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Getting the Terms Right: Green, Sustainable, or Circular Chemistry?

Hatice Mutlu and Leonie Barner**

Green chemistry, sustainable chemistry, and circular chemistry are important concepts for the modern lifestyle, current research directions, and worldwide industries. These three concepts are closely related and interconnected but cannot be used synonymously. In addition, they are addressing two different economic models, i.e., linear economy and circular economy. The current contribution focuses on the importance of these decisive chemistries for the development of a sustainable future and their role in the realm of circular economy and the planetary boundaries framework—especially for the planetary boundary of "novel entities." Researchers active in the field of polymer chemistry play an important role as plastic pollution and resource in addition to environmental depletion, caused by the still increasing production of polymers and plastics, become more and more pronounced. It is also reported that multi- and interdisciplinary approaches are needed to develop solutions for a sustainable future.

1. Introduction

When polymer chemists strive to consider sustainability in their field, understanding the differences between green or sustainable chemistry as well as circular chemistry is critical. Although closely related, these are distinctively different terms that cannot be used interchangeably, and describe different fields of research. We found that the terms green/sustainable and circular are sometimes confused by chemists in academia and allied industries, and are incorrectly used as synonyms for each other. In other words, based on our experience, some researchers lack knowledge of the distinctive central elements of those concepts and therefore clarification of these related but nevertheless different concepts would be beneficial.

For example, switching a production process from an organic solvent to water is a contribution to green chemistry as water is a benign solvent.[1] However, it might not be a sustainable process if too much water is extracted from natural sources for production or if the used water is not sufficiently cleaned when released into the sewer (which would also be in violation of green chemistry principles, see Section 2.1).[2] In addition, the product itself might not be circular, i.e., kept in material circulation as long as possible. Green chemistry and circular chemistry refer to two different economic models, i.e., the linear economic model and the circular economic model, and are based on different principles (see Sections 2.1 and 4.1). Applying the principles of green or circular chemistry to the same chemical process can result in distinguishably different outcomes for the triple bottom line, i.e., social, environmental, and economic sustainability or also expressed as "People, Planet, Profit".[3]

2. Green Chemistry

According to the International Union of Pure and Applied Chemistry (IUPAC),[4] green chemistry is closely related to the "design of chemical products and processes that particularly eliminate the use or generation of substances hazardous to humans, animals, plants, and the environment". In other words, the concept of green chemistry addresses the problem of chemical pollution and refers to the design of chemicals and their production processes reducing or eliminating the use or generation of hazardous substances.[5] Green chemistry[6,7] is centered around twelve principles (see Section 2.1) which describe how a chemical should be produced, and used in a clean and green way reducing the material use and pollution of the environment. These twelve guiding principles focus on the direct sustainability assessment of chemical reactions and are perfectly suited for the optimization of linear production routes.[8] Most of the twelve principles focus on design principles for the production of chemicals, e.g. atom and energy efficiency, use of safer solvents, and catalysts. Some relate to the environmental impact of chemical production, e.g. preventing waste, product degradability at end-of-life, as well as prevention of pollution and accidents. While green chemistry is

H. Mutlu
Soft Matter Synthesis Laboratory (SML)
Karlsruhe Institute of Technology (KIT)
Hermann-von-Helmholtz-Platz 1, Eggenstein-Leopoldshafen 76344, Germany
E-mail: hatice.mutlu@kit.edu

L. Barner
Centre for a Waste-Free World, School of Chemistry and Physics
Faculty of Science, Queensland University of Technology
Brisbane, QLD 4000, Australia
E-mail: leonie.barner@qut.edu.au

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an affirmative tool (a guiding philosophy) to reduce pollution and safeguard the environment, green chemistry still serves a linear economy model (i.e., take-make-use-dispose) and not a circular model that keeps materials in circulation for as long as possible. Still, over the past decades, fundamental research in the field of green chemistry has been significant, definitions of its principles have been refined, and green industrial processes have been implemented as reviewed by Erythropel et al.[9] and Zimmerman et al.[10]

