

# Current approaches to waste polymer utilization and minimization: a review

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## Abstract

The mass production of polymer products, in particular plastics, and their widespread use depending on the inherent advantages they have, make these materials ironically a threat to life on Earth. Polymer recycling is being considered as one of the most widely accepted remedies to the threat of growing amounts of plastic waste by both the public and scientists. In practice, recycling is associated with many difficulties, such as problems related to separation, sorting and cleaning operations, lack of fiscal subsidies, instability of selective garbage separation programs, high transport and electricity costs, etc. Still, a large section of society and the authorities agree on the necessity and importance of recycling to protect the environment, and natural habitats and resources for future generations in a balanced manner to conserve raw materials, and to reduce energy consumption, municipal solid waste production and greenhouse gas emission. The recycling effort is almost endless in itself and includes a variety of approaches such as refurbishing, mechanically reshaping, chemically treating, thermally utilizing, etc. Some novel approaches such as application in carbon capture or synthesis of carbon nanostructures from the plastic waste are among the new process technologies of recycling. From traditional and promising polymer waste utilization approaches, this review will highlight sustainable methods to reduce impacts of plastic waste on the environment.

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**Keywords:** environmental remediation; process technology; recycling; sustainable processing; waste treatment; waste minimisation

## INTRODUCTION

Polymers and plastics are seen in many areas of everyday life from household items to industrial products. A polymer is a large molecule, or a macromolecule, built up from many repeated subunits known as monomers. The term 'plastic' is originated from the Greek word 'plastikos', meaning 'moldable'. Plastics are classified as thermoplastics and thermosets. Thermoplastics can be heated and shaped several times with deterioration in the physical and mechanical properties while thermosets are produced via a setting reaction which gives them their permanent shape. The key difference between plastics and polymers is that all plastics are polymers but not all polymers are plastics. A polymer can either be biological or synthetic, while plastics are made from petroleum and its derivatives.

Both synthetic, e.g. poly(ethylene terephthalate) (PET), polyethylene (PE), polypropylene (PP), and natural, e.g. cellulose, starch, chitin, polymers play essential and ubiquitous roles in everyday life. The backbone chains of addition polymers, e.g. polyolefins such as PE, PP, polystyrene (PS) and poly(vinyl chloride) (PVC), are solely built of carbon atoms and quite resistant to degradation or hydrolytic cleavage. Consequently, these polymers are considered non-biodegradable.<sup>1</sup> The condensation polymers, on the other hand, contain heteroatoms such as O and N on their backbones. Polyamides (or nylons), polyesters (i.e. PET and polycarbonate (PC)) and polyurethanes are some examples of this class. These polymers are potentially susceptible to degradation by the hydrolytic cleavage due to hydrophilic amide or ester linkage between monomers. However, when they are disposed to landfills, condensation polymers as well will persist for at least decades, and probably for centuries.<sup>2</sup> This is mainly because commercial products resist oxidation and biodegradation due to the additives they

have, such as anti-oxidants and stabilizers. Moreover, their molecular weights are usually too high. It is reported that biodegradation begins only for molecular weight values of a few tens of thousands, which can be considered very low for many commercial polymers.<sup>3,4</sup> Crystallinity is another parameter that adversely affects biodegradation.<sup>5</sup> For the polymers listed in the first group, i.e. those synthesized by addition polymerization, degradation by natural means is even more difficult because of their chemically inert character.

Commercial plastics are made up of polymers and additives such as plasticizers, stabilizers, antioxidants, impact modifiers, reinforcing agents or fillers, pigments, compatibilizers, lubricants, flame retardants, and so on. These chemicals are added to give the polymer enhanced functionality; without the presence of additives, many polymeric materials would be of limited use. Unfortunately, concerns also exist over the additives; e.g. phthalates or bisphenol A. These chemicals are reported to be harmful to human health through some biological mechanisms.<sup>6</sup> The polymer content in a plastic can vary widely from less than 20% to

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nearly 100%.<sup>7</sup> The diversity in plastic formulations complicates recycling activities. In order to recover most of the inherent value of the pristine plastic and to produce a new product with properties mimicking those of the virgin material, the waste plastic to be recycled must have a uniform and well-defined composition. This makes separation, sorting and cleaning (for instance, separation of PET bottles from other plastic bottles, sorting them according to colors, and removal of non-PET components such as label, label adhesives, and cap) the central parts of a plastic recycling process.

The global production of plastics, excluding fibers of PET, PP, and polyamide, was approximately 322 million tons in 2015 and 335 million tons in 2016.<sup>8</sup> Around 50% of plastics are made for single-use applications, such as disposable consumer items and packaging; the rest is used for long-term infrastructures such as pipes and structural materials and for durable consumer applications with intermediate lifespan such as in furniture and vehicles in almost equal amounts.<sup>9</sup> It is estimated that the amount of plastic waste entering the ocean from land was between 4.8 and 12.7 million tons globally in 2010.<sup>10</sup> Plastic wastes, therefore, form a major component of marine litter, affecting the aquatic and the mainland life negatively.

A recently published report by the Danish Environmental Protection Agency concluded that plastic grocery bags made of low-density PE (LDPE) are more environmentally friendly than paper, bio-plastic and cotton alternatives.<sup>11</sup> The study shows that a paper bag would need to be used 43 times to achieve the same per-use resource expenditure as an LDPE-bag. Greater energy requirement for its production means more carbon emissions. In order to assure the environmental impact is similar to the LDPE-bag, a bio-material based plastic needs to be recycled 42 times, while a cotton-bag has to be reused up to 7100 times (and around 20 000 times, if the cotton is organic). In 2015, England stopped handing out plastic bags for free, and instead charged five pence per LDPE-bag.<sup>12</sup> This resulted in almost a six billion reduction in the number of plastic bags handed out in 6 months.<sup>13</sup> In 2016, France banned plastic plates, cups, and utensils with a law that will go into effect in 2020, becoming the first country in the world to do so. Exceptions will be allowed for items made of compostable, bio-sourced materials.<sup>14</sup> In 2018, Seattle became the first major US city to fully ban single-use utensils and straws in food service.<sup>15,16</sup> There will be a net benefit of discarding less single-use plastic materials, however, there are serious concerns about their replacements. Recycling plastic materials seems eco-friendlier than replacement of plastic with paper, bio-plastic or, in particular, cotton.

In many previous publications, conventional recycling technologies such as pyrolysis and depolymerization, and recycling paths of various polymers, e.g. PET and PE, or polymer classes, e.g. polyurethanes, polyamides and bioplastics, have been reviewed in detail.<sup>17–24</sup> In this review, we have dealt with traditional and promising recycling technologies in general, without specifically focusing on a certain polymer or method. We have emphasized the importance and necessity of the development of new process technologies for polymer waste utilization in order to make a positive impact on the world we live in. This review also presents some applications and, finally, suggestions on measures that can be taken on a wide scale, from end-users to manufacturers, to make recycling/utilization as widely applicable as possible.

