek at the Chemistry Department of Imperial College London, in collaboration with Professor Maria Charalambides of the Department of Mechanical Engineering, are currently investigating the development and application of compatibilisers.

Professor Maria Charalambides and her research team in the Mechanics of Materials division in the Department of Mechanical Engineering are developing material models to complement polymer blending and compatibilisation. These predictive modelling tools will inform better polymer compositions and compatibiliser choices to maximise material performance for particular applications.

Upcycling and repurposing

Another approach to improve the mechanical properties of mixed plastic recyclates from waste of varied compositions is to make reinforced composites. Upcycling of recovered polymers through integration of fibrous reinforcements or high aspect-ratio fillers takes advantage of high modulus reinforcements to improve the final properties compared to those of recycled mixed polymers alone.

This approach has been expanded to process mixed plastics wastes and a range of fibrous reinforcements, especially natural fibres.[57,58] The balance of improving mechanical properties without significantly altering processing performance of the polymers is crucial in recycling. For example, integrating sugarcane bagasse microfibres into a recycled high density polyethylene matrix as a potential construction material results in an increase in compressive modulus,[c] without significantly affecting the melting and crystallisation temperatures of the polymer.[59] Adhesion of hydrophilic natural fibres and hydrophobic polymers can be a challenge. Some properties of the individual materials may be preserved or reduced when the materials are integrated. Typically, this is addressed through the use of coupling agents to improve properties such as impact strength and tensile strength.[60,61]

Whist upcycling through reinforcing is certainly an interesting prospect, high-modulus fibrous reinforcements, such as carbon fibres, are often required to compensate for the inferior properties of recycled materials of unsorted origin. Even though this composite approach can enable an additional use cycle for recovered polymers, sorting and processing of reinforced composites within existing recycling infrastructure could be challenging. Therefore, the composite approach to upcycling or high-value repurposing is more

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[a] A block copolymer is a polymer comprising molecules with a linear arrangement of portions, called blocks, which differ in constitution from other adjacent portions, i.e. they contain a different monomer. See Figure 1.

[b] Grafted copolymers are a branched polymers in which the side chains are structurally different from the main chain, or consist of a different monomer to that of the main chain.

[c] Resistance to mechanical stress without loss of original dimensions or properties.

suitable for target applications that require durability, rather than as a broadly-applied approach.

Dr Koon Yang Lee and his research team at the Department of Aeronautics are developing composites to re-use, recycle and upcycle mixed waste plastics.

Novel reagents and delamination

The inability to separate polymers, for example in MLP, is a major limitation in using existing sorting and recycling infrastructures. Separation of the layers expands the options available for recycling, and widens the scope of blending strategies.

Compatibilisation can be used alongside combined melt-processing to effectively recycle exclusively polymeric packaging. However, packaging materials in the food and healthcare industries often include metallised layers bonded to the polymer layers, which cannot be effectively processed by compatibilisation.[62] Furthermore, metals stay in solid form when the polymers are melted into liquid form, thus may obstruct or damage polymer processing equipment.

There are options to recover the metal, whether laminated foil or metallised polymer, using novel reagents or solvents; however, the polymeric backing web is often not recovered. Applying the same reagent strategies to MLP directly would result in the undesirable loss of all the polymers in MLP. Therefore, separating just the layers containing metals from the MLP laminate is desirable to maximise available resources for recycling.

Whilst several layer separation strategies exist, such as using special inter-layers or controlling the level of adhesion between the layers, these have not transitioned into large-scale solutions to improve the recyclability of MLP laminates.

These strategies can be extended to all the layers in MLP, and they are essential for materials that pose the greatest difficulty to recycling, such as metallic layers and pigments. The main obstacles to effective exploitation of these strategies are the lack of, or disregard for, design and material guidelines to inform MLP packaging designs with improved recyclability. **Therefore, manufacturers should be further encouraged to consider overall recyclability of products and packaging, and to adhere to 'design for recycling' initiatives for MLP that preserve the greatest value materials through recycling.**

Chemical recycling

Chemical or feedstock recycling is defined as the process leading to the decomposition of plastics to their building blocks, i.e. monomers, or other valuable low molecular weight fragments.[63]