2.1. Twelve Principles of Green Chemistry

The twelve principles of green chemistry as defined by the United States Environmental Protection Agency[6] are:

1) **Prevent waste**: Design chemical syntheses to prevent waste. Leave no waste to treat or clean up.
2) **Maximize atom economy**: Design syntheses so that the final product contains the maximum proportion of the starting materials. Waste few or no atoms.
3) **Design less hazardous chemical syntheses**: Design syntheses to use and generate substances with little or no toxicity to either humans or the environment.
4) **Design safer chemicals and products**: Design chemical products that are fully effective yet have little or no toxicity.
5) **Use safer solvents and reaction conditions**: Avoid using solvents, separation agents, or other auxiliary chemicals. If you must use these chemicals, use safer ones.
6) **Increase energy efficiency**: Run chemical reactions at room temperature and pressure whenever possible.
7) **Use renewable feedstocks**: Use starting materials (also known as feedstocks) that are renewable rather than depletable. The source of renewable feedstocks is often agricultural products or the wastes of other processes; the source of depletable feedstocks is often fossil fuels (petroleum, natural gas, or coal) or mining operations.
8) **Avoid chemical derivatives**: Avoid using blocking or protecting groups or any temporary modifications if possible. Derivatives use additional reagents and generate waste.
9) **Use catalysts, not stoichiometric reagents**: Minimize waste by using catalytic reactions. Catalysts are effective in small amounts and can carry out a single reaction many times. They are preferable to stoichiometric reagents, which are used in excess and carry out a reaction only once.
10) **Design chemicals and products to degrade after use**: Design chemical products to break down into innocuous substances after use so that they do not accumulate in the environment.
11) **Analyze in real time to prevent pollution**: Include in-process, real-time monitoring and control during syntheses to minimize or eliminate the formation of byproducts.
12) **Minimize the potential for accidents**: Design chemicals and their physical forms (solid, liquid, or gas) to minimize the potential for chemical accidents including explosions, fires, and releases into the environment.

The common practice of the twelve principles of green chemistry, and explicitly, how they relate to polymer chemistry are a popular topic of discussion in the literature. Generally, principles such as maximizing atom economy (principle 2), and avoiding chemical derivatives (principle 8) along the use of catalysts (principle 9) are well adopted in the field of green polymer chemistry. Nevertheless, certain principles, such as designing less hazardous chemical syntheses (principle 3), the conceptualization of safer chemicals and products (principle 4), the use of safer solvents and reaction conditions (principle 5), the use of renewable feedstock (principle 7), and design of chemicals and products to degrade after use (principle 10) still require further research tying to the progress achieved so far. For instance, several concepts[11,12,13,14] have been investigated to design polymers with programmed degradation (principle 10) that allow both degradation to their constituent monomers or other useful intermediates. The latter was achieved via the introduction of chemically labile groups (such as esters, carbonates, amides, or acetals) and the introduction of stimuli-responsive motifs, which induce degradation of polymers, for example, by temperature, oxidation, or light-induced cleavage amongst others. Crucially, it is necessary to integrate those labile and stimuli-responsive molecules into materials such as crosslinkers, structural units, or side groups. In an exemplary manner, photosensitive *o*-nitrobenzyl moieties were introduced into a polyurethane (in which, the strong urethane linkage and often high degree of crosslinking make them a challenge to degrade without damaging the environment) as structural units.[15] Those respective light-responsive units have enabled the polymer to be broken down upon exposure to UV light. Accordingly, this specific approach is postulated to aid the development of polyurethane-based degradable packaging and adhesives. In a similar manner, completely renewable (principle 7) nonisocyanate polyurethanes (NIPUs) have been synthesized via Lossen rearrangement.[16] In particular, this approach also facilitates the utilization of safer chemicals (principles 3 and 4), as the Lossen rearrangement aids the formation of isocyanates (the main component for the PU synthesis) in situ without the use of phosgene.

