Closed-Loop Recycling of Polymers Using Solvents

Remaking plastics for a circular economy

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Recycling of plastic is an established technology contributing to a circular economy. A sustainable society requires recycling to produce high quality feedstocks from all types of reusable waste. New recycling technologies will help to improve waste management practices, for instance dissolving plastic waste in a solvent to purify and maintain its material properties. In solution it is also possible to depolymerise polymers into monomers that can be used to remake virgin-grade material. In this review the advantages and disadvantages of three solvent-based recycling processes will be considered: separation of cotton and polyester (polyethylene terephthalate (PET)) textiles, chemical recycling of polylactic acid (PLA) and dissolution-precipitation of polyvinylchloride (PVC). The current state of the art and future prospects are discussed, including a brief overview of how solvents are being used to process other types of plastic waste.

Introduction

There is an obvious and increasing need to preserve valuable resources and reduce waste and pollution. Maximising the functional lifetime of materials with reuse and recycling practices has long-term benefits (1). These themes are embodied in the circular economy concept, where materials are considered in terms of the service they provide when fabricated into products (2). Extended product lifespans deliver more service from a material, while waste represents lost potential.

The EU and China are the two regions with the most prominent circular economy strategies. Specific policies have been established since 2015 in the EU (2) and even earlier by the Chinese government (3). Although the regulatory measures are broad, encompassing critical materials and product (eco)-design, European law focuses on recycling targets. China has additional policies encouraging industrial symbiosis so responsibility for waste is shared, including heat and material (waste) outputs of one industry being provided as the input for another. Academic interest in the circular economy concept is high, ranging from policy to product design to improved recycling technologies.

Although recycling targets are an obvious, easily monitored and (potentially) enforceable legislative measure to promote a circular economy, there are many end of life options that preserve a much greater degree of product functionality. Waste avoidance is not enough, if it were then current trends towards biodegradable packaging, waste incineration and landfill reduction would be sufficient. Maintaining and extending the maximum value of limited resources is necessary for a sustainable society. In order to reuse, repair, remanufacture and refurbish products, manufacturers need to be involved in the value chain beyond production. This could be in formal partnerships with waste processing agencies or by implementing extended producer responsibility, pledging to return defunct products to use (4, 5). A change of emphasis towards valuing a product's service not its material worth prompts a

reduction in waste and better use of resources. For example, chemical leasing is a business model where payment for a service is based on productivity, not how much material changes hands (6). Under this business model it is possible to buy paint on the basis of what surface area is to be coated or industrial solvents according to how much apparatus needs to be cleaned or degreased. It is now important to the selling party to provide as little product as possible to maximise profit and in doing so minimise waste. Similar principles are being applied to consumer purchases of clothes and electronic devices on a leasing basis, rather than buying an article outright and eventually disposing of it (7).

Inevitably all products will become obsolete and the obvious way to extend the value provided by finite materials at this point is through recycling. Recycling processes for most types of material produce an inferior product that enters lower value applications, known as open-loop recycling or downcycling. Coupled with poor collection rates, this means 95% of the economic value of the plastic market is lost after a single use (8). Mechanical recycling is effective for PET, polyethylene (PE) and polypropylene (PP), whereby the waste is shredded, melted and remoulded (9). Recycling infrastructure for other polymers is more limited internationally and for composites and thermoset plastics the design and chemical composition of the material excludes conventional recycling completely as an end of life option (10). The presence of additives in many plastic products results in a recyclate with unknown impurities, some of which are toxic and they may be unnecessary or undesirable for the secondary uses of the material.

Closed-loop recycling, returning materials back to their original use, is prevented by product designs that irreversibly combine different types of materials, but also by waste collection and separation processes and the recycling processes themselves. These three aspects of waste management can be addressed by proactive product design, policy action regarding waste collection and recycling infrastructure and engineers and scientists motivated to create new recycling technologies.

Solvents can be used to selectively dissolve waste polymers at end of life for the separation of mixed wastes and composites. The advantage of this technology compared to mechanical recycling is that it is capable of returning a plastic with the same quality as virgin materials as judged by tensile strain and other properties. Recyclate specification sheets often include space for this technical information

alongside a description of its appearance (such as colour and particle size) (11). Quality control and the communication of recyclate properties is important to ensure the most value is obtained when deciding what materials are used to make products. Chemical recycling is another alternative recycling technique that takes material a step further back in the production chain by depolymerising it back to monomers (12). This is advantageous for polymers that degrade during use, including biodegradable polymers wrongly captured by recycling practices or that are unstable at the elevated temperatures used in recycling processes. Chemical recycling can potentially be solvent free but in many examples a solvent is required to homogenise the polymer with reactants and catalysts.