## PLASTIC WASTE MANAGEMENT: CURRENT STATE AND CONVENTIONAL METHODS OF RECYCLING

Recycling is the process of using recovered material to manufacture a new product or to recover energy once material enters the waste stream. Energy recovery, where the high-yielding calorific value of polymer is utilized by controlled combustion as a fuel, presents lower environmental performance than material recovery as it does not reduce the demand for new material and may cause emissions and fly and bottom ash containing toxic residues such as lead and cadmium.<sup>25</sup> Incineration has been widely assessed as ecologically unacceptable in the last decade as many environmental regulations have been implemented for a more sustainable recycling-oriented society.<sup>26</sup> In the order of decreasing environmental desirability, the main strategies for management of plastic waste can be specified as reduce, reuse, recycle (materials) and recover (energy). Landfilling is considered the least desirable management strategy and should be limited to the necessary minimum.<sup>9</sup> Yet, landfilling has also been regulated, e.g. by the European Commission with the Council Directive 99/31/EC to reduce the negative effects of landfilling on water, air and soil sources. Beyond the concerns related to collection, transportation and long-term risks of soil and groundwater contamination by toxic additives, a major drawback of landfilling arises from a sustainability aspect; that is none of the materials used to produce the plastic is recovered, the material flow is linear rather than cyclic.<sup>9</sup>

Among countries in the European Union, waste management strategies vary significantly as can be seen in Fig. 1.<sup>27</sup> Germany, the Netherlands, Belgium, and Austria recycle more than half of their waste. Almost all of the remaining waste is combusted to recover energy in these countries. In Luxemburg and France too, incineration ranks second in waste management schemes after material recycling. In Sweden and Denmark landfilling rates are less than 5%. However, for the rest of the European Union countries the least desirable management option, landfilling, is the most applied method. Especially in Croatia, Malta, Bulgaria, and Romania, more than 90% of waste ends up in landfills.

The terminology used for plastic recycling is actually complex and sometimes confusing. According to American Society for Testing and Materials (ASTM) D5033 definitions recycling includes four categories: primary (mechanical reprocessing of scrap materials with controlled history into products with equivalent properties), secondary (mechanical reprocessing of used materials into products requiring lower properties), tertiary (recovery of valuable chemical constituents such as monomers or additives) and quaternary (recovery of energy) as presented in Scheme 1.<sup>9,28,29</sup> The 'transformation' of waste to more useful products or materials is called valorization. The 'transformation' may include any process by which the waste can be transformed into valuable products via reutilization or recycling. The term 'valorizing' means producing an energy source. Therefore, the tertiary and quaternary categories fall into valorizing, while primary recycling covers reuse of an object.

### Primary recycling

Primary recycling refers to recovery and reuse of materials without being changed and usually for the very same purpose.<sup>30</sup> It ensures simplicity and low cost, however, deals only with the recycling of clean, uncontaminated and single-type waste. Only certain plastics are recycled in this manner due to purity requirements, and there is also an obvious limit on the number of cycles for each material.<sup>31</sup>

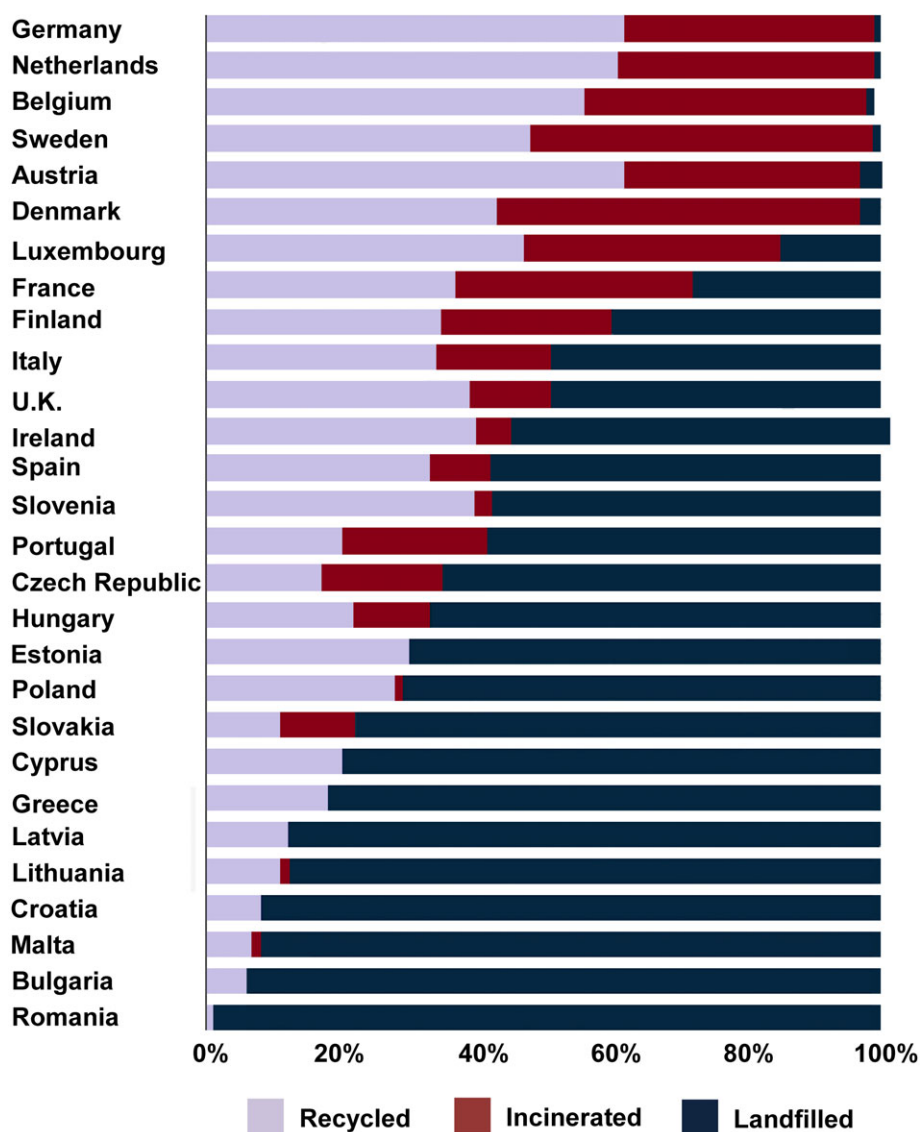


Figure 1. Recycling rates in Europe in 2011.<sup>27</sup>

Producing a new bottle from a scrap bottle is an example of this recycling category.

