

# Green Chemistry: A Framework for a Sustainable Future

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For more than three decades, Green Chemistry has provided a framework for chemists and chemical engineers to do their part in contributing to the broad scope of global sustainability. American Chemical Society journals are a great venue for these scientists to share their latest results and provide a resource to the chemistry community and beyond for understanding current problems and envisioning solutions. We believe this is an opportune time to highlight some of the leading articles on the broad theme of Green Chemistry being published today through a Virtual Issue of selected works from nine ACS chemistry and engineering journals.

The inception of this Virtual Issue is no coincidence. We have timed it to the 2021 Green Chemistry & Engineering Conference taking place virtually June 14–June 18. Now celebrating its 25th edition, we have seen progress toward more sustainable chemistries being showcased and celebrated at each GC&E conference.<sup>1</sup> The theme of this year's meeting, "Sustainable Production to Advance the Circular Economy", is particularly bold. It highlights the recognition that contributions must take into account a systems approach to reducing environmental impact through intentional design of chemical products, not just considering how raw materials are sourced and in the manufacture and use of industrial and consumer goods but also how these materials and goods may be reused, recycled, or upcycled. The embrace of life-cycle thinking as a goal among the Green Chemistry community comes at the backdrop of the realization of limited resources and a climate crisis the likes of which most of us still fail to fully comprehend. We believe these selected articles are stepping stones on the pathway to advance closed-loop economies while still serving as models for innovation at a fundamental level within their respective chemistry subdisciplines.

The drive for efficiency in organic synthesis merges the best of the idealisms of the Enlightenment and the Renaissance. Certainly, there is a premium on rationalism, with an aspiration of mechanistically sound reaction design and process development. But at the same time, the aesthetic appeal of the new ideas that culminate in the advances we now see routinely is unmistakable. There is no constraint on curiosity when we consider the boundary conditions of efficient, environmentally benign processes. On the contrary, these considerations spawn new concepts and approaches, ranging from postmodern expansions of photochemistry, reconsideration of seminal thinking about solvation, importation of physical and mechanical phenomena, to reaction development—the creativity born of efficiency considerations now drives major technology innovation in chemistry. *The Journal of Organic Chemistry* and *Organic Letters* are delighted to participate in this Virtual Issue with a selection of perspectives, research

articles, and letters that highlight just some of the most impactful science in this arena, across a very wide swath of chemical space. *Organometallics*, a journal with a long-standing history of reporting fundamental advances in organometallic chemistry, catalysis, and materials, has selected contributions to this Virtual Issue that best highlight the diverse nature of this type of organometallic chemistry and that are likely to impact development of more sustainable chemical processes and the transition to a circular economy.

*ACS Sustainable Chemistry & Engineering* is a world leader in publishing groundbreaking research that addresses the challenges of sustainability, advancing the principles of Green Chemistry and Green Engineering with global reach and impact. Key coverage includes catalysis with emerging feedstocks and synthetic methods for preparing materials and chemicals in a sustainable way to help bring critical innovations from a research setting to commercialization. The journal takes pride in its central role in promoting innovations that will enable the implementation of a circular economy. *Industrial & Engineering Chemistry Research* publishes many, many papers in the Green Chemistry and sustainability space as these are core considerations in applied chemistry and chemical engineering. The ten articles in this Virtual Issue are just the tip of the iceberg and represent the types of papers readers can find in *I&EC Research*. We would encourage those interested in the Virtual Issue to browse through other recent issues of the journal, where they will quickly find other articles aligned with the themes of the conference. Contributions from *Environmental Science & Technology* and *Environmental Science & Technology Letters* clearly illustrate the interdisciplinary systems approach that is required to address key environmental challenges that impede sustainability efforts—from remediation of pollution to design of next-generation safer and functional chemicals. Several of the high-impact contributions selected have been the subject of media coverage. Contributions from *ACS Omega*, an interdisciplinary open-access journal, were selected to highlight the potential impact of open-source publications and their importance in connecting scientists across industry and academia. With a similar goal, *Organic Process Research & Development* has a tradition of bridging industrial and academic research. Its focus on process

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chemistry as the science that enables the safe, environmentally benign, and economical manufacturing of chemicals is evident in the selected articles for this Virtual Issue, which highlight aspects of catalysis, synthetic methodology development, and synthetic strategy exploration that are needed to enable circular economies.