In this work, three important case studies will be discussed where a solvent-based process is used to recycle a polymer. The emphasis is on commercial applications, exploring their advantages and limitations. For a theoretical examination of polymer solubility and the related phenomena of gelation and swelling, other literature is available that provides the background knowledge for solvent-based recycling methods (13).

For completeness, it must be said there are less desirable end of life options for waste in a circular economy whereby the value of materials is significantly reduced or completely eliminated. This includes increasingly popular energy recovery (incineration), as well as biodegradation and landfill. Incineration offers some value and offsets energy demand that would otherwise likely be obtained from fossil fuels. Despite the additional use of waste material as a fuel, ultimately the material is lost. Carbon emissions and any other form of pollution represents a loss of resource and the material value it could have provided to society. Biodegradable products are designed to avoid litter. There are also some instances where it is impossible to collect a product for reuse or recycling. One example is lubricants. Forestry regulations require chainsaw and other 'total-loss' lubricants to be biodegradable (14). To prevent avoidable resource depletion and waste, the only articles suitable for incineration or biodegradation in a circular economy are bio-based products made only of sustainably sourced renewable materials (15).

Solvent-Based Polyethylene Terephthalate Recycling

One of the most ubiquitous forms of plastic waste is the plastic bottle. Typically made of PET, these

single use articles can be effectively recycled, although most often this is in an open recycling loop to make polyester fabrics. Despite this, the recycling rate of PET bottles in Europe is below capacity at only 57% (16), indicating flaws in collection and sorting. Product design also limits recycling. Once (recycled) PET is combined with other materials to make textile products, the inability of conventional recycling processes to separate the PET means there is no option to further recycle the material. For textiles consisting of a mix of cotton and PET, a solvent-based process can perform the separation and recovery of both components.

There are a large number of patented procedures for recycling textile waste containing mixed polyester and cotton items, typically clothes. A solvent can be applied to selectively dissolve either cellulose or PET. The remaining, undissolved polymer can also be recycled after filtration and drying or alternatively converted into a derivative compound. To selectively dissolve cellulose, the solvents used to make rayon fibres are applicable, such as *N*-methylmorpholine *N*-oxide (NMMO) which is used in the Lyocell process. The high flammability and oxidising potential of NMMO does not make it an ideal solvent from a safety point of view but it is typically recycled within processes with high efficiency. It has been reported that processes dissolving the PET component of composite textiles, for example in sulfolane (17), reduce the quality of the cellulose fibres (18). Nevertheless, the difficulty in dissolving cellulose has meant research efforts have focused on the solvent-based recovery of PET from textiles rather than the cotton.

Worn Again is a UK-based company that has developed technology for the closed-loop recycling of PET from textiles. A demonstrator pilot plant is due to be operational in 2021 (19). The principal technology describes a solvent added to blended polyester-cotton textiles at an elevated temperature (for example 100°C) (20). Suitable PET solvents include aromatic esters and aldehydes, as well as dipropylene glycol methyl ether acetate. Hot filtration removes undissolved cellulose from the solution of PET. The polyester is obtained with the use of isopropanol acting as an antisolvent. Characterisation of the separated polymers is not available, aside from a statement in the patent that the recovered PET has an identical infrared (IR) spectrum to the virgin material (20). Other works indicate that dissolution-precipitation cycles do not impact the polymer molecular weight, but the crystallinity of the recyclate is significantly lower than virgin PET (21). Here *N*-methyl-2-pyrrolidone (NMP) was used as the solvent and an alkane for the antisolvent. The use of reprotoxic NMP is not sustainable in the presence of tightening regulations (22) and the forced precipitation by antisolvents is probably responsible for the crystallinity of the isolated polymer. Greater attention is needed at the precipitation phase of the process to produce higher quality polymers.