### Secondary recycling (mechanical recycling)

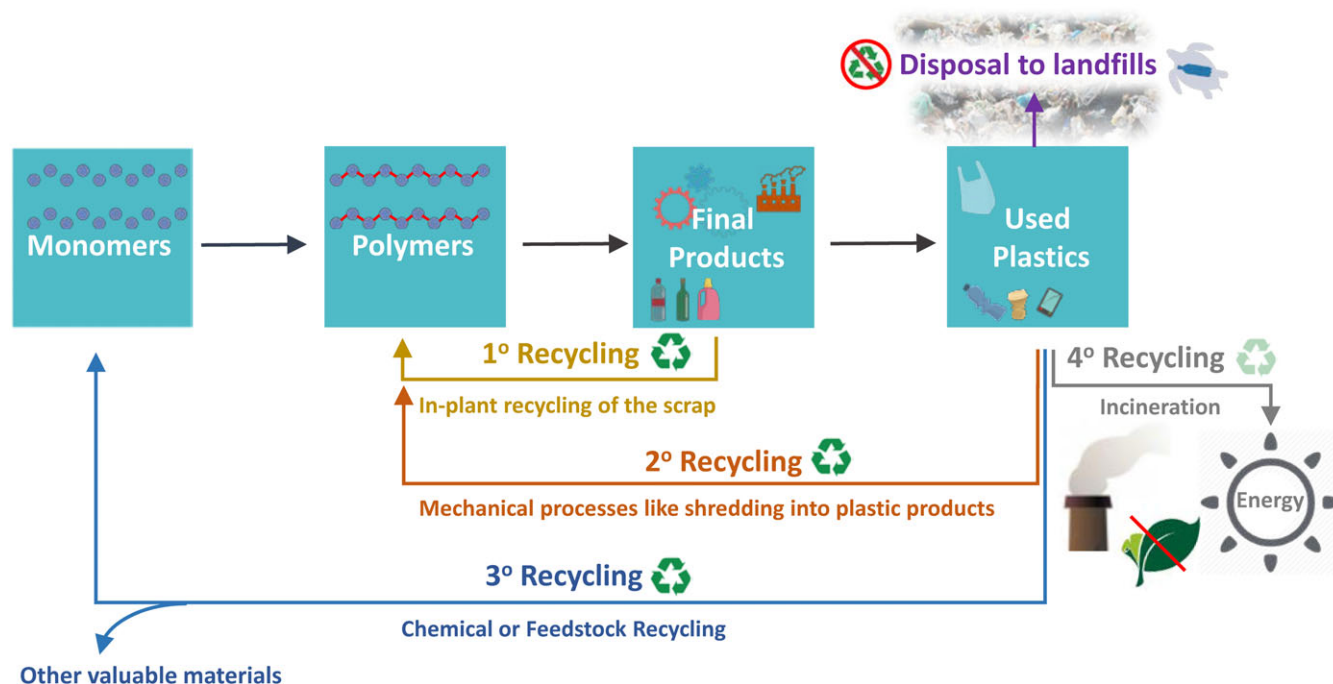
Mechanical or secondary recycling refers to reprocessing of waste plastics by physical means. The polymeric waste is generally reprocessed into granules by conventional extrusion after being separated from its associated contaminants. Collection/segregation, cleaning, and drying, chipping/sizing, coloring/agglomeration, pelletization/extrusion are the steps involved in this approach before the manufacturing of the end product. The main drawback of this method is deterioration of the properties of the product, mainly attributed to decrease in molecular weight after each cycle as a result of chain-scissions.<sup>32</sup> There are other limitations to the mechanical recycling process; temperature-sensitive plastics, composites, polymeric blends and plastics that do not flow at elevated temperatures such as thermosets and polymers with high melt viscosities, e.g. fluoropolymers, cannot be processed mechanically. Practically, only poly(ethylene terephthalate) (PET) and polyethylenes (HDPE, LDPE, LLDPE) are recycled by a

mechanical process. All other plastic wastes are either not recovered or their recovery rate is less than 1% with regard to their total production.<sup>33</sup>

### Tertiary recycling (chemical or feedstock recycling)

Chemical or feedstock recycling (tertiary 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. The monomers obtained can subsequently be re-polymerized to synthesize the original polymer.<sup>34</sup> Partial de-polymerization to oligomers or mixtures of other hydrocarbon compounds are also valuable; these materials may be used as a feedstock or as an input for the production of new plastics and petrochemicals by means of heat or chemical agents.<sup>32</sup> Tertiary recycling is attracting much attention because it is very profitable and beneficial from the sustainability point of view: it reduces the demand for energy and feedstock and the material flow is cyclic.

Pyrolysis or thermal cracking is considered as one of the most promising processes of tertiary recycling, conducted at high temperature and in the absence of oxygen (usually in a nitrogen



**Scheme 1.** Types of plastic waste recycling.

atmosphere).<sup>35</sup> During the thermal decomposition of polymers, depending on polymer type, chain scissions occur either randomly, yielding low molecular weight fractions, or from the chain-ends, yielding monomers. The decomposition products in the form of liquid oil, char or gases are all valuable as fuel or as feedstock for the production of the same or different plastic materials. If the tertiary recycling process yields energetic products consisting of high contents of carbon and hydrogen atoms with minor or no heteroatom contribution, they become valuable especially for the production of fuels such as automotive gasoline, jet fuel, and diesel products, provided that the products meet a number of strict standards.<sup>36–41</sup> Pyrolysis is conducted with or without a catalyst. Lower temperatures applied during the catalytic process yield faster degradation and a narrower fractional composition of the products. However, the main issue is economic efficiency; commercial catalysts are generally expensive and typically cannot be regenerated.<sup>42–44</sup>

There are various methods for chemical recycling. For polymers synthesized via an *addition polymerization* process, such as polyolefins (PE, PP, PS, PVC, polybutadiene, polyisoprene, etc.), acrylics (polymethacrylates, polyacrylamide polyacrylonitrile, etc.) and other vinyl types (poly(vinyl acetate), poly(vinyl acetals), poly(vinyl ethers), etc.) gasification, pyrolysis, liquid–gas hydrogenation and steam or catalytic cracking are the main subcategories of chemical recycling. The products yielded in chemical recycling of polyolefins contains numerous energetic components, hence they are especially valuable as potential fuels.

For *condensation polymers* such as polyesters (PET, poly(butylene terephthalate), polycarbonates, etc.), polyamides (aliphatic and aromatic nylons, polyimides), formaldehyde resins (phenol–formaldehyde resins, urea–formaldehyde resins, etc.), polyurethanes (polyurethane rubbers and foams, spandex fibers, etc.) and ether polymers (epoxy resins, etc.), the main subcategories of chemical recycling are hydrolysis (acidic, alkaline, and neutral), glycolysis, methanolysis, aminolysis, ammonolysis, and

