Toward cruising speed for circular plastics

Editorial Overview

Toward cruising speed for circular plastics

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Making plastics circular is a daunting challenge, though not one to avoid. Plastics circularity is quintessential as one of the building blocks towards net-zero scenarios. Carbon-based chemistry and materials will remain a cornerstone of our technological systems, yet we can no longer afford to burn them at the end of their life, giving unnecessary CO_2 emissions, nor should we let their embodied energy, which is at least partially fossil-based, go to waste. In response to increasing pressure to establish a circular economy for plastics, the science and innovation landscape has been brewing intensely in the last decades. We are, if our judgement is correct, live witnessing an enormous transition in organic materials management.

Although the amount of plastics used globally is still on the rise, while their overall carbon footprint seems to have stabilized (Cabernard et al. 2022), there were recently interesting signs that the demand for recycled plastics is soaring (Brooks 2021) while the supply cannot keep up in the short term. This confirms that many companies are both addressing the consumer calls for circular packaging, and anticipating future circular plastics policies. In response, there are numerous initiatives for new plastics recycling plants, focusing both on polyolefins and other polyaddition polymers, but also on step growth polymers, often polycondensates. The initiatives are not only restricted to state-of-the-art mechanical recycling facilities, but (thermo)chemical recycling clearly comes to the fore in recent and announced investments (Li et al. 2022). For example, given the investments by both new industrial players and the traditional (petro)chemical industry, it is reasonable to expect a multimillion tonne range capacity of mechanical and chemical recycling plants in the next few years.

In recent years, it has become clear that most recycling schemes, although often competitors, deserve their unique place in the so-called circular economy for plastics. From a policy or investor's point of view, the tradeoff should always be made between various cycles, whose sizes represent energy intensity, and the purity and quality of both the feedstock and the targeted product. For example, highly contaminated and potentially degraded mixed polyolefin films are less suited for the more conventional mechanical recycling paths, given the poor recyclate qualities obtained. Without inducing more 'advanced' recycling approaches, such as delamination and deinking, pyrolysis, etc., such waste streams would have limited potential to substitute virgin materials.

The Special Issue "Circular Technologies for Plastics", with its seven contributions from international teams of experts, shows that the recent state-of-the-art – as it should – outpaces the industrial innovations, and seems to even surpass the technological requirements for recycling or degradation of relatively abundant and well-defined plastic streams such as plastic bottles. In fact, current research efforts focus on more recalcitrant plastic fractions, such as polyesters (e.g. polyethylene terephthalate – PET) in films and trays, textiles, mixed polyolefins, polystyrenes with complex formulations, and polyurethanes. Newly explored routes span all cycles of the circular economy for plastics; advanced mechanical recycling by better prediction and understanding of extrusion technology in the case of compatibilization, to pyrolysis and solvolysis for polyaddition and step-growth polymers, respectively, to biochemical recycling, with final biological mineralization as a last resort.

Edeleva et al. (https://www.sciencedirect.com/science/article/pii/S2452223623000962) point out that molecular scale modeling of extrusion and reactive extrusion technologies are essential to expedite innovations in mechanical recycling of complex feedstock. Interestingly, there is limited knowledge available on the recycling of many bioplastics, while for most of them mechanical recycling

would be energetically favorable. The modeling tools explored, e.g., coupled matrix-based kinetic Monte Carlo (CMMC) simulations, allow to predict 3D-structural variations (molecular mass distribution, branching, a.o.) as a function of processing conditions (temperature, residence time), and hence strongly reduce the experimental requirements to validation, rather than exploration. The same holds for complex blends of bioplastics (e.g. polylactic acid – PLA, polyhydroxyalkanoates – PHA, thermoplastic starch – TPS) with traditional plastics (e.g. polyethylene – PE), which require smart compatibilization.

A similar observation is put forward, even more strongly, by Kovacs et al. (https://www.sciencedirect.com/science/article/pii/S2452223623000500) regarding chemical recycling technologies, and more specifically the formulation of products from the obtained resins. They take the case of polyurethanes (PU), and posit that recycled resins, e.g. polyols, are not expected to have the exact composition and properties of their virgin counterparts, and this limits their uptake in product formulations. To accelerate the transition to sustainable formulations, they argue, the implementation of machine learning methods combined with quantum chemical or molecular dynamics simulations, and reaction kinetics models (such as the aforementioned CMMC) will be required. This will enlarge the demand for recycled resins.