Worn Again has also patented a procedure for recycling PET packaging, including drinks bottles (23). The key innovation that distinguishes this from mechanical recycling is the removal of dyes that otherwise dictate the quality of recyclate (**Figure 1**). Synthetic textiles are also appropriate feedstocks for this process. Coloured plastics and dyed textiles are far less valuable as a secondary feedstock for products compared to uncoloured transparent materials. The Worn Again technology is based on a solvent or temperature switch to firstly dissolve any dyes (but not PET) and then the polymer is dissolved at a higher temperature or in a different solvent. It is important that the first solvent swells but does not dissolve PET under the operating conditions. For instance, dyes are dissolved in ethyl benzoate at 120°C and liberated from the swollen plastic. After removing the dye solution, a second batch of ethyl benzoate is added at 180°C to dissolve the polymer. It is necessary to implement this second step to remove any insoluble impurities. For this to be economically viable the solvent will need to be recycled and in this regard the process is simplified by using the same solvent throughout. A PET recovery of 96% is satisfactory.

Solvents described as able to dissolve PET are provided in **Table I** (20, 23). Due to solvent residue potentially trapped in the recyclate, it is important to consider toxicity as part of solvent selection. The CHEM21 solvent selection guide categorises hazards into safety (S), health (H) and environmental (E) impact using a 1–10 scale where high scores reflect severe hazards (24). Benzyl acetate and ethyl benzoate are listed as having the best health and safety profile. High boiling solvents such as these are penalised in the environmental category because recovery by distillation is energy intensive. Depending on the proposed applications of the recycled PET, residual solvent limits for food contact applications or other regulations must also be considered.
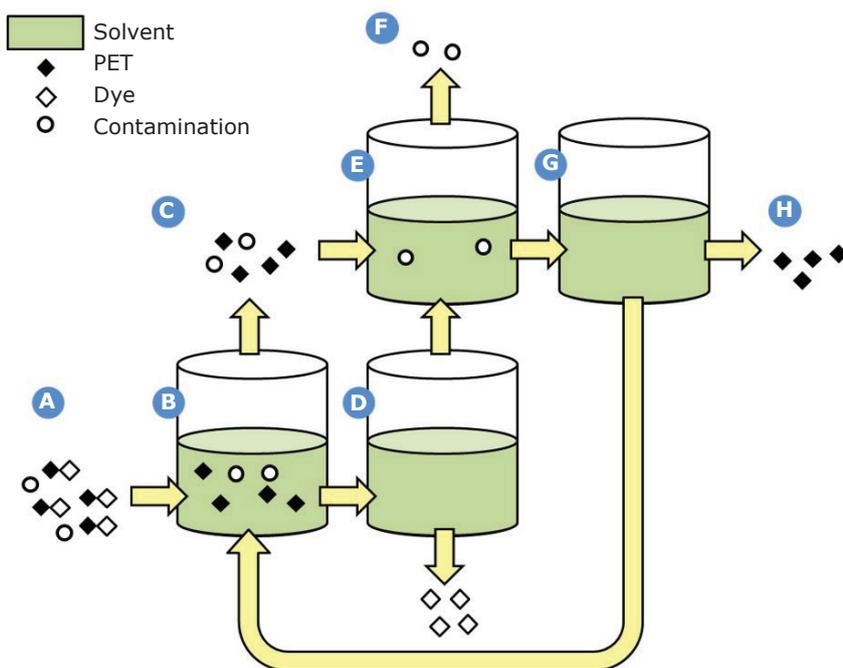
Fig. 1. Simplified schematic of the Worn Again PET recycling process; **A** contaminated and dyed PET collected; **B** dye dissolved (low temperature); **C** contaminated PET filtered; **D** dye solution removed and solvent reclaimed; **E** PET dissolved (high temperature); **F** contamination filtered; **G** PET solution cooled to precipitate PET and reclaim solvent; **H** recycled PET

Table I Patented Examples of PET Solvents, Listed Alongside Hazards (Data Compiled From REACH Registration Dossiers and Safety Datasheets)				
Solvent	**Hazards**	**S**	**H**	**E**
Benzaldehyde	Harmful if swallowed or in contact with skin. Causes skin irritation	2	2	5
Benzyl acetate	Harmful to aquatic life with long lasting effects	1	1	7
Butyl benzoate	Harmful if swallowed. Causes skin irritation. Causes serious eye irritation	1	2	7
DMEU	Harmful if swallowed. Causes serious eye damage. Suspected of damaging fertility or the unborn child. May cause damage to organs through prolonged or repeated exposure	1	6	7
Dipropylene glycol methyl ether acetate	Causes skin irritation. Causes serious eye irritation. May cause respiratory irritation	1	2	5
Ethyl benzoate	No reported hazards	1	1	7
Methyl benzoate	Harmful if swallowed	1	2	5
Cyclohexanone[a]	Flammable liquid and vapour and is harmful if inhaled	3	2	5
Ethyl acetate[a]	Highly flammable liquid and vapour, causes serious eye irritation and may cause drowsiness or dizziness	5	3	3

[a] Solvents for PET dyes only

Ultimately any disruptive PET recycling technology needs to provide significant advantages over efficient and widely practiced conventional mechanical recycling processes. The ability to separate combinations of materials is a crucial aspect of solvent-based recycling. With still much to be done to improve recovery rates of easier to recycle products, new technologies will only become commonplace if there is a political will to approach very high, near complete recycling rates, including composites.