In a related way, the utilization of one-pot catalytic transformations combining multiple synthetic steps like multi-component, cascade, domino, or tandem reactions felicitates the enhancement of energy efficiency together with the avoidance of intermediate products, waste, and toxic chemicals. Hence, such reactions fulfill three crucial principles (e.g., principles 4, 5, and 7) of green chemistry. Suitably, Meier et al.[17] have developed a novel concept that combines tandem catalysis and derivatization of cellulose (which is the most abundant biopolymer and an almost inexhaustible source of biomass) by applying a single catalyst for three transformations in the DMSO/DBU/CO_2 switchable solvent system. Applying this approach, cellulose was functionalized with four different biobased isothiocyanates, which were formed in situ via a catalytic sulfurization of isocyanides with elemental sulfur, and therefore avoiding the utilization of highly toxic isothiocyanates. It is importnat to point out that the extensive global production of elemental sulfur by hydrodesulfurization of petroleum/natural gas rafineries is corresponding to 70 million tons annually, which has prompted greater use of this abundant substance not only in organic chemistry but also in polymer synthesis.

The importance and the maintenance of the twelve principles of green chemistry as the key to a greener polymer chemistry have been recently emphasized by Dubé et al.[18,19] While they acknowledge the practicality and effectiveness of using the twelve principles of green chemistry as a guide, they also emphasize the need to reflect on the twelve principles in a holistic way. In other words, Dubé et al. stress that in many cases some of the twelve principles offer conflicting objectives. Exemplary, biodegradable polymer products are not necessarily based on bio-sourced starting material (such as lignin or cellulose). Likewise, the utilization of bio-sourced material could require synthetic approaches that are realized at the cost of significant energy inputs and perhaps exposure to other hazards. Conclusively, we quote Marc Dúbe[11] who has stated that "keeping an eye toward all twelve green chemistry principles should become standard practice for all polymer scientists".

3. Sustainable Chemistry

Although the concept of sustainability was first introduced by Hans Carl von Carlowitz (1645–1714),[20] the term sustainable chemistry was defined by The Organisation for Economic Co-operation and Development (OECD) in the 1990s as "a scientific concept that seeks to improve the efficiency with which natural resources are used to meet human needs for chemical products and services".[21] Based on this definition, sustainable chemistry is closely related to the twelve principles of green chemistry, as sustainable chemistry prioritizes production processes that promote increased product value while intersecting the goals of protecting and enhancing human health and the environment. The concept of sustainable chemistry is also reflected in the Sustainable Development Goals of the United Nations—especially in Goal 12 "Responsible Consumption and Production," which highlights the importance of "decoupling economic growth from environmental degradation, increasing resource efficiency, and promoting sustainable lifestyles."[22]

In 1999, Hutzinger,[23] an Austrian–Canadian environmental chemist, clearly addressed a fundamental difference between these two concepts of chemistry. Accordingly, green chemistry refers to the "design, manufacture, and use of chemicals and chemical processes that have little or no pollution potential or environmental risk," while sustainable chemistry reflects the "maintenance and continuation of an ecologically-sound development". In fact, experts still discuss the borders of the concept of sustainability not only in academia but also at an industrial level. Whereas keywords like safe, clean, efficient, prevention, reduce, recycle, reuse, and biodegradable have been closely related to the concept of sustainable chemistry, it is often encountered that those adjectives are also synonymously used to define the merits of green chemistry. There is also the perception that green chemistry is a characteristic of the microscopic realm of academia, hence with innovative fundamental chemistry, while sustainability[24] is concerned with the macroscopic domain of industrial chemistry. Indeed, the fact that there is no economic component implicit in green chemistry was always seen as a major shortcoming by the industry.[25] Despite the attempts to differentiate both terms, they now start to be referred to as "Green and Sustainable Chemistry."[26,27,28,29]

