waste products. One recent example is how some US plastics producers converted from crude oil or naphtha-based feedstocks to ethane, a by-product of natural gas production through hydraulic fracturing.

The transition to renewable energy opens the question of which substrates will be used for future plastics. Understanding plastics' early industrial history is important because these bio-based products established the political-economic relations of modern, conventional plastics and portended problems to come. This history also points to the insufficiency of an ahistorical technological fix, such as swapping in alternative carbon sources, which may not improve plastics' ethics, safety, or sustainability. This is especially true if the same problematic chemistry is used to modify the base plastics' performance characteristics (*13*). For example, even if viscose/rayon is sourced from Forest Stewardship Council (FSC)–certified forests, its production may still rely on carbon disulfide.

To avoid such problems, it is necessary to rethink the premises on which plastics technologies have been developed and produced. Critical adjuncts include reengineering plastics for recovery and reuse, augmenting recycling infrastructure (*14*), and source reduction and dematerialization. This means making fewer plastics by developing alternatives to their short-term, disposable uses, which presumes land access for landfills (i.e., long-term storage of solid waste or ash) (*15*). The challenge for bio-based plastics research is to account for this history and to think critically about the supply chains required by plastics currently in development, including a focus on ethical, sustainable feedstocks; toxics reduction and safer materials; and worker and community health and safety. ■

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10.1126/science.abj1003

PERSPECTIVE

Achieving a circular bioeconomy for plastics

Designing plastics for assembly and disassembly is essential to closing the resource loop

By **Sarah Kakadellis**[1] *and* **Gloria Rosetto**[2]

The visual nature of plastic pollution and the scandals of plastic waste exports to developing countries have prompted a shift in how plastics are made, used, and disposed. Plastic waste remains poorly managed, with as much as 12,000 million tonnes projected to have accumulated in landfills or the natural environment by 2050 (*1*). Although mechanical recycling was initially promoted as the solution to rising amounts of postconsumer plastic waste, its failure over the past decades has exposed the severity and scale of the plastic waste management crisis. In light of this, the recovery of plastics through chemical recycling—polymer recycling into their constituting repeat units or monomers (and oligomers)—and the development of bio-based and biodegradable alternatives have gained increasing attention. We consider the technical, chemical, and biological routes to closing the loop and argue for an integrated plastic waste management system rooted in the circular bioeconomy.

Shunning fossil-based plastics has provided a fertile ground for the emergence of alternative materials, loosely referred to as "bioplastics." Despite favorable public opinion, consumer awareness and understanding of the subtleties in the terminology is poor (*2*). The term bioplastics is an umbrella designation that captures a range of polymer chemistries, properties, and application sectors. It encompasses two distinct concepts: the bio-based origin of the raw materials and biodegradability at the end of life. Bio-based sources are necessary for divesting from fossil fuels. However, life-cycle analyses have uncovered complexities in the system, mostly owing to agricultural inputs for bioplastic feedstock production (*3*). Recent approaches using waste or coproducts from the biomass sector as feedstocks offer attractive alternatives.

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Some (fully or partly) bio-based plastics, such as bio-polyethylene terephthalate (bio-PET), are chemically identical to their fossil-based counterpart, making them suitable for the current recycling infrastructure. However, biodegradability tends to be perceived as more sustainable over (mechanical) recyclability by consumers (*2*). The biggest advantage of biodegradable plastics may not be their biodegradability per se but their compatibility with food waste, opening new streams for plastic waste management positioned around organics recycling (*3*). Nevertheless, issues associated with separation and contamination in existing mechanical recycling streams and concerns over their complete biodegradability in the current organic waste management infrastructure remain (*4*).

Although biodegradable plastics can return carbon and nutrients to the soil, the energy and resources associated with their production is effectively lost, echoing the linear flow of petrochemical plastics in single-use applications. Maintaining a closed-loop resource flow appears more sustainable. Yet, 67% of plastic waste generated in the UK consists of hard-to-recycle packaging (*6*). Across Europe, only 42% of plastic waste generated is collected for recycling (*5*, *6*). Failing market incentives for plastic recyclate have led to many plastics being exported to Southeast Asia, where they are often disposed of in illegal landfills (*7*).

Thermochemical processes, such as pyrolysis and gasification, have emerged as an alternative recycling strategy for the recovery of plastic waste—notably, hard-to-recycle plastics (*6*). Although they are often referred to as chemical recycling, these processes are not selective for monomer retrieval, producing a wide range of hydrocarbons and carbon dioxide (CO_2). Further separation and transformation steps are required that are energy intensive. By contrast, closed-loop recycling to monomers (CRM) can be seen as ultimate chemical recycling in that it ensures the recovery of a given polymer's building blocks.

The feasibility of CRM is greatly dependent on polymerization-depolymerization thermodynamics (*8*). The most prevalent feature of such polymers is a hydrolyzable functionality in the polymer backbone, such as ester, amide, and carbonate linkages. PET, the most widely mechanically recycled commodity plastic, falls under this category. Polyolefins, such as polyethylene (PE) and polypropylene (PP), pose a challenge for CRM because of their carbon-to-carbon backbone. The introduction of functional groups as break points in a PE chain presents an opportunity to address polyolefin-like polymers with potential for CRM while retaining the desired material properties, as has been demonstrated (*9*). Although challenges remain in this strategy, these technological advances could ensure that monomers are effectively recovered, preventing the issue of downgrading or downcycling, seen with mechanical recycling.