Solvent-Based Polylactic Acid Recycling

Recycling techniques primarily aim to preserve the chemical structure of materials, but polyesters, with their susceptibility to hydrolysis and alcoholysis,

are also possible to depolymerise into monomers. Alcoholysis of PET is a favourable chemical recycling approach because its reaction with ethylene glycol produces *bis*(2-hydroxyethyl) terephthalate as an appropriate monomer to remake PET. Alternatively methanol will produce dimethyl terephthalate and ethylene glycol which can be combined as they are to produce virgin PET (25). Hydrolysis creates an aqueous solution of terephthalic acid and ethylene glycol which thermodynamically discourages esterification.

Chemical recycling is appropriate when other recycling methods produce a poor quality recyclate. This could be due to contamination that is possible to remove during chemical recycling or because the polymer is prone to decomposition. PLA is thought to be responsible for both these issues by the recycling industry. As a biodegradable polymer, mechanical recycling causes degradation into shorter fibres and as a polyester it is also likely to contaminate PET recyclate. However, PLA is suited to chemical recycling. Hydrolysis or alcoholysis produces a single monomer and it is more rapidly decomposed than PET. This means PET waste destined for recycling can be pretreated to remove any PLA by chemical recycling. It has also been shown that mixtures of PLA and PET can be sequentially chemically recycled into their respective monomers in a two-step process so that the polymers no longer contaminate one another (**Figure 2**) (26). This concept proves useful where conventional sorting techniques (such as near-IR) cannot distinguish

between polyesters (27), although new analytical systems are being developed to address this (28).

Although recycling PET mechanically without the need for depolymerisation or solvents is the prevailing technology, interest in alternatives is increasing, for example by Carbios, France and DEMETO, EU Framework Programme for Research and Innovation Horizon 2020. The understanding of PLA chemical recycling is arguably more advanced, but commercialisation is constrained by the small market share of PLA and the types of product it is used in. Many PLA containing products are designed for composting at end of life (for instance, plastic lined disposable coffee cups, transparent films for food packaging and other applications). As a bio-based polymer, PLA films are suitable for composting in a circular economy (if other end of life options that preserve more value are not accessible) as there is no net loss of material or emissions from a material perspective. Having said that, there are also many other components and articles made of PLA that will not biodegrade in the conditions provided by industrial composting units (PLA is not suitable for home composting). Thicker PLA materials, such as those that result from three-dimensional (3D) printing with PLA filaments are unlikely to be adequately decomposed by biodegradation on a viable timespan. The possibility that PLA is collected together with PET waste is increasing with the advent of reusable PLA drinks bottles, creating a reason to consider chemically recycling PLA.
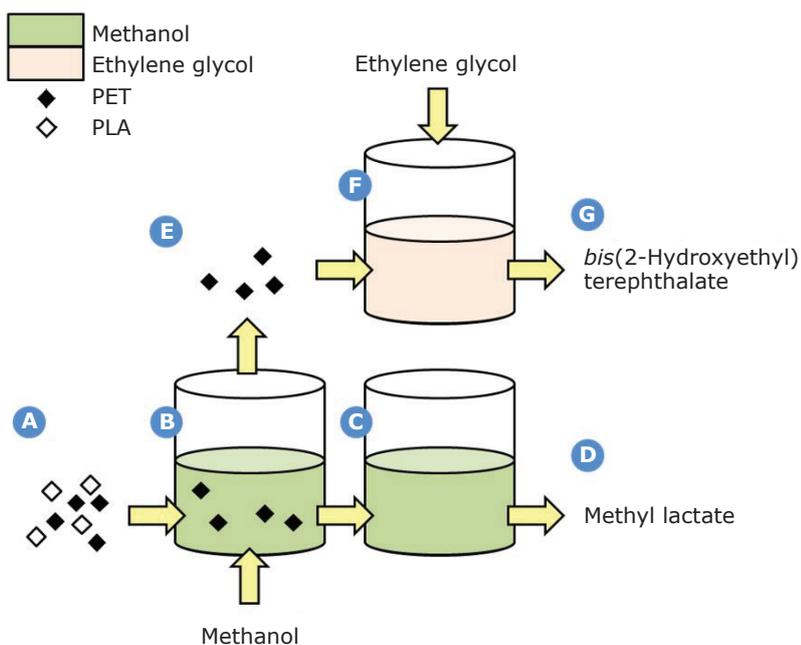