Inevitably, this conceptual delimitation between green and sustainable chemistry is strongly implied also as one of the most urgent questions in the field of polymer chemistry, particularly regarding how sustainable polymers (plastics) can be developed. Currently, most polymers and plastics are synthesized from nonrenewable or virgin resources and are discarded after their use—not only resulting in an economic loss, but often also in adverse effects on the environment.[30] Economic loss needs to be assessed not only at the production stage of a product but also at the end-of-life stage. Products that are not part of a circular economy generate economic losses due to landfill levies, maintenance of landfills, as well as the need for financial resources to clean up littering and pollution on land and in marine environment—just to name a few.

Researchers from diverse branches of polymer science and engineering are applying their knowledge toward the reduction of plastic waste as well as to improve the recyclability of plastics. In this regard, the National Science Foundation Center for Sustainable Polymers defines[31] "a sustainable polymer as a plastic material that addresses the needs of consumers without damaging our environment, health, and economy" and presents the following six metrics for the synthesis of sustainable polymers:[31]

1) Use renewable feedstocks for production.
2) Use less net water and nonrenewable energy in production.
3) Emit less greenhouse gases during production.
4) Produce less waste in production.
5) Have a smaller carbon footprint.
6) Have a facile end life.

Nevertheless, adopting those sustainability metrics is not sufficient to define polymers as sustainable. In particular because by applying those six principles, scientists encounter several challenges including the conservation of scarce natural resources, conversion of biomass feedstocks, drinking water quality, energy conversion and storage, and sustainable product design. The first metric of sustainability (see above) is clearly imposing the use of renewable feedstocks for the production of polymers. Nevertheless, within the last decade, it became obvious that in order to ensure the sustainable production of polymers and functional materials, alternative feedstocks that are not needed to produce food are also required. Particularly considering that supply chains, already disrupted by COVID-19, have been further complicated by the current situation in Ukraine, which is giving rise to shortages and price increases in vegetable or plant oils that represent renewable resource for polymer chemistry. In other words, it has been prioritized to identify sustainable building blocks that provide monomers already in use or polymers that are functional equivalents to existing macromolecules. In this regard, polymers made from elemental sulfur have emerged as a new class of materials useful in several applications[32] as they are postulated to be green in their preparation and use.[33] In fact, the preparation of sulfur polymers is an innovative example of waste valorization given the fact that elemental sulfur is known for its globally distributed raw material reserves as a by-product of the petroleum industry.[34] In a similar manner, usually the syntheses of the most elemental sulfur-based polymers are acknowledged to be atom economical and often require no solvent. Moreover, the obtained polymeric materials are recognized to have a small environmental footprint. Hence, the valorization of elemental sulfur aligns with many of the priorities of sustainable chemistry. However, this class of materials alone will not solve the problem of polymer sustainability.

Despite the existing examples which are emphasized in recent themed issues of diverse Journals on the topic of sustainability,[35,36,37] it is still problematic to define a certain polymer as a sustainable polymer, due to the absence of a universally accepted definition of sustainability with respect to materials. For example, certifications for compostability and biobased content aid to identify which products are potentially more environmentally friendly. However, without the availability of compost and recycling facilities and adequate collection streams, these certifications do not contribute to sustainability of these materials. For example, the recycling rate of so-called biopolymers in Australia is 0%[38] due to a lack of collection and recycling possibilities for biopolymers. However, it is obvious that sustainable polymer chemistry is more than green polymer chemistry, and not an alternative definition, as sometimes reported in the literature. Indeed, sustainable polymer chemistry is a comprehensive approach that includes green chemistry concepts, and inherently safer design as well, but in a general framework balanced on the pillars of economic growth and development along environment preservation and social promotion. Eventually, the successful transformation of polymer synthesis toward a more sustainable future requires not only scientific research and technological innovation, but also potentially a clear “definition”.[39]