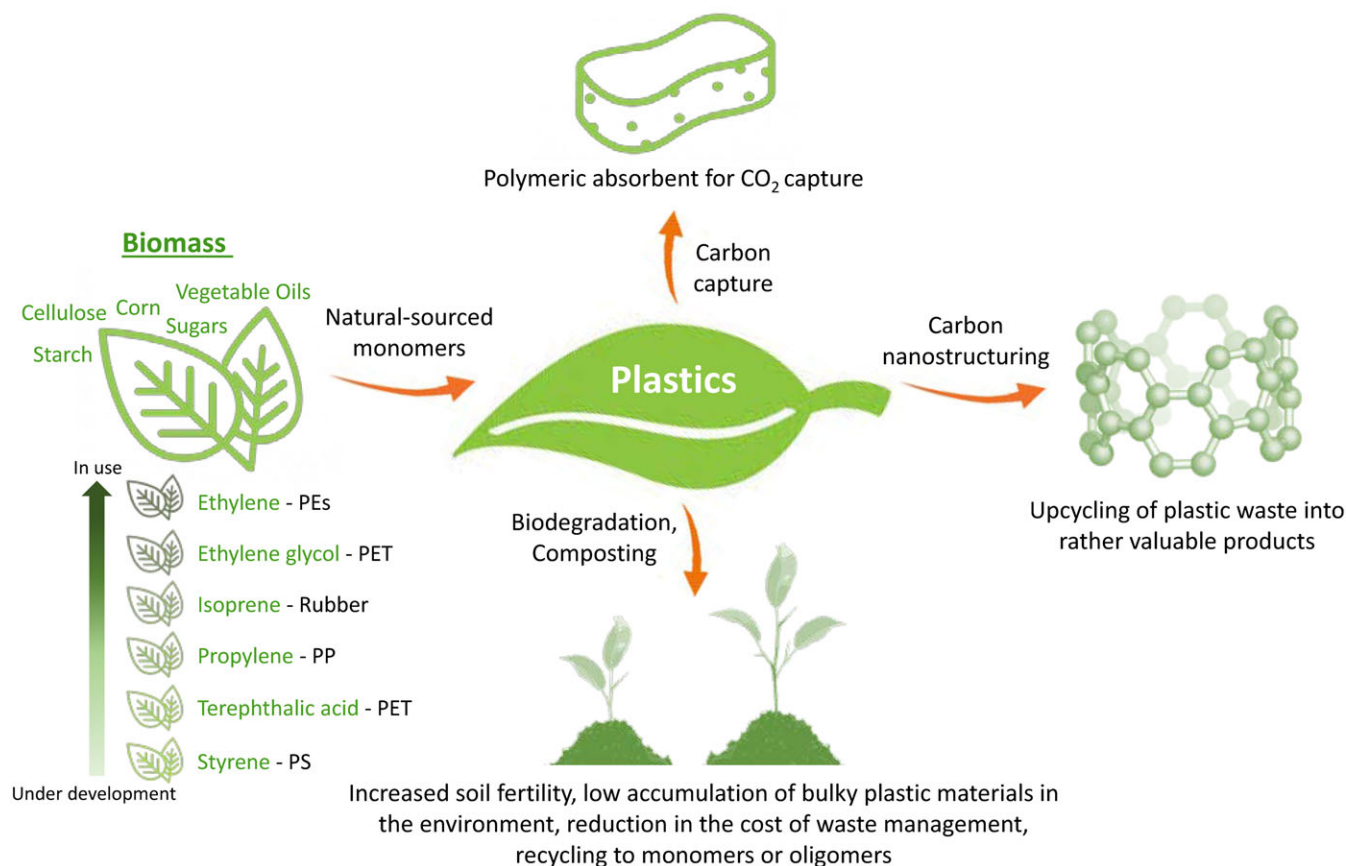
so on. Recycling of condensation polymers such as PET and Nylon by these methods yields monomer units (de-polymerization or monomer recycling) in a relatively short time.<sup>32</sup>

Other chemical recycling methods involves photodegradation,<sup>45–47</sup> ultrasound degradation,<sup>48–53</sup> degradation in microwave reactor,<sup>54–59</sup> etc. Despite all its advantages, unfortunately, chemical recycling is not a widespread method, mainly because of energy costs.<sup>33</sup> Numerous methods of chemical recycling to obtain valuable chemicals such as monomers, petrochemicals or additives from the used polymeric materials are under investigation presently, and sustainable conditions for these processes are under extensive research.

#### Quaternary recycling (energy recovery)

Recycling of plastic wastes via primary or secondary methods leads to deterioration of the properties. Therefore, continuous recycling could yield substandard and low-quality products. Chemical recycling, on the other hand, is not applied to each polymer because it is not efficient enough or economically sustainable. The end of life of plastics must be analyzed carefully because it eventually becomes economically unprofitable to recycle them further. As it is not reasonable to landfill polymeric wastes both from the environmental and economical points of view, it is beneficial to use the excellent physical properties of plastics in various applications. For instance, the mechanical properties of plastics are utilized in road,<sup>60</sup> pavement<sup>61</sup> or asphalt<sup>62</sup> constructions as modifier or reinforcing filler. Another physical property of plastics, their high calorific value, is quite valuable too; energy recovered from plastic waste can make a major contribution to energy supply. Quaternary recycling refers to the recovery of energy content of plastics by incineration. This is currently the most effective way to reduce the volume of organic materials. However, this method yields considerable toxic substances both in smoke and in ashes and is regarded as ecologically unacceptable.<sup>63</sup>





**Scheme 2.** Promising and eco-friendly plastic management approaches.

### Other valorizing options

The recycling process is divided into four parts, where primary recycling is the use of pre-consumer industrial scrap and salvage, secondary recycling is physical reprocessing, tertiary recycling is chemical processing and quaternary recycling refers to the recovery of energy.<sup>64,65</sup> In this section, a number of new approaches for the utilization of waste plastics were summarized.

Ravichandran *et al.* synthesized and characterized new polyphenolic flame-retardant material from a biodegradable and renewable starting material, cardanol, which is a by-product of cashew nut processing. Polymerization of cardanol took place in aqueous media with different types of oxidants. Promising results were obtained from preliminary studies, indicating that new types of flame retardant materials from bio-based phenols is possible.<sup>66</sup> Hollow spherical sludge carbons were prepared by Wu *et al.* from polystyrene foam sphere and sludge pyrolysis, which was then employed as an adsorbent to treat wastewater.<sup>67</sup> Conversion of waste expanded polystyrene into polymeric azo dyes with sulphonamide group was accomplished by Liu *et al.* through chlorosulfonation, amidation, hydrolysis, diazotization and coupling reactions.<sup>68</sup> Liao *et al.* reported the preparation of doped carbon catalyst for the reduction of oxygen employing precursors; polystyrene foam, melamine and iron chloride.<sup>69</sup> Using a simple technique, Fonseca *et al.* transformed waste polystyrene cups into negative electrode materials to be used in sodium ion batteries.<sup>70</sup>

There exist various materials used to reinforce asphalt. Lately, protecting the environment has become a crucial issue all around the world. Accordingly, carbon fibers and recycled waste packaging polymers seem to be good modifier candidates. For

modification of ordinary paving asphalt, Zhang *et al.* preferred waste packaging polyethylene and polyacrylonitrile-based carbon fibers over ordinary polymer modifiers.<sup>71</sup> Because of increasing environmental concerns over the last couple of decades, recycling and benefiting from non-biodegradable elastomeric wastes have become major issues for researchers. In recent years, continuous utilization of waste tires has been well studied.<sup>72–74</sup> Jeong *et al.* reported the preparation of a composite with supercritical carbon dioxide by mixing low-density polyethylene foaming and waste rubber (ethylene-propylene-diene monomer). For optimization of the process, temperature and pressure variations were studied on the foamed microcell formation. The composite prepared was found to be promising for its further use as a foaming mat for artificial turf.<sup>75</sup>

