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Reimagining plastic waste: sustainable depolymerization using mechanochemistry as a gateway to high-value applications

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Plastic materials have become essential components of modern life, found in everyday products and items.[1] They play a significant role in industrial sectors such as automotive and aerospace due to their remarkable properties.[1] However, plastics exhibit significant resistance and durability due to their polymeric structure, which consists of long chains formed by the repeated linkage of small molecular units known as monomers.[2] This remarkable resilience contributes to the plastic waste problem, resulting in the accumulation of discarded plastic materials and objects at the end of their lifecycle. It was reported that 413.8 million metric tons of plastic were produced in 2023, compared to 15 million metric tons in 1976.[3]

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The accumulation of plastic waste is evident worldwide, affecting terrestrial and aquatic ecosystems from the poles to the deep ocean and coastlines.[4] Thus, plastics are becoming a cause of concern for countries worldwide.[5] In the environment, larger plastic items break down and gradually form smaller pieces known as microplastics (MPs), which can accumulate and persist in ecosystems.[6] MPs might be consumed by different organisms, from plankton and fish to birds and even mammals, accumulating throughout the marine food web.[7] Furthermore, plastics include numerous chemical additives and absorb organic pollutants from the nearby environment.[8] Given that these substances can be transferred to organisms upon ingestion, MPs serve as carriers for additional organic contaminants.[7]

Currently, plastic waste recycling is divided into two categories: mechanical and chemical recycling. Mechanical recycling involves converting plastics into secondary raw materials or products while maintaining their original structure.[1,9] However, it faces challenges in sorting and often results in lower-quality materials.[1,9] Chemical recycling involves a range of processes that break down complex polymer structures into their basic monomers or fundamental building blocks. These building blocks can be further repolymerized to produce high-quality polymers that are comparable to virgin materials. This approach provides a more sustainable pathway toward a circular economy.[10,11]

The possibility of employing destructive mechanical forces to facilitate beneficial chemical changes is significant. The potential in polymer mechanochemistry opens the pathway to consider mechanochemical degradation and recycling as a distinctive method for plastic

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recycling.[1] Mechanochemistry is a branch of solid-state chemistry that utilizes mechanical energy to break intermolecular bonds, leading to chemical transformations.[12] Ball milling used in mechanochemistry is a useful and effective method for applying mechanical force to polymers. The polymer chains experience impacts and shear forces from the motion of the milling balls and their interaction with the walls of the milling vessel during the milling process.[13] Thus, it facilitates the intimate mixing of materials on the sub-micrometre scale, continuously generating a fresh reactive surface, and reducing diffusion limitations.[13] These mechanical stresses during the ball milling produce thermal energy, and even small elevations in temperature can significantly accelerate the rate of mechanochemical reactions.[14–17] This mechanical energy input can cause substantial molecular changes, which could lead to various chemical changes in the polymer matrix.[18] Aydonat *et al.*[19] note that the use of mechanochemical reactions for chemical recycling is a relatively new concept, though the underlying phenomena have been described for decades.[19] They emphasize the convergence of polymer and trituration mechanochemistry, wherein milling and grinding are utilized to synthesize or degrade polymers.[19]

Early research showed that mechanical forces can break polymer chains.[20] Based on this, Regel *et al.*[21] demonstrated rupture of the backbone of inert C–C backbone polymers like polymethyl methacrylate (PMMA), polystyrene (PS), and polypropylene (PP) and release of monomers due to mechanical load.[21] This pioneering observation laid the groundwork for later investigations using ball milling to generate mechanoradicals deliberately through polymer chain scission.[22] During high-energy collisions in milling vessels, mechanical forces induce homolytic bond cleavage, producing free radicals at fracture points.[23,24] These reactive intermediates undergo characteristic radical-driven processes, which include recombination, disproportionation (yielding alkenes and alkanes), hydrogen abstraction, and addition reactions, ultimately resulting in polymer fragmentation into smaller molecules.[19]