How it works

The term 'chemical' is used, as the polymer undergoes a chemical alteration or transformation. The obtained monomers can subsequently be re-polymerised to produce the original polymer, thus closing the industrial material loop.[64] The key for this type of strategy is to use the polymer or plastic as a starting reagent and generate high purity monomer using simple and efficient transformations (mainly through efficient catalytic strategies) which do not compromise the economics of the overall approach. Partial de-polymerisation to oligomers (compounds of a small number of monomers) or mixtures of other hydrocarbon compounds are also valuable; these materials may be used as feedstocks or input for the production of new plastics and petrochemicals by means of heat or chemical agents.[65]

Chemical recycling attracts significant attention because it has the potential to become profitable and improve sustainability; it reduces the demand for energy and feedstocks, and the material flow is cyclic. Of course, much research and optimisation of the current technology is still required in order for chemical recycling to become established as an industrial norm.[66,67]

The two major approaches to chemical recycling are:

- **de-polymerisation** – the inverse of the polymerisation reaction, usually mediated in organic or aqueous media under optimised temperature and pressure conditions with the aid of a catalytic system; and
- **pyrolysis** or thermal cracking (*thermolysis*) in which bulk plastics are subjected to high temperatures in the absence of oxygen.[46]

De-polymerisation

One strategy that has gained significant attention is the *de-polymerisation* of polymers to allow the recovery of high-value small organic molecules. This chemical recycling strategy generally aims to retrieve the monomers in polymerisation grade purity. This allows the recovered monomers to be re-polymerised into materials with the same quality and functionality as the original, with the possibility of becoming other high value products.

The teams of Professor Jason Hallett in the Department of Chemical Engineering and Dr Agi Brandt-Talbot in the Chemistry Department are currently working in this field.

The de-polymerisation approach is preferentially applied to *condensation* polymers, which release by-products such as water or methanol during their synthesis. These include polyesters (PET, PBT, PC, PLA), polyamides (aliphatic and aromatic nylons, polyimides), formaldehyde resins (phenol–formaldehyde resins, urea–formaldehyde resins), polyurethanes (polyurethane rubbers and foams, spandex fibres) and other polymers (e.g. epoxy resins) (Figure 7).

Different reagents can be utilised leading to various forms of chemical decomposition or *chemolysis* reactions.

These reactions provide a wide variety of small molecules from a single plastic or polymer. PET is the most prominent example, with a myriad of studies describing the chemolysis of the material.[64,65,68–75] An emerging candidate of solvents/catalysts for this procedure are *ionic liquids* (ILs).[76–78] These substances are catalytically active solvents for many otherwise insoluble polymers, including existing and novel thermosets, compatibilisers, and biopolymers such as cellulose. ILs significantly broaden the quantity of plastic that can be recycled, especially plastic contaminated with dyes and additives. Due to their ionic nature, they also have high affinity for many metal ions contained in polymerisation catalysts (e.g. zinc) that are liberated when plastics dissolve.

Several extensively-used polymers can be recycled via de-polymerisation. Nylon-6, the most popular nylon grade, is a polymer synthesised from the polymerisation of caprolactam and is one of the very few polymers for which a closed-loop chemical recycling process is already in place on an industrial scale.[1,79] PLA, another commodity bioderived plastic, which is mainly used for disposable coffee cups, has been shown to degrade to smaller useful organic precursors with high yield and purity, using non-toxic organic bases as catalysts.[80] The de-polymerisation of PC can be almost quantitative, and can lead to the synthesis