**Minimizing Dependence on Fossil Fuels.** The contributions from across these journals demonstrate that the design of methods for circularity necessitates multidisciplinary approaches and careful definition of the problem being addressed.<sup>2</sup> One of the key challenges we face continues to be decreasing our dependence on fossil fuels for chemical and fuel production. This entails both the efficient and clean transformation of renewable (biobased) raw materials into functional chemicals and fuels, as well as efficient CO<sub>2</sub> capture and conversion into fuels and chemicals. A number of exciting advances in this Virtual Issue highlight our progress on both fronts. In a critical review, Wang and Su et al. (DOI: 10.1021/acs.est.9b01453) discuss the development and application of engineered multifunctional nanohybrids of carbon nanomaterials and metal/metal oxide nanoparticles, which exhibit promising multifunctionalities for addressing the critical energy–water–environment (EWE) nexus.

More specifically on the topic of CO<sub>2</sub> capture and use, Hatton et al. (DOI: 10.1021/acs.iecr.0c04512) identify a key challenge for carbon capture and storage (CCS)—the quest for net-negative emissions. They report that bioenergy with carbon capture and storage (BECCS) with molten sorbents offers a unique opportunity to realize net-negative emissions with minimal indirect emissions and low-cost separation of CO<sub>2</sub> from other gases. BECCS could remove 300–850 kg of CO<sub>2</sub> equivalents from the atmosphere per megawatt-hour of electrical output (kg/MWh<sub>e</sub>), making it advantageous to other low carbon technologies and allowing the offset of emissions from hard-to-abate industries. In the search for efficient materials for carbon capture, Sun et al. (DOI: 10.1021/acs.iecr.0c04126) report the development of light-responsive metal–organic frameworks (LMOFs), which have tunable structures and performances. While conventional amines cannot achieve controllable adsorption separation, these LMOFs have tunable amine-based active sites, which enhance CO<sub>2</sub> capture and control adsorption and separation. Several advances also highlight the improvement of technologies for reduction of CO<sub>2</sub> based on electrocatalytic methods, including the use of Ru and Re catalysts (DOI: 10.1021/acs.organomet.9b00815) and the use of ionic liquids for this application (DOI: 10.1021/acs.iecr.0c04037), as well as exploration of Earth-abundant iron catalysts (DOI: 10.1021/acs.organomet.8b00711). Complementary to electrochemical methods, enzymatic (DOI: 10.1021/acs.est.9b05284) and photocatalytic (DOI: 10.1021/acs.iecr.0c04126) methods are also shown to have a niche in the toolbox for conversion and utilization of CO<sub>2</sub>, as highlighted in reports by Sun et al. and Jin and Kim et al., respectively.

The efficient and clean transformation of renewable raw materials into functional chemicals and fuels has blossomed from a theoretical construct to an interdisciplinary field that is fueled by fundamental innovations in synthetic chemistry and guided by practical applications of biorefineries. The requirements of biorefineries for flexibility of feedstocks, relatively mild operational conditions, and low environmental impact have resulted in highly innovative methods for defunctionalizing and refunctionalizing biomass feedstocks to value-added chemicals and materials. On the defunctionalization front,

Stephenson et al. report an organocatalytic method for photochemical C–O bond cleavage of the β-O-4 linkage in lignin scaffolds. This method offers a metal-free strategy to prior reports and is applicable to continuous flow processing (DOI: 10.1021/acs.orglett.0c03029). Cellulose-derived platform chemicals can also be obtained more efficiently from using a redox-switchable biocatalyst for controllable oxidation or reduction of 5-hydroxymethylfurfural (HMF) into high-value derivatives (DOI: 10.1021/acsomega.0c02178).