Fig. 2. Two step chemical recycling of PLA-PET mixed waste; **A** co-collected PET and PLA; **B** zinc acetate catalysed alcoholysis of PLA; **C** filtration of methyl lactate solution; **D** isolation of methyl lactate after evaporation of excess methanol; **E** PET recovered; **F** zinc acetate catalysed alcoholysis of PET; **G** isolation of *bis*(2-hydroxyethyl) terephthalate

Zeus Industrial Products, USA, has patented a process for depolymerising PLA using conditions where PET is unreactive (29). An inert solvent (chloroform) is added to the polymer, along with reactant (methanol) and catalyst (tin dioctanoate) to complete depolymerisation at 57°C. Disadvantages of this process include the use of chloroform, which is toxic if inhaled and suspected of causing cancer and reprotoxicity (24). Full depolymerisation also requires several hours (30).

The Futerro LOOPLA® process (a joint enterprise formed by Galactic, Belgium and Total, France) is another method for chemically recycling PLA. The company has expertise in PLA production as well as its hydrolysis and alcoholysis at end of life. Either chemical recycling method is potentially able to remake a feedstock suitable for PLA production (31). Hydrolysis can occur in a solution of PLA in ethyl lactate at 130–140°C (32). Ethyl lactate is a significantly less hazardous solvent than the chlorinated solvents that are often used to dissolve PLA and other polyhydroxyalkanoates (24, 33). Without the addition of a catalyst, 97% recovery of lactic acid (isolated by crystallisation) is achieved with minimal hydrolysis of the solvent. Potential contamination by PE, PP or PET is resolved because ethyl lactate does not dissolve these polymers, which can be used advantageously to separate PLA from other plastic wastes by hot filtration. If ethanol is added to the recycling process instead of water, alcoholysis occurs (34). The product is identical to the solvent, ethyl lactate, and so separation is simplified. Distillation removes excess ethanol and residues (such as pigments and contamination). An acid catalyst is required and triazabicyclodecene is preferred.

An issue with the described recycling procedures is the product (lactic acid or its esters) is subject to racemisation which produces inferior polymers with lower crystallinity (12). This must be controlled in order to perform closed-loop recycling. Furthermore, the electricity demand is too high for chemical recycling to compete with mechanical recycling (35, 36). While this is a valid concern for PET, mechanical recycling is not appropriate for PLA anyway due to its degradation (37). The first major barrier preventing chemical recycling of PLA being operated at any appreciable scale is the lack of feedstock and therefore an absence of designated PLA waste collection (38). However, the market growth of PLA products indicates future measures to capture PLA waste will need to be implemented.

Solvent-Based Polyvinylchloride Recycling

Many solvent-based recycling research projects and pilot trials have been successful, but few are viable commercial processes because of the competition from mechanical recycling and in the case of PLA the limited feedstock. The most prominent example of a successful recycling process conducted in a solvent was the VinyLoop® process, yet after 16 years of operation the plant was closed in 2018. It is important to understand the reasons why to ensure more recycling operations do not close and waste materials are not considered a burden and unnecessarily incinerated or landfilled when more value could be obtained from them.

The VinyLoop® process took PVC waste streams, often contaminated with textiles and other materials, and selectively dissolved the PVC in an organic solvent. The PVC was then precipitated by steam-driven evaporation of the solvent which itself was recycled. The PVC was said to be of the same quality as the original material. VinyLoop® was a Solvay, Belgium, technology commercialised as a joint venture in 2002 and ran until 2018 (39). The plant in Ferrara, Italy was established to recycle up to 10,000 tonnes of waste a year, primarily cable insulation (40). In 2008 the plant was updated to treat textile composites as well.