Depolymerization strategies for polyurethanes themselves are elaborated by Fonseca et al. (https://www.sciencedirect.com/science/article/pii/S2452223623000512), highlighting both chemical and biochemical pathways and the associated carbon footprint benefits. As is commonly known, most PU is thermoset, especially in the most common applications; flexible and rigid foams. This results in little possibilities for high-value mechanical recycling, making advanced (bio)chemical recycling essential for PU circularity. Although the chemical recycling of PU is gaining momentum, also industrially, the search for more benign conditions by means of biochemical processes is ongoing. Targeted enzymatic depolymerizations are explored with varying success; a true "*urethanase*" enzyme has yet to be discovered/developed. At present, the most promising results were obtained using mixed microorganism cultures. Whatever the exact catalytic mechanism exploited, the quality of the obtained products, cf. supra, plays a pivotal role though in assessing the environmental savings, which is by extension valid for all plastics recycling.

One example where high value products can potentially be combined with relatively benign processing conditions, is biochemical recycling of polyethylene terephthalate (PET), as outlined by Chen et al. (https://www.sciencedirect.com/science/article/pii/S2452223623000718). Not only are we getting much better insights into the relationships between polymer structure and enzyme degradation potency, protein engineering allows to increase the thermostability of PET degrading enzymes, and increase the degradation kinetics. They further argue that the strategies employed for PET are applicable for the biochemical recycling of other polymers. Here, we would like to raise the point of attention though, that the energy and costs associated with depolymerizations, should not be shifted covertly from high processing temperatures and solvent recovery in chemical depolymerization, towards enzyme production and isolation in biochemical recycling. The latter will need to be continuously evaluated.

The complementarity between various polymer cycles, exemplified by physical recycling, mechanical recycling and (thermo)chemical recycling, is well demonstrated by Goshayeshi et al. (https://www.sciencedirect.com/science/article/pii/S2452223623000706) for polystyrene (PS). Given its diverse applications (foams, high-impact materials, sheets, packaging) and sensitivity towards thermo-mechanical stress, conventional methods alone are inadequate to achieve PS circularity. Given the low bulk density of PS waste, defoaming seems an essential step in future recycling operations. Furthermore, rubber (from synthetic SBR blends) removal should lead to higher purity of PS recyclates,

both in mechanical and thermochemical recycling. Although the latter is in industrial demonstration (pilot) stage, several technological advances are yet to be implemented or optimized.

The short review of Kalita and Hakkarainen (https://www.sciencedirect.com/science/article/pii/S2452223622001638) even expands the view on complementary polymer cycles, by studying how biodegradation can or should be integrated into a circular economy for plastics. They clearly state that more attention should be paid to the recycling of plastics to preserve the molecular structures as intact as possible, but nonetheless biodegradability could provide an important asset for this class of materials. To this end, a solid understanding and control of biodegradation conditions is essential, as is the environmental fate of various (trace) chemicals embodied in or resulting from the polymers. Composting, anaerobic digestion, or even full biochemical mineralization are logical options for plastic materials that are highly mixed or contaminated with organics and for which sorting, separating and refining would result in excessive energy consumption.

Finally, as also demonstrated in this short editorial overview, many different terms have entered the domain of circular plastics technologies, for which no strict definitions exist. Examples of such terms are open- and closed-loop recycling, mechanical and chemical recycling, upcycling, advanced recycling, recovery, and others. As a result, some confusions may arise about the actual technological cycles meant, or, for example, the legal implications. Ragaert et al. (https://www.sciencedirect.com/science/article/pii/S2452223623001190) attempt to clarify and streamline the terminology in this special issue, at least in a European perspective. They bring several terms to the fore, along with a call to avoid terms with broad or misleading meanings.

From this article collection, it is clear that the research domain of circular technologies for plastics is reaching cruising speed, with many of the early scientific advances already getting to very high technology readiness levels. As an example, more complex waste streams are currently being mechanically recycled, including (mixed) polyolefins, due in part to a better understanding and control of extrusion technology, amongst others. Furthermore, industrial demonstration plants are being built for the pyrolysis of polyolefins and polystyrene, although several technological issues still need to be addressed. Ongoing research is focusing on more recalcitrant and mixed plastics, as well as the search for more benign recycling conditions, such as biochemical processes. In combination with the recent work of Larrain et al. (2022), we cannot help but notice that technological advances – luckily – outpace policy measures that could greatly accelerate the transition to a circular economy for plastics.

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BIOSKETCHES

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