Expanding the gamut of new platform chemicals that can be obtained from biomass, Rio et al. describe the presence of valuable phenolic compounds incorporated into lignins, such as flavonoids, hydroxystilbenes, and hydroxycinnamic amides, which behave as authentic lignin monomers and are potentially available in large amounts from the abundant in waste products from processing of agricultural or forest biomass (DOI: 10.1021/acssuschemeng.0c01109). Tomishige et al. discuss the utility of erythritol as a C<sub>4</sub> platform in biomass refinery, derived from fermentation of sugars and glycerol (DOI: 10.1021/acsomega.9b04046).

The direct valorization of lignin to functional chemicals presents potential new avenues for efficiently converting lignin to chemical products. Yin and Huang et al. report a lignin–carbohydrate complex that is a potent antioxidant for scavenging reactive oxygen species *in vitro* and zebrafish *in vivo* (DOI: 10.1021/acssuschemeng.9b05290). Lignin was also used in the manufacture of polyurethane formed by oxidative liquefaction (DOI: 10.1021/acsomega.1c00285). Many other transformations of biomass to functional chemicals and materials are discussed in two timely reviews on biomass-derived carbonaceous materials (DOI: 10.1021/acssuschemeng.8b06550) and bacterial cellulose-based composite scaffolds for biomedical applications (DOI: 10.1021/acssuschemeng.0c00125). On the critical front of transforming our polymer platform to renewable materials, consistent with the circular economy, Vodovotz et al. discuss how we can narrow the gap for bioplastic use in single-use food packaging, focusing on the most recent development successes in bioplastic materials and highlighting the “gaps” between bioplastics and their conventional counterparts with respect to their properties (DOI: 10.1021/acs.est.9b03755).

**Minimizing the Impact of Chemical Synthesis and Manufacturing.** Another key challenge in developing manufacturing infrastructure for a circular economy remains the minimization of the overall environmental footprint, which must minimize both the environmental impact of the manufacturing processes of chemicals as well as their potential hazard and persistence of the commodity chemicals produced. While the former has been a cornerstone of research in Green Chemistry from its inception in the early 1990s, it is still a fertile area of research, and we see evidence of that in this Virtual Issue. The latter will be addressed in further detail later in this Editorial.

Sustainability in organic chemistry, especially in organic synthesis, has been driving innovation for decades. With the amount of waste generated in many synthetic chemistry routes, especially at scale in manufacturing, we are faced with not only an ethical imperative to develop more sustainable chemical processes and products but also a financial imperative. Metrics to gauge our progress, including process mass intensity (PMI), have been developed that allow all aspects of a process to be compared. For example, conducting a reaction in water may not necessarily be an improvement if several volumes of an

organic solvent are needed to extract/purify the product. One could even argue that water can be problematic because its high boiling point makes recycling energy intensive, but we do not know these details without consciously thinking about them. A recent perspective highlights the need to explicitly include the assessment of sustainability using Green Chemistry metrics.<sup>3</sup>

A number of studies highlight advances in catalysis and reaction engineering that have led to waste and energy minimization for processes that still pose significant challenges. Four of the contributions from *Organic Letters* focus on use of less harmful reagents, catalysts, or routes (e.g., DOI: 10.1021/acs.orglett.1c00850; DOI: 10.1021/acs.orglett.0c03272; and DOI: 10.1021/acs.orglett.0c01043). On the catalysis front, we see an increase in the scope of synthetic transformations facilitated by supported catalysts (DOI: 10.1021/acs.organomet.9b00082; 10.1021/acs.orglett.1c01058). In one example, Pei, Lu, and Cai et al. described the preparation of a reusable magnetic Ag–Fe<sub>3</sub>O<sub>4</sub> catalyst supported on cellulose microspheres for reduction of nitrophenols (DOI: 10.1021/acsomega.0c00437). We also see increased exploration of the utility of base metals in catalysis and organocatalytic processes being performed under mild conditions and with low catalyst loading: for highly enantioselective epoxidation of  $\alpha,\beta$ -unsaturated ketones, Jarczak et al. employed an amide-based *Cinchona* alkaloid at loadings as low as 0.5 mol % as a hybrid phase-transfer organocatalyst (DOI: 10.1021/acs.orglett.0c03272).