4. Circular Economy and Circular Chemistry

"Waste," the result of the linear "take-make-use-dispose" economy, commonly refers to man-made materials that are "thrown away" – either collected as "trash" or landfilled or discarded in the environment – as materials without economic value or even with negative economic cost (e.g., landfill fees). Nevertheless, since matter can be neither created nor destroyed (Law of Conservation of Matter), the option to "throw away" is not feasible in the long term within a system. Accordingly, anthropogenic waste in all its forms is substantially contributing to a range of planetary-scale environmental crises that are evolving with escalating speed. In 2009, Rockström et al. introduced the Planetary Boundaries framework defining nine interlinked planetary boundaries, i.e., climate change, ocean acidification, stratospheric ozone depletion, biogeochemical flow, global freshwater use, change in land use, biodiversity loss, atmospheric aerosol loading, and chemical pollution/novel entities.[40,41,42]

Critically, until recently the planetary boundary for chemical pollution/novel entities was not defined and assessed. However, Persson et al. very recently reviewed the literature relevant to the planetary boundary for novel entities (NE-PB) and concluded that “the safe operating space of the NE-PB is exceed when annual production and release (of chemicals and products) increase at a pace that outstrips the global capacity for assessment and monitoring” and therefore NE-PB is in the red zone of high risk and serious impact for the Earth System.[43] Persson et al. also state that plastic pollution is a particular aspect of high concern.

Inevitably, the increasing global concern for anthropogenic driven climate change and other planetary boundaries is a major driver in the transition from a traditional linear flow of materials in a—“take-make-use-dispose”—linear economy to a circular economy. The circular economy concept and its metrics[44] have been introduced by Ellen MacArthur Foundation’s Circular Economy 100 Program, where the Material Circular Indicator has been used as a central metric. The main aim of circular economy is to reuse all compounds in a process and manufacturing lifecycle, whereas concurrently increasing their usage and reducing their waste. In fact, with the aim of “closing loops” by greater reuse and recycling of materials, the European Commission has identified the EU Circular Economy Action Plan (EU CEAP) as a top priority.[45]

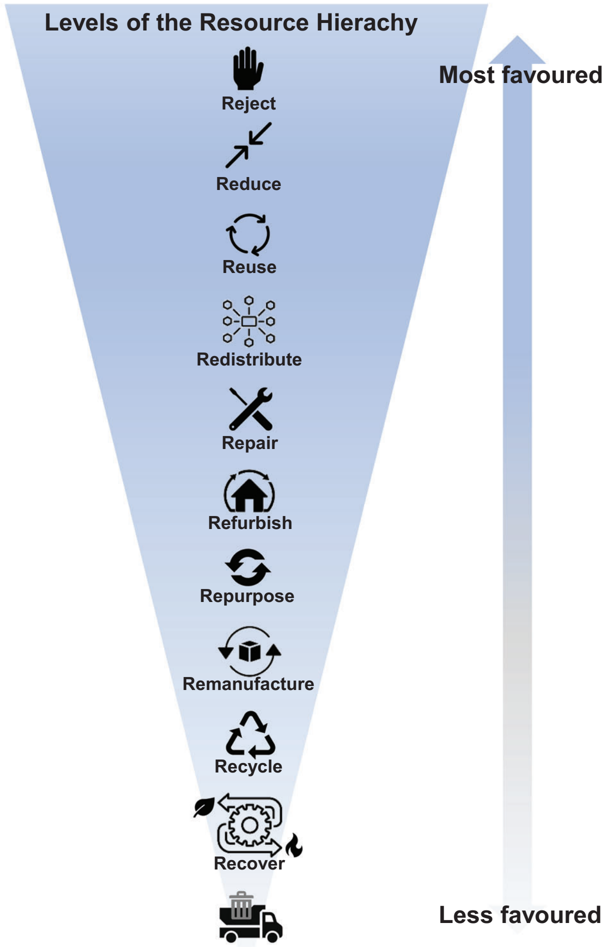

Figure 1. Levels of the resource hierarchy from refuse to recover.

