



## Introduction to “Make polymers sustainable, why and how?”

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It is estimated that more than 300 million tons of synthetic polymeric materials are being produced every year and most are made from petroleum-based feedstocks. As the global consumption of polymers increases each year, this puts an unsustainable demand on our finite and non-renewable fossil fuel resources. In addition, the ever-growing quantity of polymers becoming waste at the end of their life presents serious environmental problems due to their persistence and potential ecotoxicity.

This joint themed collection on “Make polymers sustainable, why and how?” brings together a selection of outstanding contributions, spanning reviews, perspectives, papers and communications, which collectively provide a panoramic view of the field’s current vitality. These works range from recycling of polymer wastes, substitution of polymer carbon with oxygen (or sulfur), utilization of biomass to replace petroleum-based polluting polymers,

copolymerization of CO<sub>2</sub> with other monomers, and converting polymers (either natural or synthetic ones) into platform chemicals. This editorial introduces selected works that have contributed to tackling these scientific issues. Readers will observe a variety of approaches taken by the works featured in this themed collection, ranging from developing technologies to high-efficiency synthetic process, to utilization of biomass and CO<sub>2</sub> as novel carbon banks, and finally to recycling of consumed polymers. In a word, these contributions demonstrate the central role that chemical principles play in endowing polymers sustainability. As the introduction to the themed collection, here we would like to highlight some typical works to offer readers a promising perspective in sustainable polymers.

### Utilization of biomass

To tackle challenges posed by petroleum-based general polymers, biomass constitutes another significant carbon bank, showing great potential to provide versatile monomers of many polymers.<sup>1</sup> For example, a perspective on low-carbon polymers (<https://doi.org/10.1039/D5GC02706C>) shows natural cellulose can be used as a reference (O/C molar ratio of 5/6 and weight percentage of 49.4%) for low-carbon polymers that integrate non-edible biomass-based feedstocks, CO<sub>2</sub>, and industrial carbon/sulfur waste (*i.e.*, low-carbon monomers). This study also discussed the

unique characteristics of these low-carbon polymers in comparison to conventional “carbon-rich” polymers, highlighting the representative examples of PLA, PHA and PPC that are on the way to commercialization. Fiandra *et al.* (<https://doi.org/10.1039/D5GC04056F>) used diglyoxylic acid xylose to partially replace terephthalic acid in soil-release polymers, forming a polyester copolymer. The resulting copolymers enabled enhanced cleaning of polyester and poly-spandex fabrics, presenting high-performance detergent additives with an improved sustainability profile. In another minireview, lignin and glucose (<https://doi.org/10.1039/D5PY00792E>) are confirmed to be efficient alternatives to phenol and formaldehyde in generating sustainable phenolic formaldehyde resins. In order to preserve fossil fuels and meet the demand of chemical industry, some natural polymers can be used to replace petroleum-based precursors. Wang *et al.* (<https://doi.org/10.1039/D4GC05368K>) summarized advances in converting chitin into nitrogen-containing compounds, emphasizing progress in synthesizing benzenoid aromatics and highlighting future prospects.

### Reducing carbon contents in backbones

Developing polymers with heteroatoms in the main chain is another strategy to make polymers low-carbon. Liu *et al.* (<https://doi.org/10.1039/D5GC02535D>) reported that integrating labile Si–O–C and Si–O–C(=O)

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linkages into polymer backbones enables producing silicon-based polymers, such as poly(silyl ether)s and poly(silyl ester)s. These polymers possess controlled degradation characteristics without compromising performance. By incorporating sulfur at different positions of thiolactone monomers, sulfur-containing polymers (<https://doi.org/10.1039/D4PY01442A>) are also developed with chemical recyclability as next-generation high-performance sustainable polymeric materials. Their excellent thermal stability and the mechanical properties of the resulting poly(thioether-thioester) products are superior to those of commercial low-density polyethylene. Furthermore, the introduction of sulfur brings benefits for chemical recycling of these polymers to their monomers, demonstrating good sustainability. In other research (<https://doi.org/10.1039/D5GC02799C>), the integration of 6 mol% oxygen into blends of poly(lactic acid) (PLA) and LDPE demonstrates an improvement in the interfacial adhesion and mechanical properties.