The environmentally continuous disposal and recycling of an increasing amount of electronic wastes have become a waste management matter globally. The disposal of electronic wastes is one of the major environmental challenges. The central components of many electronic devices and printed circuit boards (PCB) may contain various compositions. Serious amounts of toxic components are involved in these PCBs from TVs, laptops, mobile phones etc. together with polymers, ceramics and metals.<sup>76–78</sup> A novel study on the simultaneous use of automotive and electronic rubber waste as fillers in PP composites was reported by Kumar *et al.* This new environmentally friendly approach may enable benefiting from remarkable amounts of polymeric automotive and electronic wastes to fabricate high-value polymer compositions.<sup>76</sup>

Polymeric adsorbents can be employed for heavy metal adsorption since they can be regenerated easily and they have good

mechanical properties compared to other adsorbents like silica gel, cellulose and activated carbon.<sup>79</sup> Alsewilem and Aljlil studied cost-efficient adsorbents to remove lead from wastewater using discarded post-consumer polystyrene which was obtained cost-free. Results obtained from adsorption isotherms showed that recycled polymer compositions were a rather powerful tool compared with virgin polymer composites.<sup>80</sup>

## PROMISING APPROACHES

Current plastic waste utilization techniques are not applied at large scale. Mechanical or secondary recycling is the only widely applied approach, and unfortunately it is not being used for the treatment of thermoplastics beyond polyethylenes and PET, which represent 37% and 9% of the annual plastic produced, respectively. There is a need now more than ever to develop new recycling technologies which reduce energy consumption, increase the amount or value of products and reduce or completely eliminate toxic waste in a sustainable, economically feasible and socially responsible manner. Some eco-friendly options, summarized in Scheme 2, look promising to help the management of polymers in a sustainable way. These options are reviewed in detail below.

### Carbon capture using waste plastic sources

Carbon dioxide (CO<sub>2</sub>) emitted as a result of combustion of fossil fuels is among the main causes of global warming and climate change.<sup>81</sup> The capture and storage of CO<sub>2</sub> generated during combustion at coal- or gas-burning power plants is attractive for reducing CO<sub>2</sub> emissions into the atmosphere.<sup>82</sup> There exist economic and technical procedures that need to be followed for solid adsorbents employed in pre- and post-combustion CO<sub>2</sub> capture. The adsorbent should engage in a gas stream and leave the water–gas shift reactor at a temperature of approximately 40°C and a total pressure of around 30–45 bar. The major component here is the gas-shifter fuel, that is H<sub>2</sub>, for which the amount should be greater than 60 mol%. The temperature and pressure of the gas stream cause pressure-swing cycles to be principally attractive.<sup>83</sup> A preferred adsorbent should have good selectivity towards CO<sub>2</sub> to be able to specifically adsorb it among other components in the gas stream, have large sorption capacity in order to be considered as an economically appropriate process and have good physicochemical stability which includes steadiness against moisture, heat and acidic gases like hydrogen sulphide. The candidate materials for CO<sub>2</sub> capture and storage should also be cost-effective and their synthesis should be scalable.<sup>84</sup> Combination of high porosity and good chemical stability can be found in microporous organic polymers.<sup>85</sup> The scale-up costs are usually the prime concern in the production of porous polymer adsorbents synthesized using precious metals and catalysts, which is the case in conjugated microporous polymers.<sup>86</sup> Sneddon *et al.* reported the synthesis of a high-surface-area composite material using amine-functionalized waste PVC as the source material for the first time for CO<sub>2</sub> adsorption (Fig. 2). The synthesis scheme was based on a simple one-step reaction, which allowed it to be scalable for the further development of industrial CO<sub>2</sub> capture and storage units. Moreover, compared with un-functionalized mesoporous substrates, aminated PVC adsorbent was found to have exceptional hydrophobic characteristics which enable it to be used in elevated moisture content flue gases.<sup>87</sup>

In Europe, recovery of 69.2% of post-consumer plastics was accomplished via energy recovery processes and recycling,

whereas the disposal of the rest, 30.8%, took place together with the municipal solid wastes in landfills which in return reduced the landfill capacity as a result of low solid waste density, in 2014.<sup>88</sup> Fu *et al.* reported a technique for the synthesis of hyper crosslinked polymers using Friedel-Crafts reaction employing waste-expanded polystyrene foam as raw material and 1,2-dichloroethane as both solvent and crosslinker. With this approach, it was made possible to synthesize cost-effective hyper crosslinked polymers in a fast and facile manner. Additionally, it was found that BET surface area of the polymers could be controlled by tuning the reaction time and the weight ratio between polystyrene foam and catalyst.<sup>89</sup> The hyper crosslinked polymers synthesized showed better adsorption capacities compared to other CO<sub>2</sub> capturing materials and were found to be cost-efficient for production at industrial scales. A schematic representation of the synthesis process can be seen in Fig. 2.

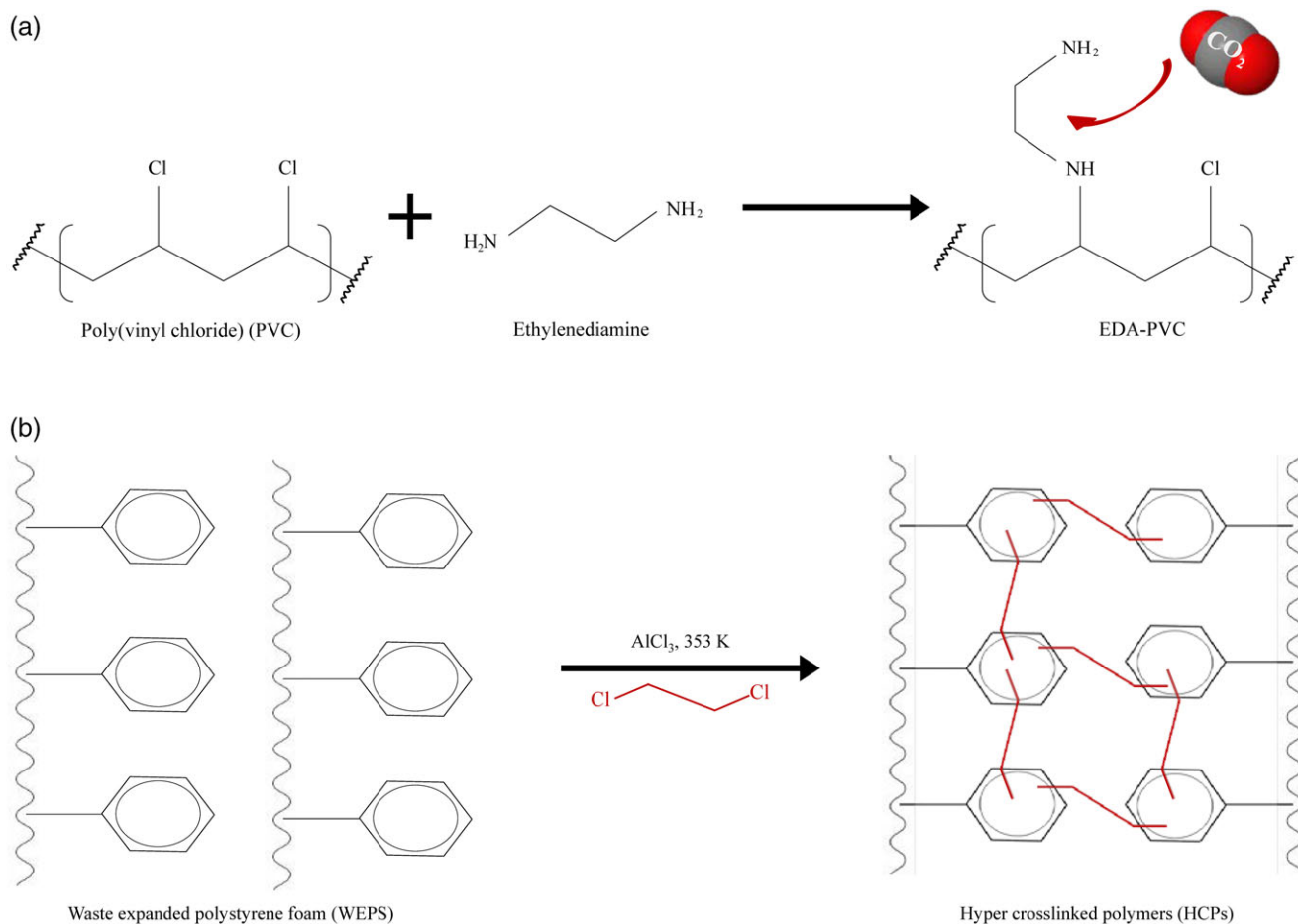
Because amine functionality boosts the CO<sub>2</sub> adsorption capacity, cost-efficient supports were advanced on these terms in order to cope with the current amine scrubbing system limitations. Most of the effort has been put into exploring an appropriate candidate with effective features to meet the energy criteria, however, their economical perspective restrain their feasible application.<sup>90,91</sup>