On the application front, the quest for reducing traditionally high *E*-factors in the pharmaceutical sector continues. In two critical perspectives, colleagues from major pharmaceutical companies highlight the need to develop less wasteful and toxic methods for peptide and oligonucleotide synthesis and purification methods, in light of the growing utility of biological peptide-based pharmaceuticals (DOI: 10.1021/acs.joc.0c02291; 10.1021/acs.joc.8b03001). In another perspective in *The Journal of Organic Chemistry*, Rossen highlights insights from process chemistry that can reduce the impact of synthetic chemistry at smaller scales (DOI: 10.1021/acs.joc.9b00344). In *Organometallics*, Leahy and colleagues provide a perspective from the pharmaceutical industry on the role of catalysis with Earth-abundant metals (DOI: 10.1021/acs.organomet.8b00566).

Catalysis with Earth-abundant rather than precious metals continues to be an active area of research and key component of sustainable chemistry research. Manuscripts in *Organometallics* focus on cobalt (DOI: 10.1021/acs.organomet.0c00241) and iron (DOI: 10.1021/acs.organomet.1c00200) catalysts for hydrosilylation and nickel precursors (DOI: 10.1021/acs.organomet.0c00485) and heterogeneous catalysts (DOI: 10.1021/acs.organomet.9b00082) for Suzuki–Miyaura coupling, one of the most widely used methods for C–C bond formation. Pidko et al. also report the role of manganese in mediating C–C bond formation from organoboranes (DOI: 10.1021/acs.organomet.0c00781).

Waste reduction also continues to be tackled on the solvent front in the development of alternative solvents, such as less hazardous task-specific ionic liquids (DOI: 10.1021/acsomega.9b04091), deep eutectic solvents (DOI: 10.1021/acs.joc.0c02039; 10.1021/acssuschemeng.8b03520), and increasing the scope and efficiency of reactions in water, such as amidation (DOI: 10.1021/acs.orglett.0c01676), amine synthesis, and transfer hydrogenation (DOI: 10.1021/acs.

organomet.0c00554; 10.1021/acs.organomet.1c00133). We also see the development of new biomass-derived solvents with greener profiles, such as glycerol-derived 1,2,3-triethoxypropane and systematic solvent selection protocols for “greener” solvents (DOI: 10.1021/acs.iecr.0c03789; 10.1021/acs.oprd.0c00326).

**Designing Chemicals with Minimal Hazard.** While minimizing the impact of chemicals through use of renewable resources and cleaner process technologies remains critical, the importance of the toxicological profile of the chemicals being circulated is now increasingly being recognized. The challenge of efficiently assessing the toxicological risks of thousands of chemicals at the design stage and for chemicals already in use requires interdisciplinary research efforts. Many toxicological mechanisms of interaction remain poorly understood and therefore unregulated. In this Virtual Issue, Shi et al. describe how *in silico* methods of molecular dynamic simulations will help to define molecular initiation events and identify toxicological mechanisms, enabling quick and effective screening of a wider range of chemicals (DOI: 10.1021/acs.estlett.9b00073). Blum et al. and Kwiatkowski et al. further propose that individual testing of thousands of chemicals is unrealistic, and chemical “classes” should be screened, using broad subclasses as required. Venier et al. (DOI: 10.1021/acs.estlett.9b00582) provide excellent examples of the regrettable substitution of polybrominated diphenyl ethers (PDBEs) with organophosphate ester flame retardants (OPFRs), resulting in undesirable toxicological outcomes, and Kwiatkowski et al. consider the scientific bases for dealing with perfluoroalkyl (PFAS) chemicals as a class (DOI: 10.1021/acs.estlett.0c00255).