## Recycling of post-consumer polymer waste

With the increase in production, a significant portion of plastic waste accumulates in landfills, oceans, and incinerators, leading to severe environmental and health risks. As a result, much attention has been paid to recycling of polymer wastes, broadly including mechanical recycling and chemical recycling.<sup>2,3</sup> In this collection, the recycling of polyethylene terephthalate, *i.e.*, PET or plastic #1, has been studied. A tutorial review (<https://doi.org/10.1039/D5GC03083H>) summarized recent advancements in the catalytic reductive conversion of post-consumer PET into fuels, value-added chemicals, and degradable polymers, with a particular focus on heterogeneous catalysis. Furthermore, synthetic textiles (<https://doi.org/10.1039/D4GC04768K>) are also capable of being depolymerized into added-value reusable monomers using chemical catalysts. In terms of method-

ology, several innovative techniques are reported, including mechanochemical ball milling (<https://doi.org/10.1039/D5GC03507D>), photocatalytic depolymerization (<https://doi.org/10.1039/D5GC01218J>), and so on. Catalysts play a significant role in depolymerization of polymer waste. In this collection, many excellent catalysis systems are reported for chemical recycling of spent polymers, such as CeCl<sub>3</sub> (<https://doi.org/10.1039/D5GC01218J>) and carbon-supported single site molybdenum dioxide (<https://doi.org/10.1039/D4GC05916F>) for depolymerization of PET.

## Innovative preparation processes

In order to reduce energy consumption and pollutant emissions, some contributions tackle the front line of highly efficient synthesis processes. Kurita *et al.* (<https://doi.org/10.1039/D4PY01169D>) developed a copper-catalyzed azide-alkyne cycloaddition (CuAAC) process to generate cyclic peptide-based [2]rotaxanes from cyclo (PG)<sub>4</sub> and monocationic ammonium threads. The pursuit of novel sustainable materials is driving advancements in polymer science. Using a similar principle, aliphatic polyester block copolymer blends of poly(L-lactide)-*block*-poly(γ-methyl-ε-caprolactone)-*block*-poly(L-lactide) (LML) and poly(L-lactide)-*block*-poly(γ-methyl-ε-caprolactone) (ML) have also been synthesized (<https://doi.org/10.1039/D5PY00506J>). The tackified pressure-sensitive adhesive (PSA) demonstrates competitive adhesion properties when compared to established styrenic PSAs. Grilo *et al.* (<https://doi.org/10.1039/D5PY00485C>) developed a melt polycondensation technique to successfully synthesize a small library of polyesters using various aliphatic diols and a furfural-derived cyclobutane diacid as precursors. The resulting polymers exhibited good thermal stability and char-forming properties, along with tunable *T<sub>g</sub>*. The halophilic microorganism *Halomonas bluephagenesis* has been considered as a production platform strain for various PHAs, chemicals and proteins under

open nonsterile and continuous conditions. Chen *et al.* (<https://doi.org/10.1039/D5GC02422F>) engineered *H. bluephagenesis* to synthesize a copolymer of short chain length PHA (SCL PHA) and long chain length PHA (LCL PHA), called SCL-*co*-LCL PHA. Parra *et al.* (<https://doi.org/10.1039/D4PY01076K>) developed a photo-surfactant-free polymerization (photo-SFEP) process to synthesize polymeric nanoparticles under mild conditions. In their work, the rapid photolysis of TPO-Na is involved, generating anionic radical species to initiate the polymerization of glycidyl methacrylate (GlyMA). Such photo-SFEP processes share advantages in ensuring colloidal stability over several months, even under varying environmental ionic strengths.

Besides innovative synthetic processes, structural design is also an efficient strategy to develop sustainable polymers. Nguyen *et al.* (<https://doi.org/10.1039/D5PY00401B>) converted 2,3-dihydroxyterephthalate into a reactive monomer that polymerized into polyesters that exhibit adhesive properties. Elsewhere, taking advantage of the synergistic effect of stereomicrostructure and architecture engineering, four-arm star-shaped poly(3-hydroxybutyrate) (P3HBs) (<https://doi.org/10.1039/D5PY00691K>) consisting of stereo-perfect isotactic, iso-rich, and syndio-rich stereomicrostructures, as well as their linear counterparts of comparable molar mass as controls, have been synthesized through stereochemically and architecturally controlled polymerization. The overall material properties demonstrate dramatic enhancement, including the thermal transitions, crystallization behaviors, mechanical performance, and rheological and gas barrier properties. Reversible bond networks are expected to improve mechanical properties and recyclability of polymers. By introducing a dynamic imine bond network and hydrogen bond network, the mechanical properties (tensile strength and modulus), recyclability and reprocessability of polyurethane vitrimers show great improvement. This property enhancement can be ascribed to the incorporation of

