

## EDITORIAL

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## Moving mechanochemistry forward: reimagining inorganic chemistry through mechanochemistry

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Inorganic chemistry encompasses one of the broadest and most diverse domains in the chemical sciences. Comprising most elements in the periodic table, it spans molecular main-group and coordination compounds to organometallic complexes, extended solids, and functional materials. As a result, inorganic chemistry plays a central role not only in shaping and advancing the field of chemistry but also in cutting-edge technological, energy, and therapeutic applications.

Traditionally, inorganic syntheses relied heavily on solution techniques and high-temperature solid-state methods, which are invariably energy- and solvent-intensive. In recent years, mechanochemistry has emerged as a transformative force, providing a powerful alternative to conventional solution methods.<sup>1,2</sup> This shift enables not only more sustainable routes to known compounds, but also opens up entirely new reactivity landscapes, particularly well suited to the often chemically heterogeneous, structurally complex world of inorganic systems.<sup>3,4</sup>

The following highlights the opportunities of mechanochemistry for inorganic synthesis and calls on the community to help shape a cohesive framework for its future. Given the breadth of the field and limited space in this editorial, selected examples illustrate its growing impact.

Mechanochemical methods have opened exciting possibilities for the synthesis of compounds in the s- and p-block,<sup>5</sup> as well as coordination and organometallic complexes that are challenging to prepare by traditional solution chemistry.<sup>6–8</sup> Under mechanochemical conditions, reactants can be combined without the need for dry solvents or low temperatures, avoiding decomposition pathways common in solution synthesis.<sup>5</sup> The absence of bulk solvent also enables the formation of solvate-free compounds,<sup>9</sup> and allows highly reactive species, including low-valent compounds, to be generated and manipulated as solids.<sup>10</sup> Solvent-free conditions enhance the influence of non-covalent interactions (NCI) during mechanochemical transformations. Understanding the role of NCIs will transform the understanding and design of complex inorganic molecular solids and novel catalytic processes.<sup>11,12</sup>

Mechanochemistry readily activates zero-valent metals, facilitating their use in synthesis, contrasting conventional solution-based methodologies.<sup>13</sup> For instance, mechanochemistry facilitates

the direct formation of Grignard reagents, and harnesses the reactivity of alkali metals,<sup>14–16</sup> eliminating the need for ether solvents.<sup>17,18</sup> Resulting species can be employed *in situ* for subsequent organic transformations.<sup>19</sup> In the p-block, mechanochemistry enabled using bulk metals and metal oxides as entry points to complexes, circumventing toxic, highly reactive metal chlorides.<sup>20</sup> This approach is not only greener<sup>21</sup> but particularly appealing for notoriously poorly soluble reactants or those with high lattice energies.<sup>7,22,23</sup>

Mechanochemistry has been a transformative force for transition-metal chemistry.<sup>24–28</sup> For instance, tandem and cascade reactions, commonly reported in organic chemistry, can be facilitated in a ball mill.<sup>29,30</sup> Although still an emerging area, such mechanochemical cascades offer exciting possibilities for constructing organometallic and coordination compounds through sequential bond-forming steps, while avoiding solvent and thereby simplifying purification.

Mechanochemical catalysis<sup>24,31,32</sup> has often employed homogeneous and heterogeneous catalysts originally developed for solution-based methods.<sup>32,33</sup> Although most catalysts have been optimised for solution-phase reactivity<sup>34</sup>—where solubility and reactivity are intertwined—mechanochemistry has frequently outperformed conventional processes despite the lack of specific

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solvent-free media catalyst development.<sup>24,31</sup> Recent advances in catalytic CO<sub>2</sub> valorisation and ammonia synthesis illustrate how mechanochemical strategies can bypass the need for harsh thermal or high-pressure conditions.<sup>35–38</sup> These developments point to promising avenues for energy-efficient technologies suited to low-infrastructure economies and for distributed chemical manufacturing with a reduced environmental footprint.<sup>39</sup>

An area where mechanochemistry is at the interface of molecular coordination compounds and solids is the assembly of metal–organic frameworks (MOFs).<sup>40,41</sup> Using mechanochemical routes, the production has been reported of advanced porous materials, including benchmark structures such as MOF-5, ZIF-8, HKUST-1, MIL-101, and UiO-66, with surface areas comparable to those resulting from conventional methods, as well as multimetallic MOFs and previously not reported phases.<sup>40–42</sup>

Mechanochemistry has emerged as a game-changer in materials science, providing a powerful tool for synthesising novel materials with structural and functional properties that may not be achievable through conventional synthetic routes. The “green” aspects of mechanochemistry have been illustrated, for example, in the synthesis of nanostructured and non-equilibrium complex oxides,<sup>43</sup> where highly defective and metastable nanostructured complex oxides with distinctive functionalities can be mechanochemically synthesised in a single processing step at ambient temperature, without needing solvents and/or calcination under a controlled oxygen fugacity.

The green metrics of mechanochemical processes contrast sharply with those of traditional solid-state methods, which involve multiple stages such as powder precursor homogenisation, reactant compaction, and extended heat treatment at high calcination temperatures or under a reducing atmosphere. Thus, the appeal of mechanochemistry lies in its simplicity, energy efficiency, and environmental friendliness. Despite promising developments in this field, the facile synthesis of inorganic materials, particularly in nano-sized, nanostructured, and

far-from-equilibrium states remains a significant challenge.