Methyl ethyl ketone (MEK) is a good PVC solvent and in the VinyLoop® process was used with the cosolvent *n*-hexane (**Figure 3**) (41). In a typical example of the process, 9.3 kg of 82% MEK, 5% water, 13% hexane was added for every kilogram of PVC. After mixing at 100°C (2.8 bar) for 10 min, a dispersant was added (0.2% relative to PVC of METHOCEL™ K100, a cellulose ether). The dispersion agent was needed to make fine particles of PVC. Then the temperature and pressure were reduced and steam injected (3.6 kg per kilogram of PVC). The addition of water allowed evaporation of a MEK-water azeotrope. Precipitation of PVC occurred at 64–65°C, below the boiling point of the azeotrope. Over 99% of the recovered PVC was able to pass through a 1 mm sieve. The water-MEK-*n*-hexane mixture was also collected. The presence of *n*-hexane improved the separation of the organic phase from water for reuse. An earlier patent describes the addition to salts to achieve the same effect (42).

The PVC waste being processed had been plasticised into flexible products. The VinyLoop® process maintained the additive composition of
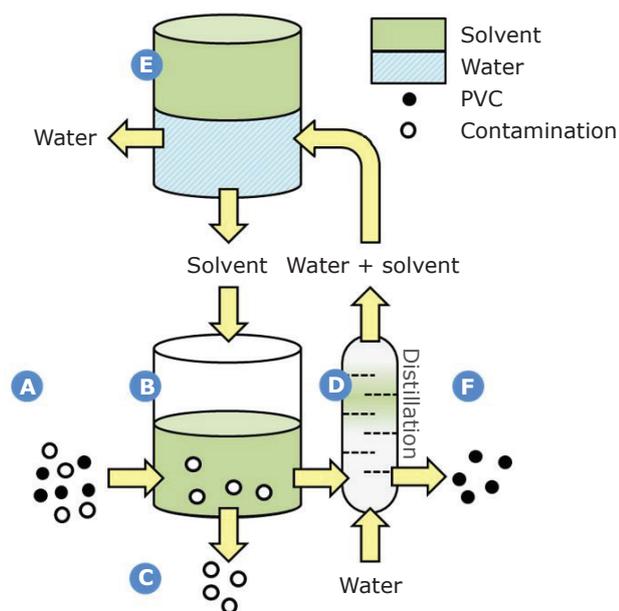
Fig. 3. A simplified schematic of the VinyLoop® process; **A** collection of PVC containing waste; **B** selective dissolution of PVC; **C** filtration of contamination; **D** steam distillation; **E** recovery of solvent; **F** recycled PVC

the PVC, which in theory may be advantageous for closed-loop recycling, but in practice the ability to introduce new additives to create new products for contemporary markets and meet changing regulatory requirements would have been preferable. It was the latter that caused the closure of the VinyLoop® plant. Phthalate esters are used extensively to plasticise PVC. The toxicity of phthalate esters has prompted action by the European Chemicals Agency (ECHA), resulting in a ban on many phthalates, including *bis*(2-ethylhexyl) phthalate, since 2015 (43). The European Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH) regulation that dictates the nature of bans or restrictions on chemical use requires any company producing, importing, using or isolating *bis*(2-ethylhexyl) phthalate (present above 0.1%) to have obtained authorisation to do so (44). Recycling of materials containing substances subject to authorisation is also within scope of the REACH regulation. How much this is appreciated, adhered to and policed in Europe is a subject of interesting debate with significant consequences. The operators of VinyLoop® did have authorisation (45), but these permits are time limited, in this case less than three years. As the expiry date of the authorisation drew near, the recycling plant was closed. The expectation is that most companies will stop handling the banned substances and find alternatives where possible because authorisation is very expensive to obtain. For a recycler, they are subject to the nature of waste produced by others, including legacy materials and plastics produced by manufacturers with authorisation to include otherwise banned plasticisers. A further complication is that medical products are exempt from the plasticiser ban and so there is the possibility of materials containing *bis*(2-ethylhexyl) phthalate still entering recycling streams.

This case study raises some important questions. How much PVC currently in use contains banned plasticisers? Many articles such as the cable insulation that was recycled by VinyLoop® has a long lifetime and was made before the EU phthalate bans were implemented. Can solvents remove additives in a compliant way? This question can be addressed by studying the solubility of PVC and phthalate esters. *bis*(2-Ethylhexyl) phthalate is a liquid expected to be miscible with a large number of organic solvents. Techniques for phthalate determination use solvent extraction methods, albeit on a small analytical scale. Therefore it is logical to add a pre-step to future recycling methods where phthalates (if present) are extracted by swelling but not dissolving the PVC or by dissolving both polymer and additives but later selectively precipitating the PVC. Distillation, as practiced by VinyLoop®, leaves non-volatile components unseparated (i.e. PVC and *bis*(2-ethylhexyl) phthalate). A condition of handling substances subject to authorisation in Europe is not to isolate or store refined batches of the chemical(s) in question without a permit. For recycling this is an issue as what can be considered an impurity cannot be removed without destroying it *in situ*. Incineration or chemical transformation may be suitable and legal approaches.

