

News & views

Sustainable chemistry

Closing the loop on recycling bioplastics

Charlotte K. Williams & Georgina L. Gregory

Plastics are invaluable materials, but they use up petroleum resources and persist in the environment. A high-performance plastic derived from renewable oils has been designed at the molecular level to be truly recyclable. See p.423

Stark images of plastic mounting up in landfill sites and oceans has prompted a reconsideration of its use. Not only does plastic production consume depleting crude-oil resources, but most plastics are not effectively recycled and are environmental pollutants. There are many types of plastic, but all contain polymers. Solving the plastics problem will require many different approaches, but, fundamentally, the polymer chemistry of plastics must be redesigned to improve their sustainability¹. Key targets are to diversify the raw materials used to make plastics beyond fossil fuels; to conserve the embedded energy and valuable resources in their structures; to fully maintain their useful properties through multiple recycling loops; and to design plastics whose molecular structure can be completely disassembled when necessary^{2–4}. On page 423, Häußler *et al.*⁵ report a plastic that has the potential to meet all of these criteria.

High-density polyethylene (HDPE) is a widely used plastic, featuring long, straight chains of polyethylene – a polymer consisting of repeated CH_2CH_2 units. When crystallized, HDPE has excellent properties for diverse applications, including as electrical insulators, pipes and detergent bottles. HDPE is regularly recycled mechanically by melting and reprocessing⁶ (Fig. 1a). Nonetheless, current plastic-waste management is ineffective. The US Environmental Protection Agency found that, in 2018, less than 10% of all plastics, and only about 30% of HDPE bottles, were being recovered from mixed-plastic waste streams and recycled (see go.nature.com/3jw8meq). Mechanical recycling can also yield inferior materials after each round of recycling and, in the case of HDPE, it can be challenging to control the crystallinity of the recycled products^{7,8}.

An alternative to mechanical recycling is

chemical recycling (also known as closed-loop recycling), in which long-chain polymers are deconstructed after use to produce the same molecular building blocks (monomers) that were originally used to make them. The advantage of this approach is that these monomers can be repeatedly re-polymerized to produce materials that have the same high performance and properties as the original. Unfortunately, such a strategy is ineffective for HDPE because a lot of energy is required to break its carbon-carbon bonds. The strength of these bonds

also explains the environmental persistence of polyethylene and its recalcitrance to enzyme degradation.

Häußler *et al.* now report plastics that have many of the key properties of HDPE, but that are also designed for complete closed-loop recycling. The authors developed high-yielding chemistry (greater than 95% reaction yields) for transforming oils derived from plants or microalgae into polymers with high molecular weights. The polymer chains contain a small fraction of regularly placed carbonate or ester linkages (Fig. 1b). Well-established ‘solvolysis’ reactions with water or common alcohols can then be used to completely break up all of the polymer chains, enabling almost full recovery (96%) of the monomers and closed-loop recycling. The authors report that isolation of the monomers from the solvolysis reactions is straightforward, and that the monomers can be successfully re-polymerized to produce materials that retain the properties of the original plastic.

The key advance of this work is that it simultaneously solves many long-standing and difficult challenges that have dogged the field of sustainable polymers. Researchers from the same group as Häußler *et al.* have pioneered, over several decades, the chemistry now used to transform natural oils into useful monomers⁹.

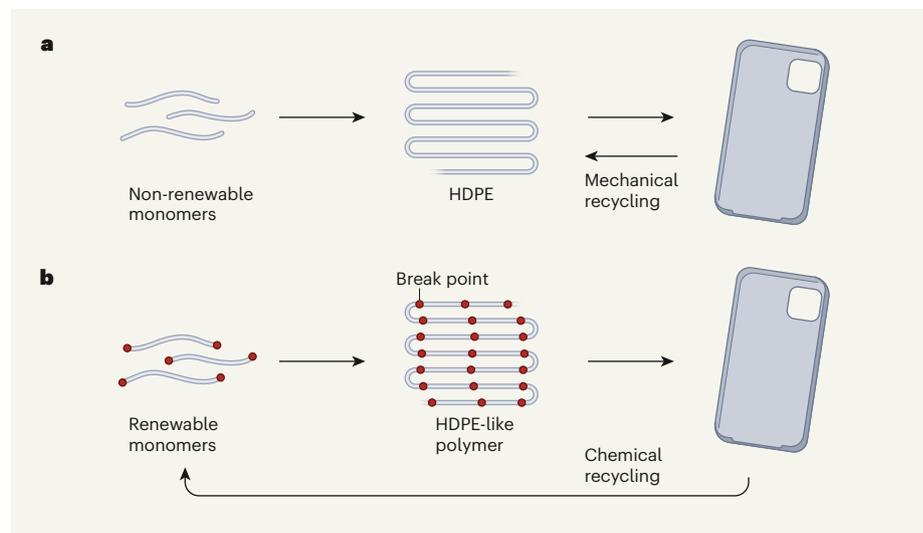


Figure 1 | Redesigning high-density polyethylene (HDPE). **a**, Commercial HDPE is a widely used plastic that consists of a chemically inert polymer, formed from non-renewable petroleum-based monomers. Although it can be recycled mechanically (by melting and reprocessing), this can lead to a reduction in the material’s performance. **b**, Häußler *et al.*⁵ report a plastic that has comparable properties to HDPE but that is formed from renewable plant- or microalga-derived monomers. The monomers have chemical groups at each end that form ‘break points’ in the resulting polymer chains. These break points allow the polymer to be chemically recycled back to its monomers, which can then be used to remake the plastic. The authors make various objects from the plastic, such as a mobile-phone case, and show that the properties of the plastic do not degrade after each recycling loop.