Nevertheless, a sustainable plastics value chain extends beyond monomer recovery. The accumulation of plastic waste points toward a design flaw in the plastics value chain and the need to think systemically about closing the loop of the circular economy. If resources are cheap, the impetus to produce single-use products from virgin materials is high. Suspending trade of low-quality plastic waste from developed to developing countries and introducing taxes on fossil resources can encourage the substitution of raw resources with recycled materials and investment in waste management infrastructure (*7*, *10*).

Although these measures may increase the value of recycled polymers, the quality of recycled materials will remain a substantial challenge, especially for plastic packaging. In the context of a circular economy, the value of durable plastics needs to be recognized, but in conjunction with modularity in polymer and product design. Yet, there seems to be a lack of directionality around plastics-focused policies. If left uncoordinated, the promotion of biodegradable plastics within a bioeconomy framework on one side and of closed-loop recycling from a circular economy perspective on the other may lead to conflicting priorities.

The distinction between biodegradable and recyclable plastics suggests that biological and chemical routes to plastic waste management cannot be merged, perhaps misleadingly so. Most biodegradable plastics are or could be chemically recyclable because they can be fully metabolized by naturally occurring microorganisms. Developing a system in which plastics are designed for both chemical recycling and biodegradation is not only sensible but helps to overcome the artificial dichotomy emerging from current policies (see the figure). Thus, a waste management infrastructure for plastics to be collected and recycled should be prioritized while also fulfilling an end of life in applications for which biodegradability is needed.

Chemical polymer manufacturing and recycling is already technically feasible, cost effective, and scalable. But, development of the chemistry to design out recalcitrant petrochemicals and improve recycling efficiency is still needed. These challenges should be supported with a combined push in both chemistry and biotechnology. More recently, enzymatic hydrolysis of polymers has emerged as a potential bioremediation strategy (*11*). Enzymatic recycling has been demonstrated for PET (*12*, *13*), with the need for other enzymes for metabolizing a greater range of polymers. Obtaining monomers from CO_2 fixation would ultimately decouple production from raw materials (*13*).

The consideration of alternative waste treatment strategies for plastic waste is undoubtedly only part of the bigger issue of a linear economic model. The fallacy of mechanical recycling has already taught us that technology alone will not and cannot solve the plastic pollution crisis. No silver-bullet solution exists for the multifaceted nature of plastic pollution. The answer instead lies in a blend of approaches. Pre- and postconsumer stages need to be more aligned, from a strong regulatory framework and the investment in effective waste collection and management infrastructure to the development of polymer chemistries, life-cycle design, and consumer behavior. Only through committed action and coordination across the value chain will a sustainable future for plastics be secured. ■

Closing the plastics loop

Plastics (polymers) could be designed so that the monomers they are built from can be retrieved. Some plastics can also biodegrade for certain niche applications or when environmental leakage occurs. Monomers may be feedstocks for synthesizing added-value products such as surfactants or new polymers or turned back to their original polymer. Enzymatic or chemical catalysis can prevent property deterioration from this process.

1 Design polymers for monomer recovery

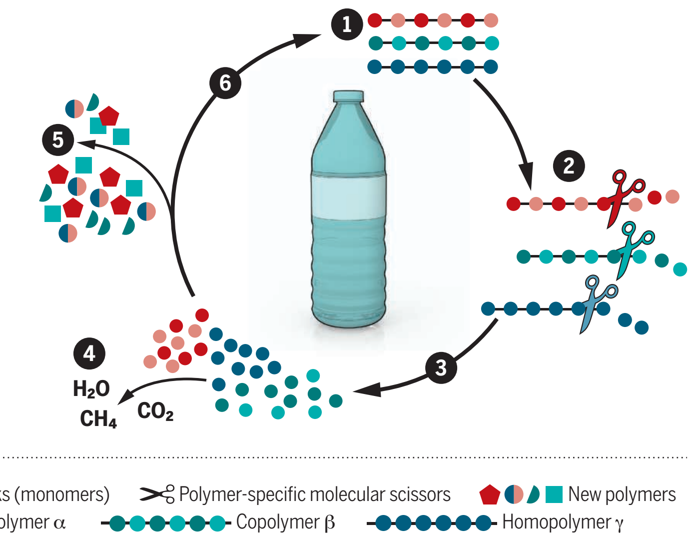
2 Site-specific bond cleavage

3 Further disassembly into monomers

4 Biodegradation in the biosphere

5 Synthesis of added-value products in the technosphere

6 Synthesis of polymers from recovered monomers



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ACKNOWLEDGMENTS

The authors acknowledge C. K. Williams, Z. M. Harris, and J. I. Jimenez for providing invaluable insight throughout the revision process. S.K. was supported by a London Interdisciplinary Social Science Doctoral Training Partnership studentship awarded by the Economic and Social Research Council (ES/P000703/1). G.R. was supported by SCG Chemicals.

10.1126/science.abj3476

GRAPHIC: N. CARY/*SCIENCE*

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Science, 373 (6550), • DOI: 10.1126/science.abj3476

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