If a process were to be developed that could remove additives in a compliant way, the cosolvents MEK and *n*-hexane may no longer be the ideal combination for PVC recycling. This creates scope to reduce the hazards posed by *n*-hexane in particular. In solvent selection it is important to know what solvents are restricted or subject to authorisation by REACH of course. Recently some ether and chlorinated solvents have been subjected to authorisation and a large number of restrictions on how many others can be used are also in place (22, 33).

Solvent-Based Polyethylene and Polypropylene Recycling

The polyolefins PE (high and low density grades) and PP are produced in greater quantities than any other synthetic plastics. As for PET, mechanical recycling is viable because of the availability of the waste and the quality of the recyclate is appropriate for large markets. However, the high calorific content of these hydrocarbons means they are favoured as a feedstock for energy recovery plants (46). Plastic pyrolysis to make oils suitable for refining into fuels and base chemicals is being investigated as a more flexible alternative to incineration (47). The technology is proven on a multi-tonne scale (48, 49). BASF has now used pyrolysis oils made from waste plastic to feed the steam cracker at its primary chemical production plant (50). This indicates there is tangible interest in diversifying the uses of waste polyolefins.

It is also feasible to recover polyolefins from solution. Pappa *et al*. found xylene at 85°C dissolves PE but not PP (51). The undissolved PP could be removed by filtration and then the PE precipitated with an antisolvent (propanol). Recovery on a 3 kg scale was greater than 99% (**Figure 4**). The authors report no loss in performance attributes of the recovered polymers and actually an increase in crystallinity. This is unusual compared to the previous case studies (12, 21). Other research also reports that the elastic modulus of PE and PP increases while other properties are the same or slightly improved after solvent-based recycling (52). One explanation is that while recovery is high, the small losses probably represent the more soluble lower molecular weight polymers with less desirable properties.

Solvent-based recycling can offer a major advantage when it is used for separation of wastes. Extraction of polymers from mixed waste streams with selective solubility has been known for decades (53), but it is not cost competitive with flotation and near-IR sorting. However, multilayer materials cannot be separated effectively with current technology. This must be considered as a design flaw in a circular economy, which if impossible to resolve by product designers must be addressed by recyclers. Multilayer packaging typically contains a film of aluminium and a number of plastic layers, including PE sealing layers. The use of switchable-polarity solvents can delaminate these materials by dissolving the PE (54, 55). The principle of a switchable-polarity solvent is based on a hydrophobic amine that is converted into an
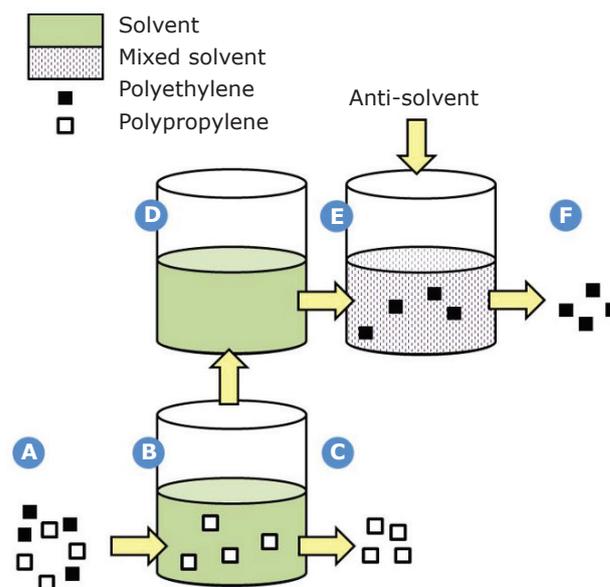

Fig. 4. Separation of polyolefins; **A** mixed PE and PP feedstock; **B** selective dissolution of PE; **C** PP recovered; **D** filtration to give a PE solution; **E** addition of anti-solvent; **F** isolation of PE by filtration

ammonium bicarbonate solution with the addition of water and carbon dioxide (**Figure 5**) (56). The resultant hydrophilic antisolvent precipitates the PE. Releasing the carbon dioxide pressure then reforms the original amine ready for reuse.