### Synthesis of carbon microspheres and nanotubes from plastic wastes

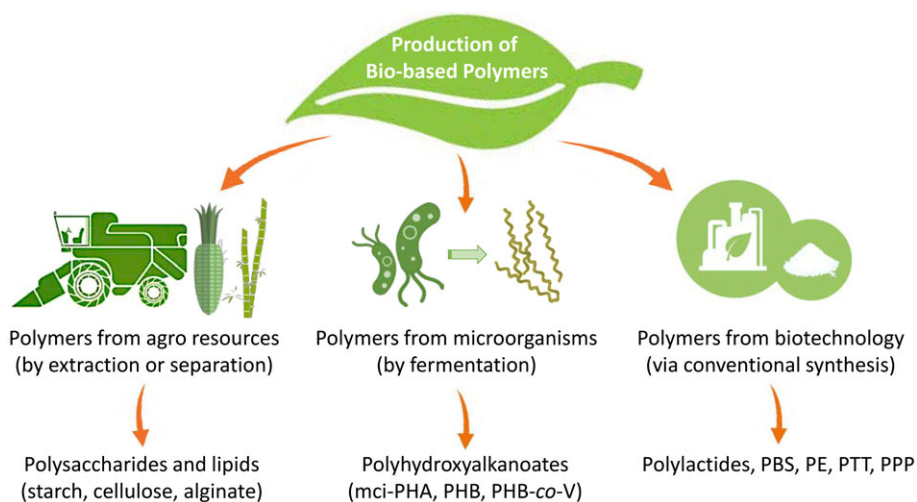
Upcycling refers to the conversion of a waste material into a valuable product. It is a sustainable concept and prevents waste being directly thrown into landfill or an incinerator. It also is quite useful since it makes use of inexpensive materials. It is commonly true that the more innovative the transformation of an upcycle entity, the more vendible it is. Therefore, upcycling is a constructive method due to cost benefits and waste reduction.

Pol claimed that waste plastics can be completely converted into high-purity carbon microspheres that would be crucial industrially.<sup>92</sup> Carbon microspheres are promising tools for various applications such as in printers, batteries, toners, paints and tires. Yang *et al.* reported the synthesis of amorphous carbon nanospheres with sizes ranging between 140 and 200 nm through treatment of poly(tetrafluoroethylene) (PTFE) in supercritical water at 550°C with Ca(OH)<sub>2</sub> as de-fluorination agent.<sup>93</sup> Zou *et al.* successfully degraded PTFE with a mild and cost-efficient method at low temperature.<sup>94</sup>

Carbon nanotubes (CNTs) can be classified depending on their graphene sheets as single-walled CNTs (SWCNTs) and multi-walled CNTs (MWCNTs) which are continuous cylinders of a graphene sheet with hexagonal units consisting of sp<sup>2</sup>-bonded carbon atoms and multiple rolled graphene sheets, respectively. Waste plastics are among the carbon sources through which CNTs can be produced. This way, both the waste problem can be solved and resourceful product can be produced. Plastics pyrolysis is another technique to produce high-value CNTs with bizarre physical and chemical properties. This technique has been employed for some time and follows mixing plastics with catalyst in a single reactor or passing the pyrolysis products of plastic through a second stage catalyst reactor.<sup>95–98</sup> Wu *et al.* reported the production of hydrogen and high-value CNTs from waste plastics, employing a pyrolysis-reforming method consisting of a two-stage reaction system in the presence of steam and Ni-Mn-Al catalyst. The waste plastics comprised commercial waste high-density polyethylene (HDPE), re-granulated HDPE containing PVC and plastics from motor oil containers. The results showed that a



**Figure 2.** Schematic representation of chemical processes developed by (a) Sneddon *et al.*<sup>87</sup> and (b) Fu *et al.*<sup>89</sup>



**Scheme 3.** Bio-based polymer manufacturing route.

pyrolysis-reforming process produces hydrogen and in the meantime, CNTs were formed on the catalyst.<sup>99</sup>

Other than waste plastics and hydrocarbon gases, pyrolysis oils can also be employed to produce CNTs. In a study by Quan *et al.*, high phenol-containing pyrolysis oil was obtained, from a printed circuit board. Then, hollow-centered and straight CNTs were produced with the pyrolysis oil.<sup>100</sup> It should be noted that during

waste plastic CNT transformation, along with CNT, producing hydrogen is also quite beneficial. For instance, Nahil *et al.* studied a number of Ni-based catalysts for the co-production of CNTs and hydrogen through pyrolysis of waste plastics.<sup>101</sup> Then, in-line catalytic reforming of pyrolysis vapours was performed employing a two-stage reaction system in which the first-stage pyrolysis was performed at 500°C and second-stage catalytic reforming was