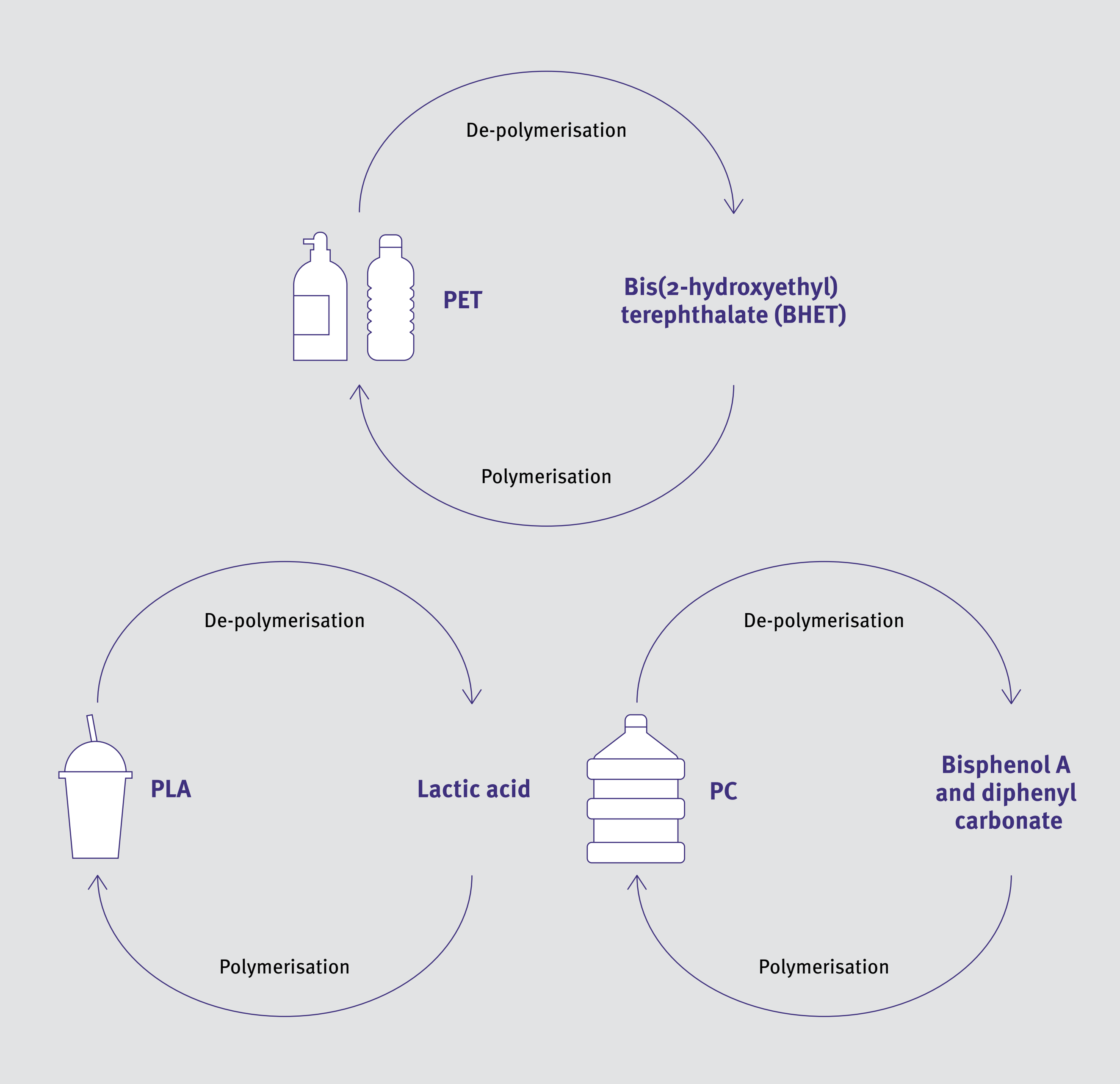

Figure 7: Graphical depiction of examples of closed-loop chemical recycling of modern plastics.

of a wide pool of useful starting materials (monomers) for different applications.[81]

Pyrolysis

The second approach for treating post-consumer plastics relies on their direct conversion into high calorific value fuels and platform chemicals through *pyrolysis*.[67,82–84] This is the process of thermally degrading long chain polymers synthesised via an *addition* polymerisation process into smaller, less complex molecules through heat and pressure. These plastics are mainly polyolefins[d] such as PE, PP, PS, PVC, polybutadiene, polyisoprene; acrylics such as polymethacrylates, polyacrylamide, polyacrylonitrile, and other vinyl types such as poly(vinyl acetate), poly(vinyl acetals) and poly(vinyl ethers).

Non-catalytic or *thermal* pyrolysis is a process that requires intense heat (temperature range 300–500°C, varied depending on polymer type in order not to produce gaseous waste) with short duration and in the absence of oxygen (to avoid incineration).[83]

The three major products of pyrolysis are oil, gas and char, which are valuable to the industrial sector. For example, the liquid oil from catalytic pyrolysis can be used in several energy-related applications such as electricity generation, transport fuel and heating source. Moreover, process by-products, such as char, could be used as an adsorbent material for the removal of heavy metals, pollutants and odour from wastewater and polluted air, while the gases produced could be used as energy carriers.[84]

Pyrolysis of different types of plastic has been carried out extensively in the past, including PE and PS. Only a few studies have focused on PVC, PET, PMMA and PU pyrolysis. Thermal pyrolysis of PS is easily carried out in comparison to HDPE, LDPE and PP, as they require higher temperatures for degradation. Furthermore, in the absence of a catalyst, PE is converted into wax instead of liquid oil.[85] Thermal pyrolysis oil contains large carbon chain compounds and has low quality due to its low octane number, the high concentration of solid residues, and impurities such as sulphur, nitrogen, chlorine and phosphorous.