Recent advances have revitalized Regel's approach,[21] achieving over 80% monomer recovery for polystyrene (PS) through optimized ball-milling protocols.[21] Mechanochemistry is now emerging as a solvent-free method to enhance the value of these resistant materials.[19] Notably, the solvent-free nature of mechanochemical reactions aligns with key green chemistry principles, reducing waste generation and minimizing environmental impact while enhancing process efficiency.[25] Thus, polymer transformation in a mechanochemical ball-milling reactor achieves material conversion under ambient conditions, avoiding the extreme temperatures required by conventional chemical recycling methods like pyrolysis.[19,26,27] This substantially reduces energy consumption and environmental impact.[19,27]

The depolymerisation of PMMA through mechanochemistry produced a high-yield monomer.[27] In addition, great progress has been made in the mechanochemical degradation of chemically inert polyolefins like polyethylene and polypropylene.[27] The mechanochemical depolymerization of polyethylene terephthalate (PET) has been successfully demonstrated, achieving complete conversion of PET into its monomeric components through a solid-state reaction with sodium hydroxide in ball mill reactors.[28–30] The efficient depolymerization of these materials into their monomeric building blocks addresses plastic waste accumulation and creates valuable feedstocks.[30]

The monomers obtained from the mechanochemical depolymerization of plastics represent more than simple waste-to-resource outcomes; they function as fundamental building blocks for high-value industrial applications. This concept can be further expanded to high-value applications to transform waste into products of economic value. In the production of pharmaceutical packaging, thus enabling the production of high-grade bottles and containers that meet European Union (EU) and Food and Drug Administration (FDA) standards, ensuring product integrity and consumer safety without relying on virgin plastics.[31]

Furthermore, these recycled monomers from plastics can be used to create resins for 3D printing applications and can be utilized to synthesize polymer-based hydrogels, enabling the production of customized implants or drug delivery devices with precise control over shape and drug distribution.[32] Monomers derived from bioplastic-based nanoparticles (NPs) can be used as drug carriers in pharmaceutical formulations.[33] These NPs enable targeted delivery, sustained release, and even passage through the blood–brain barrier (BBB) *via* surface modifications, enhancing therapeutic efficacy.[33]

While these high-value applications demonstrate the potential of mechanochemically recycled monomers, it is essential to scale them up to meet industrial demand. Recent advancements have improved the scalability of mechanochemical processes, which can be achieved using attritor mills,[34] twin-screw extruders,[35] and drum mills.[36] These developments are crucial for establishing design principles for scaling up mechanochemical recycling, especially since large quantities of plastics will need to be recycled in the near future.[37]

Currently, there is great potential in the market for recycled plastics. The chemical recycling of plastics globally created a return estimated at around USD 14.82 billion in 2023 and is expected to grow at a compound annual growth rate (CAGR) of 9.4% from 2024 to 2030.[38] Alongside shifts in consumer behaviour, different government policies promoting plastic recycling and an increase in the utilization of chemically recycled plastics across various sectors—including automotive, electronics, packaging, textiles, and construction—have played a significant role in this change.[39]

This transition towards a circular economy presents a multitude of challenges and opportunities that must be addressed. A primary challenge lies in the need to redesign our value chains to incorporate the principles of circularity. This involves not only integrating these principles with competitive business models but also promoting their adoption among consumers and enterprises.[40] Thus, an effective collaboration throughout the entire value chain is essential for success.

Ultimately, mechanochemical degradation of polymers represents a promising selective chemical recycling strategy in today's world, where most plastic waste is not managed sustainably. This approach aligns closely with sustainability goals and circular economy principles while effectively breaking down plastics into monomers, which can be further converted into high-value feedstocks with minimized energy and environmental impact.

Conflicts of interest

There are no conflicts of interest to declare.

Data availability

There is no additional data associated with this article.

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