dynamic imine bonds, which results in an increase of the number of hydrogen bonds in the PU vitrimers, leading to an improvement in the degree of micro-phase separation between soft and hard segments (<https://doi.org/10.1039/D5PY00728C>). In order to improve upcycling potential, CO<sub>2</sub>-binding alcohols (CO<sub>2</sub>BALs) were used as nucleophiles for PVC functionalization (<https://doi.org/10.1039/D5PY00350D>).

## CO<sub>2</sub>-based polymers

Owing to its abundance, low toxicity, nonflammability, and availability, carbon dioxide (CO<sub>2</sub>) is regarded as a renewable one-carbon building block for chemical synthesis. The incorporation of CO<sub>2</sub> into the polymer backbone presents a transformative approach to converting greenhouse gases into useful polymers.<sup>4–8</sup> Taking the work of Meng *et al.* (<https://doi.org/10.1039/D5PY00272A>) as an example in case, the copolymerization of propylene oxide (PO), phthalic anhydride (PA), and CO<sub>2</sub> in combination with PLA is capable of yielding semi-crystalline copolymers, PPC-P-co-PLA. This is achieved through transesterification reaction catalyzed by Lewis acid–base pairs in a one-pot/one-step method. Compared to PPC-P, the copolymers deliver a higher glass transition temperature and an elevation in thermal decomposition temperature. The optimal mechanical and rheological properties of the PPC-P-co-PLA multi-block copolymers are achieved at a PLA concentration of 8 wt%. By copolymerization of CO<sub>2</sub> with various biobased polyols (*e.g.*, cashew nutshell liquid, vegetable oil, and lignocellulose), a series of self-blown isocyanate free polycarbonate foams are generated (<https://doi.org/10.1039/D4GC06429A>). As a forward-looking solution to address environmental challenges, this foam effectively supports the germination of different types of vegetable seeds. In order to realize enantioselective Ring-Opening Copolymerization (ROCOP) of cyclohexene oxide (CHO) and CO<sub>2</sub>, Rusconi *et al.* (<https://doi.org/10.1039/D5GC00523J>) systematically investigated

the factors affecting copolymerization of CO<sub>2</sub> and *meso*-epoxide *via* DFT calculations combined with an activation strain model and noncovalent interaction analysis. They identified key factors influencing ROCOP enantioselectivity, including monomer deformation, ligand steric effects dictated by the number of chiral centers, and noncovalent interactions, all contributing additively to the observed selectivity.

## Conclusion

This themed collection is a platform to showcase the recent significant achievements in the science and technology of sustainable polymers. By bringing together excellent global expertise, these articles demonstrate the shared commitment from the global chemical science community to address one of the most enduring challenges to create a more sustainable future. Looking forward, several key opportunities are poised to shape the next stage of this field. Despite plenty work having been done toward a sustainable plastics economy, there is still much work to be done, including developing new sustainable monomers and new technologies to deconstruct polymers into meaningful products. Furthermore, currently the recycling of polymers is mainly focused on thermoplastic ones. Much attention should be paid to recycling thermosets, particularly those that do not contain inherent dynamic bonds. Furthermore, a wide range of engineering, economic, and environmental factors should be taken into account for creating a green perspective in the polymer industry. Finally, for many processes, it is necessary to establish standards to guide operations.

This collection features numerous valuable contributions that could not be covered in this editorial due to space constraints. It is impossible for us to achieve success in such a hot themed collection without excellent contributors and peer reviewers. On this occasion, we would like to extend our gratitude to all contributors for providing groundbreaking research to enrich this collection and

all reviewers for voluntarily providing professional comments to facilitate the high quality of this collection. Meanwhile, the editorial office has also made a great contribution to this collection, so we are also grateful to the editorial offices of *Green Chemistry*, *Polymer Chemistry* and *RSC Sustainability*. Hopefully, this themed collection will provide a picture of the current status of sustainable polymer research and development.

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