Understanding solid-state reactions in molecular-inorganic solids and extended lattices is essential to establish mechanochemistry as a central synthetic method in inorganic chemistry and materials science. Once viewed as a “black box”, an understanding of the field has advanced through real-time, *in situ* analytical techniques. Synchrotron X-ray powder diffraction, Raman spectroscopy, and solid-state NMR enable direct observation of phase transitions, intermediate species, and reaction kinetics during milling. These insights enhance our mechanistic understanding, and time-resolved analysis will be crucial for developing predictive models and guiding reactivity precision.<sup>44–46</sup>

The application of various comprehensive spectroscopic and electron microscopy techniques has provided detailed, complementary atomic-scale insights into the nature of local short-range (dis)order in nanostructured materials.<sup>47</sup> It has been demonstrated that the interfacial regions of as-prepared nanomaterials exhibit a myriad of unique physico-chemical features, including structural deformations (e.g., distorted structural units, tilted polyhedra, ruptured polyhedral bonds, reduced coordination of cations, cation anti-site disorder, oxygen vacancies, random cation distribution over crystallographic sites). These structural imperfections give rise to a wide variety of electronic and magnetic phenomena, including defect electronic bands or magnetic anomalies such as non-collinear spin arrangements and the collapse of long-range magnetic ordering.<sup>48,49</sup>

The accumulated knowledge on the nature of short-range disordering phenomena in solids has contributed to understanding their role in mechanochemical syntheses. The mechanism of mechanically-driven syntheses of non-equilibrium phases is interpreted as arising from impact-induced nucleation and growth processes, spatially confined to the structurally disordered and chemically reactive regions found at the strained contact points of the precursor interfaces.<sup>50–52</sup>

Far-from-equilibrium mechanochemically synthesised nanostructured

materials exhibit unique macroscopic properties that are typically unattainable through conventional synthetic methods. Both better and worse magnetic and electronic properties of nanoceramics produced *via* mechanochemistry have been shown, emphasising the clear-cut explanation of the interplay between their short-range structural disorder and magnetism.<sup>48,49</sup> Mechanochemistry has also been shown to be important in the preparation of photocatalytically and electrochemically active oxides.<sup>53</sup>

Inorganic materials prepared through mechanochemical routes are inherently unstable due to their far-from-equilibrium structures. Their stability is primarily determined by the nature of structural disorder, confined to interfacial and near-surface regions of the as-prepared compounds. For example, the range of thermal stability of mechanochemically-made nano-oxides is shown to extend to about 500–700 K.<sup>43</sup> Heated above these temperatures, the nanostructured oxides undergo gradual structural relaxation, reaching an equilibrium state similar to that of their bulk counterparts prepared *via* conventional routes.

Moreover, mechanochemistry is increasingly recognised as a methodology to design novel energy-efficient, low-cost routes to existing, as well as new materials. Its applications span energy storage,<sup>54</sup> OLED materials,<sup>55</sup> nanocomposites,<sup>56</sup> nanocrystals,<sup>57</sup> thermoelectric materials,<sup>58</sup> metal hydrides,<sup>59,60</sup> nanomaterials,<sup>61</sup> hydrogen-storage materials,<sup>62</sup> chalcogenides<sup>63</sup> and perovskites,<sup>64,65</sup> at different scales.<sup>66</sup>

Mechanochemistry may not replace traditional methods, but will be a transformative tool in inorganic synthesis. It enables new reactivities, promotes sustainability, and unlocks access to materials that are otherwise difficult to obtain. Beyond its scale-up potential, mechanochemistry fosters discovery-driven research. Its solid-state environment decouples solubility from reactivity to open opportunities to explore unconventional reactivity, bonding motifs, and elusive compound classes.

As mechanochemical platforms continue to evolve—with enhanced temperature control and integrated



monitoring systems—their practical applicability and broader adoption are expected to increase. This, in turn, will support the development of more standardised protocols, parameter databases,<sup>67,68</sup> and predictive tools (such as machine learning and artificial intelligence),<sup>69–71</sup> improving reproducibility and accelerating discoveries across the diverse landscape of inorganic chemistry.

Realising this potential will require close collaboration among researchers, technologists, and engineers. In that respect, the International Mechanochemical Association (IMA)<sup>72</sup> – one of the organisations associated with IUPAC – has emerged as a crucial player that can catalyse the implementation of the international cooperation needed to enhance and harmonise research in mechanochemistry worldwide. Furthermore, the newly formed EuChemS “Working Party on Mechanochemistry”,<sup>73</sup> which builds upon the legacy of the European COST Action CA18112 “Mechanochemistry for Sustainable Industry”,<sup>74</sup> represents an important step forward for the mechanochemistry community in Europe. These initiatives are particularly important at a time when mechanochemical processes are being investigated at both laboratory and large scales, targeting potential applications across various industrial market sectors.

Currently, several new global initiatives are underway aimed at further promoting mechanochemistry to realise its full potential for science and technology. Examples are the project IMPACTIVE (Innovative Mechanochemical Processes to synthesise green ACTIVE pharmaceutical ingredients),<sup>75</sup> funded by the European Union, and the NSF Center for the Mechanical Control of Chemistry (CMCC),<sup>76</sup> funded by the National Science Foundation. We hope these efforts will inspire the further creation of dedicated research centres and funded initiatives worldwide, especially where mechanochemical research is just emerging or underrepresented.

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