Catalytic pyrolysis is carried out using a catalyst and shows promise for the conversion of plastic waste into liquid oil with improved quality at lower temperatures and shorter reaction times compared to thermal pyrolysis. These factors can reduce the process energy requirements and optimise overall process efficiency.[86] Nevertheless, despite all the potential advantages of catalyst-enhanced pyrolysis, some limitations such as high process energy demand, catalyst costs, and low reuse of catalyst remain. The recommended solutions for these challenges include exploration of cheaper catalysts, catalyst regeneration, and overall process optimization.[84]

Improved packaging

Better use of additives

The effect of additives and adhesives on re-use and recycling processes must also be considered during design. The chemical and thermal stability of additives should be suitable for combined melt-processing in the recycling stage as this is currently the dominant route. Essentially, components that result in low performance of recyclates – such as excessive pigments, thermosets, metal foils and metallised films in the case of MLP – are being targeted in research for substitution and possible displacement. Where these are unavoidable, new developments must integrate them into MLP in a way that allows them to be easily separated without compromising functionality. Design solutions can be applied to layers containing print and metals to keep them separate from plastic recycling, to maximise the value of material recovered from MLP.

Implications

Around 26 million tonnes of plastic waste are generated in Europe every year.[87] Unfortunately, only *ca.* 30% of that amount was collected for recycling in 2018. Of this portion, a significant share is exported outside of the EU to be treated in countries where different environmental standards may apply.[87] The growth in exports of recovered plastics largely reflects the strong demand from lower- and middle-income countries (LMIC), where manufacturers' demand for polymers has grown at an average annual rate of almost 20% over the past few years. At the same time, in Europe, landfilling and incineration rates of plastic waste remain high, at 31% and 39% respectively, and while landfill has decreased over the past decade, incineration has been constantly increasing. According to estimates, 95% of the value of plastic packaging material, i.e. between €70 billion and €105 billion annually, is lost to the economy after first-use.[1] Demand for recycled plastics today accounts for only around 6–8% of plastics demand in Europe. In recent years, the EU plastic recycling sector has suffered from low commodity prices for virgin resins and uncertainties about market outlets. Investments in new plastic recycling facilities and methodologies have been hampered by the low projected profits.

Virgin polymers are the materials produced from petroleum derivatives, after the most commercially useful components, such as fuel for transport and heating applications and natural gas, have been extracted. Prices for processed recovered polymers are closely correlated with prices for virgin PET and HDPE. Indeed, market reports suggest that

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[d] Polyolefins are polymers produced from a simple olefin, also called an alkene, which is a hydrocarbon that contains a carbon–carbon double bond. For example, polyethylene is the polyolefin produced by polymerising the olefin ethylene; polypropylene is produced by polymerising the olefin propylene.

recovered polymers are traded at a significantly lower price compared to the pristine new material (Figure 7). Virgin polymer prices are one of the key determinants of recovered plastics prices.[88]

The most common route for recycling recovered plastic is for it to be used in a different application from the source product. This is known as repurposing, 'open-loop' or 'cascade' recycling, although 'closed-loop' routes, such as bottle-to-bottle recycling, are gradually being developed. The main end products for recycled HDPE include pipes, pots, crates, and other moulded products, while recovered film is turned into sacks, bags, and damp-proof membranes. The majority of recovered PET is used in the polyester fibre industry, although there is gradually growing demand for closed-loop packaging of PET.

