



## Introduction to Plastic Conversion

Cite this: *Catal. Sci. Technol.*, 2023,  
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DOI: 10.1039/d3cy90036c

rsc.li/catalysis

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Decades of polymer science have yielded a variety of functional materials essential to modern life. These materials extend the shelf-life of food, improve the hygiene of products, allow us to have modern medical treatment and are used in our transition to more sustainable energy sources. The diversity of polymers, their light weight, low cost, good barrier properties, electrically insulating and persistent nature make them hard to replace. However, the focus on application of plastics has often led to a neglect of end-of-life use, leading to more and more plastic

waste accumulating in landfills and if not managed properly, leakage into the environment. For chemists, this calls for innovation both in material design as well as in polymer conversion routes. In this cross-journal collection we highlight outstanding contributions representing the switch from an emphasis on application to a focus on developing versatile and robust ways to chemically convert waste polymer to high value chemicals thereby enabling the circular economy. Both approaches in unison can lead to the design of more recyclable and sustainable solutions.



Ina Vollmer

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*Science and Technology (graduation 2019). After a two-year postdoc on chemical recycling of plastic and nanoplastics detection at Utrecht University, she was appointed as a tenure track Assistant Professor in 2021 at the same university.*



Haritz Sardon

*Haritz Sardon has been an Associate Professor at the University of Basque Country since 2021. He graduated from the University of Basque Country in 2011 with honors before joining the group of Dr Hedrick at IBM-Almaden Research Center as a postdoc in 2012, where he spent 2 years. In 2014 he returned to Spain with a Spanish Ministry grant and joined POLYMAT as group leader. Haritz Sardon has participated in 125 peer-reviewed*

*publications, with more than 60 as corresponding author. The impact of his work can be measured by the increasing number of citations, near 1500 in 2022. He has received several awards including the ACS Macromolecules Young Investigator Award, Excellence of Young Researcher in Chemistry Award by the Spanish Royal Society (2021) and the Excellence of Young Researcher in Polymers Award by the Grupo Español de Polímeros (2020).*

Chemical recycling of existing plastic can replace chemical production of fuels and chemicals from fossil resources and is already commercially applied as one piece in the sustainability puzzle. The breadth in polymer variations however calls for both single polymer applications as well as non-selective all-consuming solutions like pyrolysis. In both cases, catalysts can play an important role in reducing processing temperature and time as well as increasing product selectivity. In addition, chemical solutions for contamination removal are crucial. The complexity of a circular economy has been described by Sharmila and Banu (<https://doi.org/10.1039/D2CY02066A>), who also review examples of novel ways for plastic conversion using nanocatalysts, biotechnology, microwave assisted pyrolysis and plasma assisted gasification as well as photoreforming. A review focussed on hydroconversion of polymers is provided by Liu *et al.*

(<https://doi.org/10.1039/D2CY01886A>). For specific plastics, the development of innovative catalytic systems will facilitate more efficient upcycling of polymers. For example, Wang *et al.* explored the use of the  $\text{Mg}(\text{HMDS})_2$  catalyst in a closed-loop chemical recycling process for poly( $\epsilon$ -caprolactone) (PCL) (<https://doi.org/10.1039/d2py00953f>). Using process intensification, this study achieved the “ $\epsilon$ -CL to PCL to  $\epsilon$ -CL” cycle and the “PCL to  $\epsilon$ -CL to PCL” cycle. Guerrero-Sánchez *et al.* developed a catalytic glycolysis system for poly(ethylene terephthalate) (PET) utilizing a thermo-responsive polymer catalyst ( $[\text{PIL}]_m\text{Zn}_n\text{Cl}_n$ ), which provided the benefits of both homogeneous catalysis and heterogeneous catalysis by taking advantage of the catalyst's upper critical solution temperature (UCST) behavior in glycolic solvents (<https://doi.org/10.1039/d2py01520j>). This system not only realized the highly efficient catalytic