The design of plastics to be both biodegradable and sustainably produced from nonpetrochemical sources is another critical circular design area requiring urgent research input. The *ES&T* paper by Napper and Thomson (DOI: 10.1021/acs.est.8b06984) reports a lack of biodegradable plastic formulations used for carrier bags that offered rapid rates of decay compared to conventional plastic bags. For environmental concerns, Nguyen et al. (DOI: 10.1021/acs.estlett.8b00671) provide a novel and simple method in *ES&T Letters* to assess the quantities of microplastic fragments in environmental matrices, using hydrophobic iron nanoparticles to extract microplastics from soil, sediments, and water magnetically, and could be used as a sustainable remediation tool.

**Sustainable Water Resources.** The sustainable supply of potable drinking water is a global imperative and was identified as a United Nations Sustainable Development Goal in 2015. Just last month in *ACS Omega*, Gude et al. provided a critical evaluation and perspective of two major routes for developing more sustainable and circular-economy-based wastewater treatment and showed that integrating both concepts may result in superior energy and resource efficiency for wastewater treatment systems (DOI: 10.1021/acsomega.0c05827). Zodrow et al. (DOI: 10.1021/acs.estlett.0c00019) report on the use of self-healing bacterially formed cellulose fiber networks creating sustainable biological membranes (Live Filtration Membranes) capable of filtering water for drinking water purposes. Alternative energy-efficient and sustainable remediation approaches include the application of UV/sulfite water treatments to offer effective degradation of persistent PFAS chemicals in raw water for onward drinking water use, reported by Liut et al. in *ES&T Letters* (DOI: 10.1021/acs.estlett.0c00236).

Environmentally sustainable and energy-efficient nanotechnology membranes also have water purification applications, often in combination with other environmental uses, including energy harvesting, environmental sensing, and remediation. Desalination technologies with superior high efficiency are based on selective desalination membranes (Geise et al.; DOI: [10.1021/acs.estlett.9b00351](https://doi.org/10.1021/acs.estlett.9b00351)) and reduced graphene oxide membranes with minimal impact of nanowrinkles (Zhang et al.; DOI: [10.1021/acs.estlett.0c00143](https://doi.org/10.1021/acs.estlett.0c00143)). In addition to desalination, the degradation of persistent organic pollutants is also a key priority for water purification. Li, Li, and Zhang et al. (DOI: [10.1021/acs.est.8b05685](https://doi.org/10.1021/acs.est.8b05685)) report on the use of a porous, coral-like nanostructure photoelectrode for this purpose while simultaneously producing electricity. Luo et al. describe mechanisms of carbo-catalysis in carbon nanotubes with persulfate oxidation for water treatment applications (DOI: [10.1021/acs.est.0c02645](https://doi.org/10.1021/acs.est.0c02645)).