Where they can be freely substituted for one another, the demand for recovered plastics is determined by the residual demand unsatisfied by virgin polymer supply. **Recovered plastics are, in some cases, imperfect substitutes for virgin materials owing to inferior quality and mechanical properties.** However, this is changing as closed-loop recycling technology (mostly through *chemical recycling* pathways) becomes established, and as the advertised recycled content in plastic products becomes a marketable tool, for example for corporate social responsibility purposes. The strength in virgin polymer prices over the past 15 years has, until recently, supported the development of the UK's plastics recovery infrastructure as a cheaper alternative. The outlook for polymer prices has tended to remain strong, aligned with polymer demand, particularly in LMIC, and is expected to continue to grow. However, significant international expansions in the petrochemical industry are expected in the next few years. Although some of this will replace a large portion of the retiring European manufacturing capacity, a sharp increase in supply could affect virgin polymer prices. If prices for recovered plastics were also to fall, this could reduce commercial incentives to further increase plastics recovery.

Rethinking plastic manufacturing

Biodegradable plastic

Current biodegradable plastics

One strategy developed in the past two decades to improve sustainability is the use of **biodegradable polymers**, for example PLA, PBS and PHB. These have a wide variety of applications, especially for short-term usage products such as packaging materials, foils, and utilities in the agricultural sector.[89,90] Currently however, adequate, industrial-scale biodegradation treatment of any mass-produced biodegradable plastics has not achieved a significant market share.

Box 5: Degradable and high-performance

Replacing plastics employed in single use packaging with renewable and degradable alternatives is attractive from an environmental perspective. However, the majority of synthetic polymers are designed for performance and durability, not for degradability and recyclability, which has resulted in significant growth of disposed and unrecovered polymer wastes over the past few decades.[97–99] This is because **it is difficult to design materials that are both degradable and high-performance.** Resistance against chemical deterioration is one characteristic that helps to define a high-performance polymer. In many applications, the terms *degradable* and *high-performance* are considered to be mutually exclusive.

Where biomass from plants is used as the renewable raw material, the polymers are referred to as *bioderived* or *biobased*.[91] It is important to recognise that **some petrochemical polymers are biodegradable, and that not all bioderived polymers will biodegrade.**

Biodegradable polymers are mainly derived from renewable sources (e.g. starch and cellulose) and can be degraded by enzymes or hydrolysis[e] into simpler building blocks, such as CO_2, water, methane, and organic soil components, leading to a closed circular system.

To date, significant advances have been achieved in the synthesis of biodegradable polymers *via* organometallic, organic and enzymatic catalysis,[f] leading to a significant scientific and industrial interest.[92–95] Although biodegradable polymers are often considered environmentally friendly polymers, their degradation profile is economically and environmentally suboptimal at present, as they usually end up in landfill, where the actual material value is lost. This is because compostable polymers are only a small subset of all commercial polymers, and only a small fraction of compostable material is returned to composting facilities. Polymers designed to be composted (i.e. decomposed by soil bacteria in the presence of oxygen) such as PLA, may not degrade as planned in landfill. This leads to the formation of potentially harmful substances (e.g. methane gas), depending on the environmental conditions.[96]

[e] Hydrolysis is a chemical reaction in which water molecules are used to fragment chemical bonds.

[f] Degradation can be achieved using organic compounds or enzymes as catalysts, or, in the case of organometallic catalysis, chemical compounds containing bonds between an organic molecule and a metal.

Although the creation of bioderived durable plastics will help address consumer interest in renewable products, these materials suffer from the same end-of-life issues as traditional petroleum-derived plastics. Although some bio-based compostable polyesters such as PLA are now being commercialised, the development of this industry has been hindered by a lack of a composting infrastructure.[13]