glycolysis of PET, but also the recovery of the catalyst. Another zinc-based homogeneous catalyst system for PLA and PET solvolysis was developed by Mazzeo *et al.* (<https://doi.org/10.1039/D2CY01092E>). The authors developed a homoleptic  $\text{Zn}(\text{II})$  complexes based on tridentate phenoxy-imine pyridine ligands that was more active at lower concentration and shorter reaction times than the benchmark  $\text{Zn}(\text{OAc})_2 \cdot 2\text{H}_2\text{O}$  catalyst. The authors also demonstrated the activity of these catalyst complexes for ring opening polymerization reactions. To enrich the types of degradation products, Wang *et al.* carried out the glycolysis of PET with bio-based cardanol diol to obtain bio-based degradation products, providing a technical path for the diversification of PET upcycling (<https://doi.org/10.1039/d2py01506d>).

In addition to chemical recycling of commercial plastics, the direct synthesis of highly recyclable plastic products is



**George W. Huber**

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**Zhibo Li**

*Prof. Zhibo Li obtained his B.S. (1998) and M.S. (2001) degrees from the University of Science and Technology of China (USTC). He then completed his Ph.D. at the University of Minnesota under the supervision of Prof. Tim Lodge and Prof. Marc Hillmyer in 2006 before joining the group of Prof. Tim Deming in UCLA as a postdoctoral fellow. In late 2008, he became a faculty member in the Laboratory of Polymer Physics and Chemistry*

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another strategy to solve the plastic challenge. Kohsaka *et al.* described a new monomer (2-methylene-4*H*-benzo[*d*]-[1,3]dioxin-4-one, MBDO) that can be used to produce degradable polymers through vinyl polymerization (VP) and ring-opening polymerization (ROP) (<https://doi.org/10.1039/d2py01181f>). All the polymers could be easily degraded into the starting monomer, thus enabling efficient cycling between monomers and polymers. Similarly, Zhu *et al.* synthesized aromatic monomers (DHB-R and DHN-R) to prepare polymers with high thermal stability through a ROP reaction, which could be selectively depolymerized into the corresponding monomers (<https://doi.org/10.1039/d2py01491b>).

The raw materials used to create plastics can also be shifted from non-renewable fossil fuels to renewable resources. For example, Grignard *et al.* discussed how CO<sub>2</sub> can be used as a feedstock to produce polycarbonates (<https://doi.org/10.1039/d2py01258h>).

These approaches provide promising avenues for the development of sustainable plastic products. One example for making biosourced terephthalic acid is presented by Saraci *et al.* (<https://doi.org/10.1039/D2CY01337A>). Biomass-resources can also provide new functionality to polymers that are traditionally produced from petroleum monomers. For example, Saito *et al.* took advantage of the photo-responsive property of the  $\alpha,\beta$ -unsaturated ester moiety of the *p*-hydroxycinnamic acid structure synthesized from renewable lignin-oxidation compounds, to develop switchable adhesives (<https://doi.org/10.1039/d2py01474b>).

The implementation of self-healing plastics possesses the potential to mitigate the plastic waste produced. By introducing Cu-S dynamic reversible bonds into brittle plastic poly(cyclohexene carbonate) (PCHC), Wang and co-workers developed a novel elastomer, which possesses self-healing properties that allowed for reprocessing

(<https://doi.org/10.1039/d2py01002j>). Slugovc *et al.* demonstrated the feasibility of the oxa-Michael reaction using a model system and evidenced the self-healing properties of poly(ethers) derived from oxa-Michael reactions (<https://doi.org/10.1039/d2py01345b>). Chan-Seng *et al.* designed interfacial agents for polymer blends to enhance the mechanical properties (<https://doi.org/10.1039/d2py01495e>). The extension of the service life of plastics will fundamentally limit the generation of waste plastics.

The recycling of plastics is of significant economic and social value. As demonstrated in this collection, the efficient degradation of plastic waste, the rational development of new recyclable plastics, using renewable resources to synthesize plastics, and the enhancement of plastic product properties to extend service life are crucial measures in improving the overall recyclability and environmental sustainability of the plastics industry.