Conclusion

Current policies and investment for waste collection, separation and recycling limit the circularity of materials. Product design, consumer choices and conventional business models also share the blame. Despite academic interest in novel polymers designed to self-heal, rapidly biodegrade or depolymerise on command, they are met with resistance by established petrochemical plastic markets. The major reason is that new, synthetically complex products will be more expensive. The introduction of new plastic materials also increases the complexity of the plastic waste market and that is generally unhelpful for recycling practices. Recycling rejection rates are overall already increasing in the UK, now standing at over 4% of post-consumer material collected from households (57). At end of life, small volume plastics are contamination in PET, PE and PP recycling streams, which increases the likelihood that waste is not returned to use because of the low quality of the recyclate. We see this in the
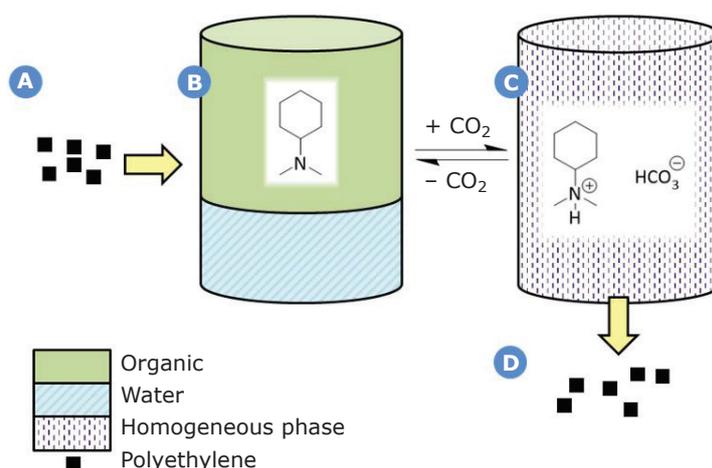
Fig. 5. A schematic of a switchable-polarity solvent being used to process PE; **A** PE is collected; **B** hydrophobic amine solvent dissolves PE (water may or may not be present at this stage); **C** addition of carbon dioxide (and water) forms a hydrophilic solution; **D** precipitation of PE

recycling of PET, where the presence of PVC at 100 ppm can cause discolouration and degradation of the recyclate (58). Solvent extraction makes it possible to remove PVC from PET (59), in the same way that it might become necessary to remove PLA from PET waste in the future (27).

The potential of polystyrene recycling is also high (60), but recycling rates of consumer waste are low due to the very few districts willing to collect it. Significant barriers to polystyrene recycling include its smaller market size compared to the other major plastics and its low density. Expanded polystyrene is uneconomical to collect, transport and sort for this reason. A number of solvent-based approaches have been proposed to dissolve and densify polystyrene, which in turn could make recycling more economical. Limonene is an effective solvent (61, 62) and Ran *et al*. recently reported the use of binary solvent systems to dissolve polystyrene (63). The use of switchable-polarity solvents is also known for this purpose (64), but no commercial plants are operational at this time.

The potential for solvent-based recycling to make a significant contribution to a circular economy depends on willingness to invest in end of life processes that recycle difficult waste streams. Start-up and maintenance costs are certainly higher than a conventional recycling plant. There is a social benefit to recycling composites and layered materials that relates to the avoidance of litter, including topical concerns about ocean pollution and microplastics. Waste management of electrical and electronic equipment is infamous for exports to Africa exploiting vulnerable people and exposing them to toxic substances (65). The Basel Convention now makes this practice illegal. With responsibility now placed on treating this waste domestically, research has shown solvents assist the separation and recovery of the complex and valuable components found in these articles (66–69). Removing or at the very least monitoring additives will become hugely important to the recycling industry. Addressing brominated flame retardants is a key step in the reprocessing of electrical and electronic equipment (70, 71). Solvent-based recycling processes have been shown to successfully remove brominated flame retardants from plastics by firstly dissolving the waste and then adding a second solvent to selectively precipitate the polymers (72, 73). Ultimately the possibility of future feedstock shortages and subsequent price increases, coupled with countries' refusal to accept foreign waste (74), will demand a change to recycling practices beyond simply increasing the capacity of conventional processes. Whether this will occur in the short term or many decades from now depends on the prioritisation of a circular economy in the ambitions of world leaders.

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