**Table 1.** Natural-sourced monomers: transformation into commodity plastics

Source	Natural-based monomers	Modification	Application	Reference
Bagasse, corn cobs, oat hulls	Furfural	Conversion to adipic acid and hexamethylene diamine, reaction with phenol	Production of Nylon, 66 and thermosetting phenolics	107,108,111
Rosin	Terpene, levopimaric acid, etc.	Chemical conversion and polymerization	Thermoplastic resins, high temperature resistant resins	107,108,111,118,119
Vegetable oils, e.g. soybean oil, castor oil, rape seed oil, etc.	Fatty acids such as oleic acid, linoleic acid, erucic acid, etc.	Extraction, epoxidation, catalytic reactions, etc.	A large range of polymers from soft rubbers to ductile or rigid plastics, e.g. polyurethanes, polyesters, polyethers, etc.	120–126
Sugar waste	Glucose	To Sorbitol	As Polyol	107,108,111
Molasses	Sucrose	To hydroxymethyl furfural and sucrose methacrylate	Variety of chemicals and polymers	
Carbohydrates	Lactic acid and levulinic acid	Extraction, catalytic reactions, etc. and polymerization	Poly lactide copolymers	107,108,111
<i>Rhus fernisifera</i>	Urushiol	Oxidative polymerization	Specialty coatings	107,108,111
<i>Anacardium occidentale</i>	Cardanol	Chemical modification and polymerization	High performance polymers	107,108,111
Crude cells containing PHA	Microbial synthesized monomers	Pretreatment, extraction, and purification	Poly(hydroxy alkanooates) (PHAs)	127–129
Suberin	Suberin monomers	Crosslinking	Suberin polymers	107,108,111
Sugar	Ethylene	Dehydration of ethanol obtained from sugar into ethylene	Polyethylene	130
Sugar	Propylene	Catalytic conversion of ethanol into propylene	Polypropylene	130
Isopentenyl diphosphate	Isoprene	Biosynthesis using bacterial sources	Polyisoprene	130
Sugars and cellulosic biomass	Ethylene glycol	Hydrolysis, hydrogenolysis, catalytic conversion	Synthesis of PET	131–133
Sugars and cellulosic biomass	Terephthalic acid	Catalytic aerobic oxidation	Synthesis of PET	131
Glucose	Styrene	Microbial biosynthesis	Polystyrene and styrene-based copolymers	134
Biomass, e.g. glucose sources	Succinic acid	Extraction, fermentation	Polyamides, polyesters, poly(ester amide)s	135–137
Polysaccharides such as starch, cellulose	Isosorbide, isomannide and isoidide	Depolymerization, hydrogenation, dehydration	Polyesters, polycarbonates, polyethers and polyurethanes	138–140
Lignin	Vanillin, vanillic acid, terephthalic acid	Extraction, derivations	Polyurethanes, PET, various aromatic polymers	141–145

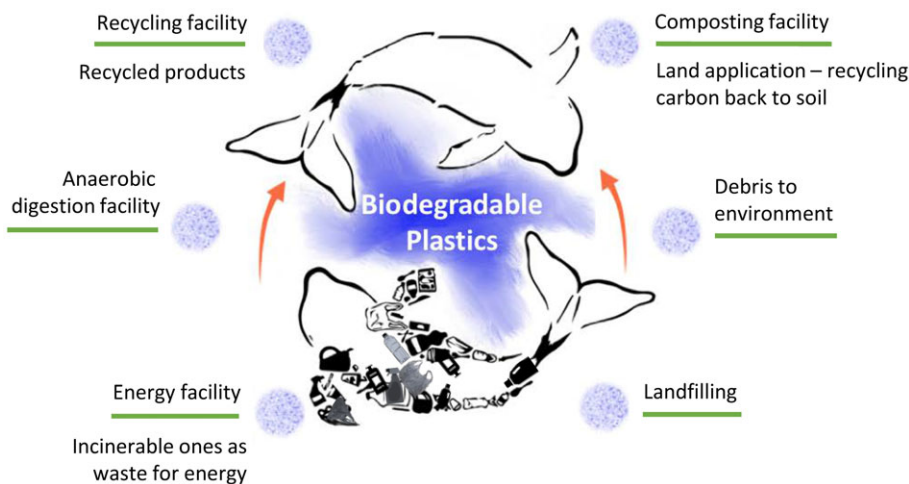
carried out at 800°C. They also revealed that addition of Mn into Ni-Al catalyst dramatically increased the CNT production and the hydrogen yield was also found to be high. The suggestion was that the weak interaction between metal particles and catalyst support had a significant role in CNT growth. A comparable process was employed in the conversion of real waste plastics into CNTs and hydrogen.<sup>99</sup> Results obtained by Borsodi *et al.* showed that the presence of Cl (0.3 wt% PVC in waste HDPE) had an adverse effect on CNT formation, whereas the presence of S had less influence with respect to CNT morphologies.<sup>102</sup> El Essawy *et al.* reported a cost-efficient solvent-free procedure to convert PET bottles into carbon nanomaterials through thermal dissociation inside a closed system under autogenic pressure in the presence of additives and/or catalyst, which acted as cluster nuclei for carbon nanomaterials like CNTs and fullerenes. This study was successful in the production and control of the different forms of

micro-structured carbon nanomaterials from PET waste through optimization of parameters with regard to amount of catalyst, additives and time.<sup>103</sup> It is clear that the developed processes are limited to laboratory studies and more effort should be made to scale them up. Main challenges encountered by researchers are reactor design and conversion and high operating temperatures, in spite of the sustainable course of waste plastic management. For a sustainable scale-up procedure these challenges and related parameters should be adjusted.<sup>104</sup>

#### Natural-based monomers for commodity plastics

In order to decrease the negative effect of plastic waste on the environment, one approach would be to decrease the production and use of petroleum derived polymers. Synthesizing polymeric materials from bio-based feedstocks is of interest being more eco-friendly compared with what are currently being used,





**Scheme 4.** Biodegradable plastic integration.

namely fossil-based feedstocks. Accordingly, this concept was also included into our review. Several natural-polymer based industries were able to compete against the strict competition of synthetics and this success is mainly due to developments in the chemical modification of natural polymers. Among the most important ones are the generation of methods to tune structure, shape and size of the polymers to combine a set of functions and properties for certain applications. There have been trials to apply this knowledge to natural polymers and monomers in order to accomplish required properties, leading to new opinions on possible alternative source to produce polymers.<sup>105–107</sup> The dramatic decrease of fossil resources along with the increase in oil prices induced the search for alternatives, focusing on renewable resources for polymer production.<sup>108,109</sup> Except polymers existing naturally, there exist various monomers that occur naturally, either in free or combined form and can be obtained by cleavage, extraction of de-polymerization from biomass.<sup>110</sup> Agricultural and forest products are widely available and vary structurally, which creates the requirement to reappraise their applications. Huge amounts of effort have been put into the synthesis and characterization of macromolecular materials, synthesized employing renewable resources. Therefore, monomers like furans and terpenes, oligomers such as tannins and rosin and polymers including proteins, cellulose, macromolecules synthesized via microbes have been proved to have features to create various materials through intelligent utilization.<sup>111</sup>

To obtain high-value polymers, studies on applying molecular and process design principles to polymers and monomers obtained naturally or from renewable resources are increasing. Various novel materials are being developed to take the place of petrochemical derivatives.<sup>111–117</sup> Table 1 shows a few of the modifications to some natural-based monomers and their polymers.