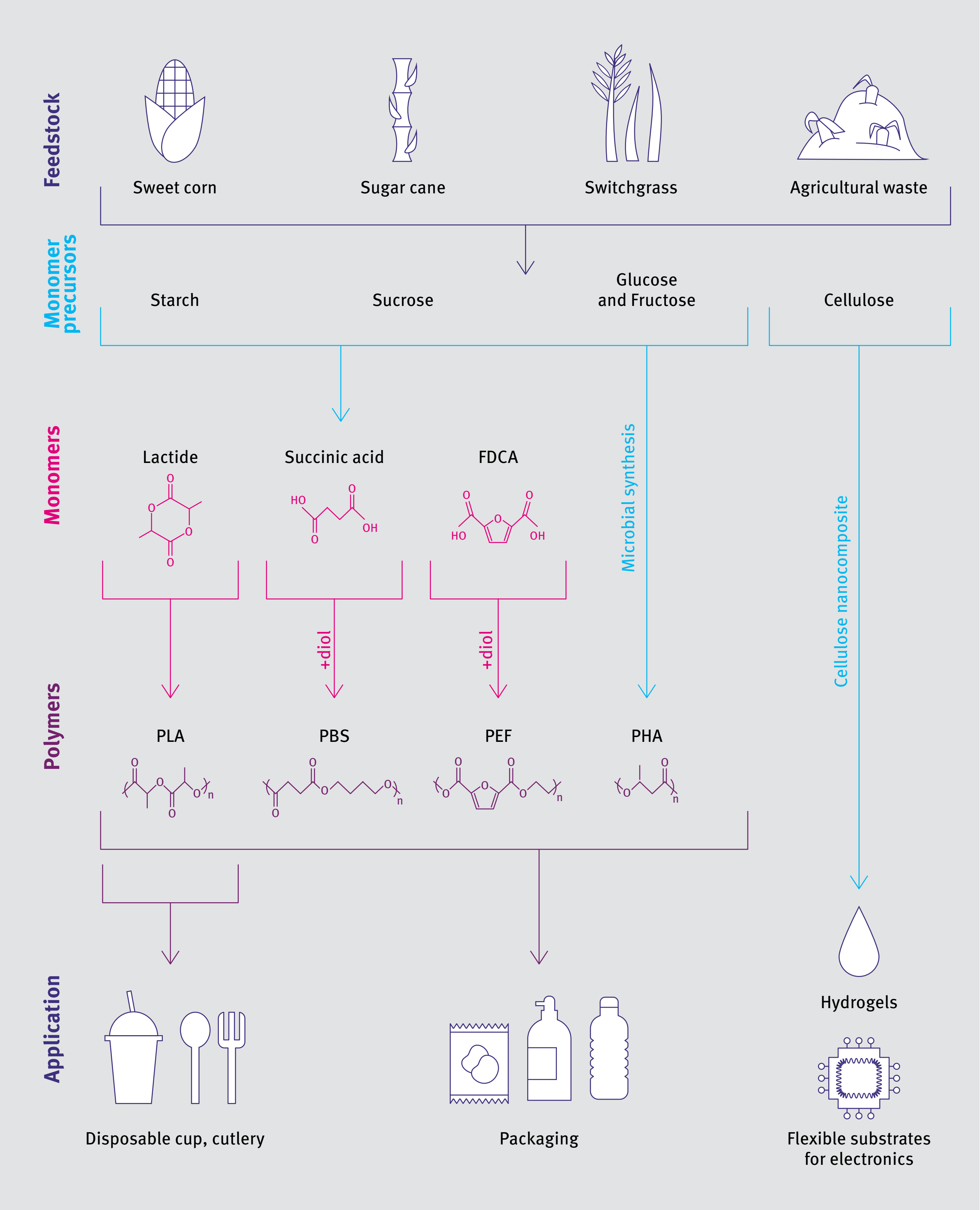

Figure 8: Possible polymers synthesised based on bioderived sustainable monomers.[91]

The future of biodegradable plastics

Considering the diversity of biomass, and the large pool of organic compounds this encompasses, a large number of alternative **bioderived monomers and novel polymers** are potentially accessible (Figure 8).[100,101] The earth produces approximately 170 billion tonnes of plant biomass every year, although only 3–4% is currently used by humanity.[102] It is estimated that, within 15 years, up to 30% of all chemical feedstocks will be derived from plant biomass.[103,104]

Lignocellulose production does not compete with food production and is readily available from various waste streams, for example from the forestry and paper industries. **Therefore, lignocellulose is likely to be the main renewable source of carbon to substitute fossil hydrocarbons in future.**

Recent work has focused on biopolymers derived from natural sources which are chemically modified to adjust the properties. For example, polyhydroxyalkanoates (PHAs) are produced naturally by bacteria as a form of energy storage.[105,106] PHAs were initially commercialised in the 1980s and were distributed under the name 'Biopol' in the USA; they remain prominent in the bioplastics market.

New Generation plastics

Smart polymers

The commercial success of new renewable and degradable polymers will depend on the development of low-cost **materials that can change their structure**. Ideally, the degradation rate of these materials should be matched to their desired applications.

An appealing strategy is to design so-called **'smart polymers'**. These contain chemically responsive switches that can be tripped at end-of-life to trigger degradation, or to enable either reprocessing or chemical recycling (Figure 9).

