

Cite this: *RSC Mechanochem.*, 2025, 2, 783

# Moving mechanochemistry forward: mechanochemical polymer synthesis and recycling

Jeung Gon Kim 

DOI: 10.1039/d5mr90029h

rsc.li/RSCMechanochem

Mechanochemistry is no longer merely viewed as an emerging methodology, but is increasingly being recognized as an established chemical synthetic technique.<sup>1,2</sup> What began as a green alternative to solution-phase reactions—emphasizing solvent-free conditions and reduced energy input—is now maturing into a platform capable of enabling entirely new classes of chemical reactions that might not be achievable in solution,<sup>3–6</sup> and in doing so, is quickly advancing the boundaries of modern chemistry and chemical manufacturing.

Historically, polymer mechanochemistry was largely associated with chain scission: as early as the 1930s, a period generally considered as the birth of polymer chemistry, it was shown that mechanical forces could break the covalent backbone of macromolecules.<sup>7,8</sup> This foundation later inspired the concept of mechanophores, in which a mechanical force is used to selectively trigger chemical events.<sup>9–11</sup>

In contrast, the mechanochemical synthesis of polymers from monomers developed more slowly. A pioneering study by Kargin and co-workers, in 1959 reported the ball-milling-induced polymerization of methyl methacrylate and styrene,<sup>12,13</sup> followed by the work of Simionescu and Oprea, and co-workers, in the 1980s<sup>14,15</sup> and Kuzuya and co-workers, in the 1990s,<sup>16,17</sup>

which spurred interest in this field. At the same time, the domain of reactive extrusion—where molten polymers undergo chemical transformations inside screws—should also be considered part of polymer mechanochemistry.<sup>18,19</sup>

Since the 2010s, coinciding with the growth in interest in green chemistry, research on mechanochemical polymer synthesis has accelerated, particularly using ball milling.<sup>20–25</sup> Initially, these studies directly transferred solution-based reactions into mechanochemical platforms. Such efforts led to insights into which aspects of polymer synthetic chemical strategies derived from solution chemistry remain valid under mechanochemical conditions, and where the rules diverge.<sup>26–30</sup> Notable examples include studies that control the delicate balance between chain growth and chain scission, as well as those that exploit unique mechanochemical activation pathways to access new modes of polymer formation.<sup>31–34</sup> Today, however, the field has significantly matured, with sufficient experience and methodologies having been accumulated; and the community now looks toward new frontiers. Three directions show particular promise.

## Exclusively mechanochemical polymer synthesis

One of the unique strengths of mechanochemistry lies in its ability to operate

without solvents, allowing reactions to proceed beyond the solubility limits that restrict conventional solution chemistry.<sup>35,36</sup> This offers a powerful solution to the challenge of copolymerizing monomers with drastically different solubilities for which no common solvent can be identified.<sup>37–40</sup> Mechanochemical approaches thus open unique routes to synthesizing previously unexplored copolymer compositions and architectures. Furthermore, in cases where low solubility leads to premature precipitation, hindering the formation of high-molecular-weight polymers in solution, mechanochemistry provides a robust alternative.<sup>26,41,42</sup> By bypassing solubility limitations, it expands the landscape of polymer structures and properties that can be accessed, paving the way for the discovery of entirely new classes of functional materials. In addition, growing attention should be directed toward combining mechanochemistry with other emerging hybrid activation strategies, such as photocatalysis and electrochemistry.<sup>43–45</sup> The integration of these complementary approaches may unlock new reactivities and enable transformations that are inaccessible to any single methodology.

## Polymer recycling

Plastic waste remains one of the most pressing challenges for polymer

Department of Chemistry, Jeonbuk National University, Jeonju, 54896, Republic of Korea. E-mail: jeunggonkim@jnbnu.ac.kr



chemistry, and society at large.<sup>46</sup> Among various solutions, the chemical degradation of waste polymers into monomers or other valuable products—so-called chemical recycling—has emerged as a promising route.<sup>47</sup> Mechanochemistry has begun to play a pivotal role in this area.<sup>48,49</sup> Its ability to promote chemical transformations under solvent-free and energy-efficient conditions could decisively influence the economics of recycling processes, which often struggle to compete with petrochemical routes. Recent advances in the solvent- and catalyst-free depolymerization of polyesters and polycarbonates,<sup>50–55</sup> as well as the successful mechanochemical degradation of vinyl polymers whose strong covalent bonds typically demand high energy input, demonstrate the breadth of this potential.<sup>55–58</sup> Such achievements highlight how mechanochemistry may empower chemical recycling beyond regulatory-driven initiatives, offering pathways that combine sustainability with economic feasibility.

## Matching scalability to industrial demands

The amount of chemical products and the scale of the industries involved span a wide range across different applications. While gram-scale production may suffice for the synthesis of highly specialized fine chemicals, and kilogram quantities are often adequate in the pharmaceutical sector, the polymer industry, both for production and recycling, typically requires the manufacture of the order of tons per day. For niche, high-value polymers, the current mechanochemical equipment may already meet industrial needs.<sup>59,60</sup> However, for consumer polymer applications, entirely new approaches will be necessary to meet the demand. Emerging technologies such as twin-screw extrusion and resonant acoustic mixing represent promising avenues for scaling up mechanochemical syntheses.<sup>61–69</sup> The outcomes of these methods in organic and materials chemistry may pave the way toward a viable polymer mechanochemistry industry, enabling the transition from gram- to ton-scale production and aligning mechanochemistry with the

practical realities of industrial polymer manufacturing.

As highlighted throughout the literature, mechanochemistry not only improves upon existing solution-based methodologies, but also opens up entirely new domains of chemical reactivity. Yet, the understanding of the underlying mechanistic phenomena remains incomplete.<sup>70–73</sup> Progress will require the collective insights of many researchers, drawing from diverse disciplines to achieve deeper understanding and innovation. These efforts will undoubtedly extend to polymer synthesis and recycling, where mechanochemistry has the potential to make lasting contributions. We hope this Editorial will inspire and encourage more researchers to join the field of polymer mechanochemistry and look forward to moving the field forward together.

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