However, Kirchherr and colleagues state that at least 100 possible definitions and many different interpretations of the concept of circular economy exist,[46] also indicated by the rapid growth of peer-reviewed articles on the topic.[47,48] The “3R” principles of circular economy—reduce, reuse, and recycle and depicted by the Mobius symbol—first emerged in the 1970s, with a strong emphasis on “reduce,” which subsequently paved the way toward the concept of green chemistry from the early 1990s. Inevitably, in the last decade, circular economy has gained momentum based on the eleven hierarchy levels depicted in **Figure 1** (i.e., a resource hierarchy showing various strategies for waste avoidance), and has encouraged a change of mindset from waste management to resource management, including the prioritization of the sustainability of resources and emphasizing waste prevention option. In other words, it prioritizes the focus on resources and incorporates waste as valuable starting material. Regarding polymers and plastics, the objective of the waste hierarchy is to reduce the ecological and carbon footprint of plastics by executing the highest-level strategy on a hierarchy of decreasingly favorable choices. The top strategy of the waste hierarchy is refusing the product and packaging (most desired); followed by the reduction of the amount of plastic used in the product and packaging, along reuse of the product and package. Whereas recycling and recovering and the disposal of plastic in a managed landfill or in a mismanaged way are the least desired strategies. In a similar manner to green chemistry and sustainability, replacing fossil-derived plastics with plastics based on renewable resources (totally or partly) (also referred to as "biopolymer") and the use of renewable energy in the production of fossil-derived plastics present some alternative options to reduce the carbon footprint of plastics in regard of circular economy of polymers. However, green chemistry prioritizes using renewable feedstocks based on biomass without explicitly considering the potential environmental, economic, and societal issues associated with increased production and transport of the biomass needed to supply those feedstocks. Therefore, it is important to realize the extreme potential of end-of-life plastic as a resource for new products. In addition, strategies for plastics that are removed as pollution from the environment (e.g., marine plastics) need to re-enter the circular economy of polymers and concepts for the avoidance of plastic pollution need to be developed and implemented.

It is also obvious that there can be no single or simple approach to circular economy as the challenges of minimizing waste and achieving material circularity in a sustainable fashion can be different for each material and each process considered. These challenges lead to the question of how chemistry can address and support the circular economy. In 2019, Keijer et al. introduce the twelve principles of circular chemistry as shown below, which combine concepts from chemistry with circular economy and sustainability.[49]

4.1. Twelve Principles of Circular Chemistry

The twelve principles of circular chemistry as introduced by Keijer et al.[49] are:

1) **Collect and use waste**. Waste is a valuable resource that should be transformed into marketable products.
2) **Maximize atom circulation**. Circular processes should aim to maximize the utility of all atoms in existing molecules.
3) **Optimize resource efficiency**. Resource conservation should be targeted, promoting reuse and preserving finite feedstocks.
4) **Strive for energy persistence**. Energy efficiency should be maximized.
5) **Enhance process efficiency**. Innovations should continuously improve in- and post-process reuse and recycling, preferably on-site.
6) **No out-of-plant toxicity**. Chemical processes should not release any toxic compounds into the environment.
7) **Target optimal design**. Design should be based on the highest end-of-life options, accounting for separation, purification, and degradation.
8) **Assess sustainability**. Environmental assessments (typified by life cycle assessment (LCA)) should become prevalent to identify inefficiencies in chemical processes.
9) **Apply ladder of circularity**. The end-of-life options for a product should strive for the highest possibilities on the ladder of circularity.
10) **Sell service, not product**. Producers should employ service-based business models such as chemical leasing, promoting efficiency over production rate.
11) **Reject lock-in**. Business and regulatory environment should be flexible to allow the implementation of innovations.
12) **Unify industry and provide coherent policy framework**. The industry and policy should be unified to create an optimal environment to enable circularity in chemical processes.