In theory, such approaches could be used for thermosets as well as thermoplastics.[108] Thermosets (cross-linked polymers) represent *ca.* 15–20% of global polymer production.[107] Traditional recycling of these materials by mechanical reprocessing is impractical because their structures prevent flow even at elevated temperatures (they do not melt). Their insolubility also hinders reprocessing using solution-based methods. For these reasons, most thermosets are incinerated, landfilled or unaccounted for. However, their high strength and chemical stability render them essential for use in tyres, engineering composites, foams, adhesives, and many other applications.

Developing strategies to recycle thermoset materials without compromising performance represents a financial and environmental challenge.

Hybrid materials address this challenge by combining two types of polymers: linear or branched polymers are **physically networked** via reversible, physical bonds such as hydrogen bonds or ionic interactions. This represents an interesting class of materials able to adapt to changing

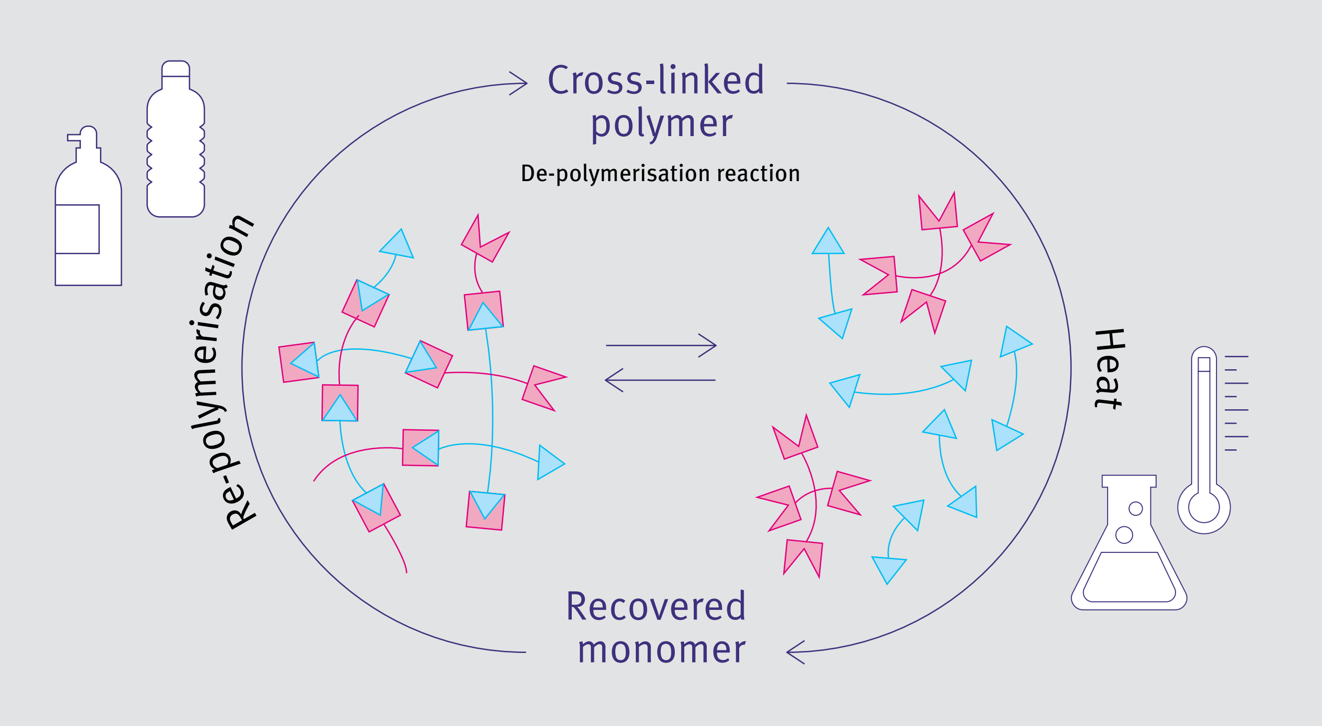

Figure 9: Association and dissociation of dynamically cross-linked thermosets complemented by the stimulus-triggered de-polymerisation possibilities.[107]

conditions, including applied stress during use, depending on the timescale of the stress compared to the lieftime of the physical network. For instance, if the stress is applied for much longer than the lifetime of their structural network, then the material will flow like a thermoplastic, whereas if the stress is applied for a timescale that is much shorter than the lifetime of their structural network, then the material will be elastic and behave largely like a thermoset *elastomer.*[g] The desirable combination of a crosslinked polymer structure with the capability for a triggerable reversion of the crosslinking has recently been named a **Covalent Adaptable Network (CAN)**. These CANs have all the state-fo-the-art mechanical benefits of a traditional thermoset while their unique dynamic chemical bond structures enable them to respond selectively to stimuli applied to alter their structure, properties and shape, thus creating self-healing capabilities and transforming them into recyclable structures.[109] Other aspects of sustainability (renewability and degradability) can also be integrated into their development.