Researchers found various applications with polysaccharides and proteins in biodegradable products. There is a lack of natural enzymes in nature for degrading natural polymers. The formation and degradation rate of the final metabolites and the structural complexity of the materials as well as environmental conditions chosen for the degradation are interdependent.<sup>111,146–150</sup> There are enormous opportunities and challenges for the next decades regarding the utilization of non-renewable chemical sources for the synthesis and further construction of polymers. Renewable material sources can be counted as phenolic compounds

occurring naturally such as wood, fiber, shell,<sup>151</sup> seed,<sup>152,153</sup> bagasse,<sup>154</sup> empty fruit bunches,<sup>155</sup> acids like lactic acid and alcohols like glycerol and soybean oil.<sup>156–158</sup> There has always been and still is a requirement for the development of new techniques to benefit from renewable resources and synthesize new monomers from to help enable a greener future.

#### Biodegradation and composting opportunities

Latest trends in the field of biodegradable polymers led to important developments with regard to innovative design strategies and engineering to procure high-performance advanced polymers. Many definitions of biodegradation are based on the application field of the polymers, such as natural environment or biomedical areas.<sup>159</sup> The definition of biodegradation made by Albertsson and Karlsson was if it was an event taking place through the action of enzymes and/or chemical decomposition regarding living organisms and their secretion products.<sup>160</sup> Generally, synthetic polymers are non-biodegradable. Although, polymers with hydrolysable backbones (poly(ester amide)s, poly(amide enamine)s, poly(ortho ester)s, polyurethanes, polycaprolactone, polyamides, polyesters, vinyl polymers) are sensitive to enzymatic biodegradation and hydrolysis.<sup>159</sup> In the simplest definition, biopolymers are developed in nature and are polymers resulting from growth cycles of organisms. Therefore, they are also termed as natural polymers. Synthesis of biopolymers usually involves enzyme catalyzed chain growth polymerization reactions with activated monomers that generally occur within cells through complex metabolic processes. Scheme 3 shows the bio-based polymers' manufacturing route created by Shiam.<sup>161</sup>

It is of no biodegradable value if the product does not culminate in a waste management system using the biodegradability properties.<sup>162,163</sup> Scheme 4, adapted from Song *et al.*<sup>163</sup> shows biodegradable plastic integration with disposal substructures which employs the biodegradable function of the plastic product.

Biodegradable polymers can be classified as bio-based or petrochemical-based depending upon their origins. The former version is based on being produced from natural origins and biodegradable by nature. These natural origins like microorganisms, animals, and plants can be proteins like wheat gluten, casein, gelatin, lipids like animal fats, plant oils and polysaccharides like cellulose, chitin, and starch. An example of this



**Scheme 5.** Suggested measures for the management of plastics.

category can be natural rubber and some polyesters produced from microorganisms or plants like poly(3-hydroxybutyrate) and polyhydroxyalkanoates or from bio-derived monomers like polylactic acid. Petrochemical-based biodegradable polymers like aliphatic polyesters such as polycaprolactone, polyglycolic acid or polybutylene succinate and aromatic copolyesters such as poly(vinyl alcohol) and polybutylene succinate terephthalate are synthesized via petrochemical refining derived monomers.<sup>163,164</sup> Polylactic acid (PLA) is currently being employed in the packaging of short-shelf life bottles, films and thermoformed containers.<sup>165</sup> A thermoplastic biodegradable polyester poly( $\epsilon$ -caprolactone) (PCL) can be synthesized through chemical conversion of crude oil followed by ring-opening polymerization.<sup>165</sup> PCL has good resistance to chlorine, solvent, oil, and water, has low viscosity and melting point and can be thermally processed easily.

Thermosets are rather tricky in recycling terms since they cannot be remolded after they are once cured and cannot be decomposed thermally with high-temperature heating. Therefore, to recycle and redeploy thermosets, one that can be naturally de-polymerized would be quite useful to lead the way. In a study by Garcia *et al.* recyclable thermosets and organogels were successfully synthesized through one-pot, low-temperature polycondensation between 4,4'-oxydianiline and paraformaldehyde, forming hemiaminal dynamic covalent networks that can cyclize at high temperatures further to produce poly(hexahydrotriazine)s.<sup>166</sup> This study has potential in terms of meeting the requirements for thermoset polymers.

Biodegradable polymers can be used in various fields. Eco-friendly polymers are required in many sectors such as packaging, medicine, controlled drug release and automotive. Bioplastic polymers are promising to serve in the reduction of land-fill, material recovery and use of renewable resources. To be able to benefit fully from these materials, effective substructure for strict control of composting, separation, collection and certification as well as widespread public awareness are of significance.

## CONCLUSIONS

Plastic products need to be responsibly managed throughout their life cycle so that they are prevented from escaping into the environment by proper waste management, mindful product

design and responsible behavior. A number of measures outlined in Scheme 5 can be implemented on a wide scale, from end-user to manufacturers, to manage the design, use and recycling of plastic materials.

Reduce, reuse, recycle (materials) and recover (energy) are the main plastic management strategies in the order of decreasing environmental desirability. The most preferred strategy, reduce, includes source reduction activities, such as reducing the quantity of materials used in production of plastic goods, e.g. using concentrated detergents, extension of the life of products to postpone disposal, avoiding unnecessary use where possible, and replacing single-use products in the market with eco-friendly ones, e.g. using natural-based superabsorbent polymers<sup>167</sup> in disposable diapers instead of petroleum-based ones. Along with these measures, reducing the number of components of plastics, e.g. using a single polymer as much as possible instead of blending multiple polymers in plastic formulations, and reducing the amount of additives used or standardizing them to achieve easy removal from plastics via well-defined procedures may significantly increase the recycling rates.

Some simple measures such as stamping the information written on the labels instead of using labels and label adhesives may facilitate separation, sorting and cleaning operations which are the central and the toughest parts of a plastic recycling process. Government actions are also needed to achieve a broader engagement affecting both the consumers and producers, e.g. plastic products may contain a certain amount of recycled material depending on the field of use.

These measures may not always be economically feasible; however, research should continue to develop proper methods to replace the current ones. There are pros and cons for all plastic management approaches. Ideally, complementary practices of all actions and conservation would be the most ecologically friendly and economically feasible in the long run. We suggest that research should continue to develop sustainable methods of recycling of polymers along with appropriate sorting and identification methods. For instance, new catalysts must be developed to increase the chemical recycling rates of plastics in an energy-saving manner. Most plastics are immiscible with each other; even small amounts of contamination with a

different plastic may potentially hinder use of the recycled material. Designing new compatibilizers enhancing the miscibility and physical properties of polymer mixtures is, therefore, important. Almost all current recycling technologies focus on certain thermoplastics, e.g. PET. The rest, in particular cross-linked thermosets, are ignored in recycling operations. Sustained research efforts are important to expand recycling to a diverse array of polymer materials. It seems not to be greener to replace plastics with frequently-mentioned alternatives, such as paper, bio-plastic or, in particular, cotton.<sup>11</sup> Therefore, plastics are indispensable in our lives and will continue to be so; we have to find out better ways to responsibly and sustainably design, recycle and recover them.

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