**Closing the Loop.** The environmentally sustainable recovery of resources from wastes is also a burgeoning area of research. In a highly cited account, Suh and Scott et al. report benchmarks for degradation rates of common plastics, which will help prioritize future research efforts on chemical degradation to close the loop (DOI: [10.1021/acssuschemeng.9b06635](https://doi.org/10.1021/acssuschemeng.9b06635)). In *Organometallics*, O'Hair and Ryzhov et al. explore the depolymerization of ethylene with palladium catalysts to understand approaches to remediation of polyolefins (DOI: [10.1021/acs.organomet.0c00782](https://doi.org/10.1021/acs.organomet.0c00782)). Li et al. report on the recovery of lithium from spent batteries using an acid-free mechanochemical process that utilizes salt and sodium carbonate as the sole reagents (DOI: [10.1021/acs.est.9b01919](https://doi.org/10.1021/acs.est.9b01919)). Lo et al. provide a critical review on the use of selective sorbents for the removal and recovery of phosphates from water and wastewaters (DOI: [10.1021/acs.est.9b05569](https://doi.org/10.1021/acs.est.9b05569)). Munasinghe-Arachchige and Nirmalakhandan (DOI: [10.1021/acs.estlett.0c00355](https://doi.org/10.1021/acs.estlett.0c00355)) describe the recovery of nitrogen from sewage sludge and livestock manure as offering some circularity to the high global demand for nitrogenous fertilizers. The capture and conversion of carbon dioxide to bicarbonate is reported by Kim and Jin et al. (DOI: [10.1021/acs.est.9b05284](https://doi.org/10.1021/acs.est.9b05284)) using an environmentally simple system of stabilized carbonic anhydrases loaded onto electrospun polymer nanofibers.

**Lifecycle and Systems Thinking.** The ability to apply life-cycle thinking to prioritize research and propose truly long-term, sustainable solutions is critical for identifying feasible ways to address circularity. Two common life-cycle assessment (LCA) methods—process LCA and economic input–output LCA—both suffer from different shortcomings. As a result, many hybrid methods are used to improve the overall accuracy. Luo and Ierapetritou analyze different hybrid LCA methodologies through a case study of two biomass-based *p*-xylene production technologies and provide key insights applicable to future LCA analysis (DOI: [10.1021/acs.iecr.0c04709](https://doi.org/10.1021/acs.iecr.0c04709)). LCA analyses of the synthesis of widely used titanium dioxide nanoparticles (DOI: [10.1021/acs.est.8b06800](https://doi.org/10.1021/acs.est.8b06800)) and butyl acetate (DOI: [10.1021/acs.iecr.0c04233](https://doi.org/10.1021/acs.iecr.0c04233)) provide theoretical guidance for more sustainable production of both. Parkinson and Hunt apply such an approach in proposing the conversion of agricultural land in groundwater-stressed areas from irrigated crop types to rain-fed crop types combined with diversification to solar harvesting, so-called “agrivoltaics” (DOI: [10.1021/acs.estlett.0c00349](https://doi.org/10.1021/acs.estlett.0c00349)).

**Future Research Needs in Developing a Circular Economy.** The selected publications in this Virtual Issue exemplify the diverse and interdisciplinary contributions surrounding the theme of this year's conference. They also allow us to identify areas that will require further emphasis in order to strategically advance the field toward applications that facilitate circularity. Some of these areas include:

- Depolymerization and defunctionalization methods for existing chemicals that can allow circularity, especially for plastics;
- Design of circular systems with consideration of human health and ecotoxicity, ideally via rational design of benign commodity chemicals;
- Systematic application of life-cycle analysis, or thinking, and process metrics in developing new manufacturing/synthetic routes;
- Implementation of machine learning and other big-data methods to drive innovation toward new paradigms of circularity.

In order to allow future contributions to highlight the potential application to circular economies, authors may want to consider explicitly addressing some of following questions in their manuscripts. First, consider the source of the materials being used, and if it is fossil-fuel-derived, consider if there is a biobased alternative that could provide the same chemical insights. If elements are used that are facing limited supply, consider whether the synthetic routes and processes could help reduce the use of such materials. Second, discuss whether the method described could impact different stages of a chemical's life cycle. For example, is it a step that is potentially reversible based on kinetics and thermodynamics? If it produces byproducts, could those be a valuable resource? And finally, consider whether the method or product constitutes a safe product that can be either recycled, reused, or remanufactured. Addressing such questions forces the authors to apply life-cycle thinking and to broaden their discussion of the prior art most closely related to the advances presented. We look forward to future fruitful contributions to this field from the innovative authorship of the ACS family of journals.

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## Notes

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