At Imperial, Dr Charles Romain's team in the Chemistry Department is currently developing an exciting version of thermally reversible bio-sourced dynamically crosslinked polymer networks.

Conclusions

Plastic pollution is one of the main socio-environmental challenges that we face today. We are regularly discovering new, worrying consequences of plastic pollution. Plastic production is expected to double again in 20 years and almost quadruple by 2050, therefore it is a necessity that we act now to reduce the impact of plastic pollution on our health and the environment.

Packaging plastics, particularly in food contact applications, represent a potentially substantial contribution to public health hazards, while plastics used in fast-moving consumer goods are hazardous to wildlife and contaminate the food chain. The poor recyclability of certain plastic products, in addition to insufficient waste management and infrastructure, can be linked to leakage of plastics into the environment.

Items with high material value in recycling streams, such as PET bottles, are typically reclaimed from waste streams as there tend to be some financial incentives to recirculate them into the economy. By contrast, there are no established recycling solutions for low-density flexible items.

It is critically important that we not only gain a wider understanding of the health impacts from plastic products, but also address the health consequences of making and discarding plastics.

Making plastic products more reusable and recyclable through novel material combinations, processing methods, overall design – in combination with resource efficiency policies – could make a significant contribution to reducing generation and leakage of plastic waste.

Outlook

Tackling plastic pollution will require a multi-pronged, medium-term strategy that implements the right incentives for each stage of the innovation cycle of both the early-stage and more established technologies described in this Briefing Paper. Market take-up of environmentally beneficial options will require more than the right science and engineering. For instance, early stage chemical recycling research requires further support for R&D. More established techniques, such as mechanical recycling, require a more convergent approach that considers better use of additives and design for recyclability as well as the right infrastructure, coherent guidelines and public participation.

Current research efforts at Imperial College London are contributing to these developments by investigating:

- design and recycling routes for multi-layer plastics;
- the properties of combinations of recovered fibres and post-sorting plastic waste;
- new approaches for preserving the value of monomers through compatibilisation;
- new processes and solvents to enable more repeatable recovery of monomers in chemical recycling;
- nano-scale imaging and analysis techniques to study the potential impact of plastic and fibre particles in the human body;
- systems to increase reuse and resource efficiency;
- environmental and innovation policies to bring all these technologies closer to market.

The molecular science and engineering approach[110] is essential to ensure the success of these research challenges.

A coordinated approach to deploying the materials, technologies and policies discussed in this Briefing Paper has the potential to alleviate the detrimental effects on health and the environment caused by current practices in production, use, recovery and (mis)management of plastic materials.

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[g] An elastomer is essentially a polymer which displays rubber-like elasticity. It can be a thermoplastic, easily mouldable and re-processable, or a thermoset with a crosslinked network structure.

Glossary and List of Abbreviations

CAN	Covalent Adaptable Network
HDPE	High-density polyethylene
ELV	End of Life Vehicles
LDPE	Low-density polyethylene
MRF	Material recovery facility
MLP	Multi-Layered Packaging
MSW	Municipal solid waste
NIAS	Non-Intentionally Added Substances
PBS	Poly(butylene succinate)
PBT	Poly(butylene terephthalate)
PC	Polycarbonate
PE	Polyethylene
PET	Poly(ethylene terephthalate)
PHA	Poly(hydroxy alkanoate)
PHB	Poly(hydroxy butyrate)
PLA	Poly(lactic acid)
PMMA	Poly(methyl methacrylate)
PP	Polypropylene
PS	Polystyrene
PVC	Poly(vinyl chloride)
PU	Polyurethane
RECOUP	A national charity developing plastics recycling in the UK
Recyclate	Product of recycling process
Sugarcane bagasse	The remaining residue after sugarcane is crushed to extract its juice
WEEE	Waste Electrical and Electronic Equipment

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