Interestingly, these twelve principles of circular chemistry are covering aspects of chemistry, but also aspects of the economy, policy, and environmental science highlighting the importance of the interconnectivity between these areas and the need for trans- and multidisciplinary approaches, research, and practice.

The twelve principles of circular chemistry clearly imply that (polymer) chemists equipped with the tools of green chemistry will play a central role in the development of circular (polymer) chemistry. Accordingly, polymer scientists will need to focus on improving the overall circularity (and sustainability) of the developed polymeric materials. Critically, it is important to consider and implement circularity at the design state of polymeric and plastic materials development. Indeed, addressing only waste management and recycling issues of plastics and polymers is a clear indication that the concept of circular polymer chemistry is still in its early stage, hence resulting in a large knowledge gap. We submit that significant advances in circular chemistry will rely on the further development of principles of circular economy and sustainable design as well as realizing the enormous potential of polymeric and plastic waste as a resource for new products, i.e., keeping a fossil resource in circulation as long as possible. In addition, commodity polymers are not used in their pure form, mostly they are mixtures—called plastics—of polymer(s) with a variety of additives depending on the desired properties of the product and the requirements for their production. In order to achieve a circular economy of plastics, the focus of research and development needs to be on the polymers as well as the additives, i.e., plasticizers, flame retardants, pigments, antioxidants, stabilizers, antistatic, and nucleating agents. The reader is referred to a publication from Aurisano et al. summarizing the challenges and knowledge gaps that need to be overcome to enable a circular economy of plastics.[50]

5. Conclusion

In summary, it is important to distinguish between green, sustainable, or circular chemistry, particularly within the field of polymer chemistry. Green chemistry and circular chemistry are both important concepts; however, they serve different economies, i.e., linear and circular economy. In similar manner, polymers and plastics are important materials with a forecast production increase that goes hand in hand with more and more environmental and social deterioration caused by pollution and waste. Green and sustainable chemistry can address these challenges to some extent. However, ultimately a shift from green polymer chemistry to circular polymer chemistry with a strong emphasize on how to maximize sustainability is urgently needed.

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The authors declare no conflict of interest.

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Hatice Mutlu studied chemistry at Marmara University and Bogaziçi University (Turkey). Subsequently, she obtained her Ph.D. from the Karlsruhe Institute of Technology (KIT) in the group of M. A. R. Meier. After post-doctoral stays in the groups of J.-F. Lutz (ICS-CNRS, Strasbourg) and C. Barner-Kowollik (KIT), respectively, she is currently working as a senior researcher and acting as the Deputy Director of the Institute of Biological Interfaces 3 (IBG3) at KIT. She was selected as one of the 2020 Emerging Investigators in Polymer Chemistry by *Polymer Chemistry*, RSC. She is also a recent member of the International Advisory Board of *Macromolecular Chemistryand Physics*. Her research interest spans the synthesis of complex macromolecular architectures and functional polymers to the sustainable development of new polymer-forming reactions and novel conjugation chemistries with a particular focus on design of novel sustainable sulfur-based in addition to self-reporting polymeric materials.

Leonie Barner is the inaugural director of the Centre for a Waste-Free World at the Queensland University of Technology (QUT, Brisbane, Australia). She received her Ph.D. in physical chemistry in 1998 (Georg-August-Universität, Göttingen, Germany). During her career, she worked in industry, German research institutes (Fraunhofer, Helmholtz), and Australian universities (University of New South Wales, QUT). Besides her research interest in macromolecular synthesis and characterization, she leads a transdisciplinary research center that follows a holistic approach to develop technologies and processes to reduce waste, while using social science knowledge to catalyze change and reduce barriers to participation and adoption.