The authors use highly efficient catalysis (reaction yields 80–90%) to selectively install chemical groups at the ends of the monomers; these groups form the basis of the desired ‘break points’ in the polymers. The monomers are then polymerized using well-established methods. The authors find that using a particular co-monomer (diethyl carbonate) in the polymerization reaction enables the formation of high-molecular-weight polymers. This is essential for making a plastic that matches the thermal, mechanical and processing properties of HDPE.

Häußler and colleagues demonstrate that the new plastic can be processed using common industrial techniques such as injection moulding and 3D printing, and to include colourants or carbon fibres (which are widely used as additives to strengthen polymers). They also show that solvolysis of the new plastic occurs selectively when it is mixed with conventional plastics such as commercial polyethylene terephthalate (PET), which is widely used in drinks bottles and is also a candidate for chemical recycling by solvolysis. This proof-of-concept result hints that selective recycling of the new plastic might be possible in the future.

Although the findings hold great promise, it is important to recognize that this is still early-stage, fundamental research. Polymer-to-polymer recycling has been demonstrated for only 20 grams of the new material, and much work will be required to translate this into industrial-scale processes and products. Major engineering challenges remain to be solved in other parts of the polymer’s life cycle, including finding ways to produce the biomass-derived monomers at large scales and the development of industrial processes for making, forming and recycling the plastic.

Furthermore, economic considerations overshadow these endeavours. Plastics used in industry, such as HDPE, are produced on the multimillion-tonne scale, and usually sell at US\$1–3 per kilogram. It would be unreasonable to expect a new plastic to be cost-competitive immediately, but such price issues make the introduction of new plastics highly challenging.

Questions also need to be answered about how well the new plastic integrates with existing waste-management systems – if it is to replace HDPE, it must be shown to be compatible with all the methods used for separating waste plastics across multiple facilities and geographies. Unlike most current recycling strategies, the type of chemical recycling reported by Häußler *et al.* requires a chemical plant. Encouragingly, however, the reported chemistries seem well suited for use with industrial methods. Moreover, the reported system seems consistent with European legislation that requires manufacturers to take responsibility for the plastics in their

products after consumer use.

Häußler and colleagues’ work is exciting and inspiring, because it is extremely challenging to come up with plastics that can be derived from renewable resources, have outstanding properties, are compatible with large-scale manufacturing and processing techniques, and are fully recyclable – few materials meet all these criteria. The authors’ work is an excellent example of how scientific innovation can solve all the facets of a problem, rather than just individual components. The next step must be to build on the life-cycle assessments presented in the current work, to provide even greater improvements in sustainability. More broadly, society must also demand that manufacturers provide equivalent life-cycle assessments and evaluations of all the environmental impacts of currently used plastics, so that the priorities for replacements become clear.

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Evolution

The role and rule of relatedness

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Inclusive fitness theory shows that social partners must be related for altruism to evolve, yet some models suggest that relatedness is not needed. An analysis concludes that assumptions in those models build in a role for relatedness, after all.

Scientists aim to work on important, open problems: their papers and grant proposals tell us so, and, of course, such a focus is entirely proper. But if a field forever declares a problem to be open, it can give the impression that progress is never made. The study of social evolution runs this risk, because many researchers repeatedly assert that how altruism evolved remains an unsolved puzzle. In fact, in the early 1960s, the evolutionary biologist W. D. Hamilton came up with a solution to this ‘problem of altruism’ with his inclusive fitness theory^{1,2}, which shows that it is possible for altruism to evolve if socially interacting individuals are related. Writing in *Proceedings of the National Academy of Sciences*, Kay *et al.*³ conclude that multiple attempts to find alternatives to Hamilton’s solution have simply rediscovered it.

Biological altruism is defined as any social behaviour between individuals in which an action by an altruistic individual decreases its lifetime direct ‘fitness’ (that is, reduces the number of offspring it has) but increases

the lifetime direct fitness of the recipient. By contrast, another type of social behaviour, termed reciprocal altruism, does not qualify as altruism in this sense. Reciprocal altruism occurs when social benefits are exchanged between interacting individuals⁴, and such cooperation evolves only when both partners experience a net gain in lifetime direct fitness⁵.

A prime example of biological altruism is the case of eusocial insects (those that have a worker caste), such as bees (Fig. 1), and in some of these species the workers have lost their ability to reproduce entirely. Such cases encapsulate the problem of altruism. By what means can natural selection, a process based on out-reproducing competitors, lead to self-sacrificial sterility?

According to Hamilton^{1,2}, the answer lies in interacting individuals being positively related, meaning that they are genetically more alike than any individuals associating together randomly within a population. If altruism is directed towards relatives (kin),