

Microdroplet Mechanochemistry

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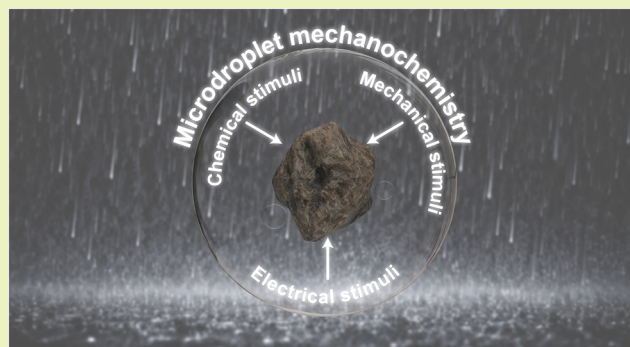
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ABSTRACT: Water, mother of all fluids, long considered passive, is now shown to be highly active in the form of droplets, where chemical reactions requiring intense stimuli, happen rapidly. Discoveries from our lab in the recent past have shown that hard, common minerals such as quartz and ruby disintegrate spontaneously to form nanoparticles in charged water microdroplets. This occurs due to a combination of intense electric fields, reactive chemical species and mechanical forces in the droplets, acting in concert on the minerals. Subsequent studies have shown that this is a generic process occurring in a variety of materials. This evolving field of microdroplet mechanochemistry presents a low energy, reagent-free and ambient method for water-based transformations of materials. As the process occurs in milliseconds rather than the millennia conventionally associated with mineral disintegration, it effectively compresses geological time scales. The possibilities of such a process as a new method of materials discovery and green manufacture is outlined here. This positions water as a green factory making materials sustainably.

KEYWORDS: microdroplet chemistry, mechanochemistry, mass spectrometry, materials, environmental science, catalysis, multiscale modeling, sustainable chemistry



INTRODUCTION

When Water Becomes Active. Water dissolves, disperses and solvates matter but rarely constructs it directly. A medium that is typically a silent observer is now beginning to be very active at small scale, in the size regime of microdroplets. At the interface of charged, levitating, sprayed, and evaporating microdroplets, large electric fields^{1,2} in the range of 10^8 V m⁻¹ are reported. Consequently, reactive chemical species and mechanical stresses are observed and they collectively make the confined droplet to a microreactor, figuratively a dynamo. Our discovery³ of disintegration of common minerals to nanoparticles extends the microdroplet chemistry^{4–6} to the solid state, unveiling a new field of science—microdroplet mechanochemistry.

Microdroplet Mechanochemistry (MDMC) is defined as the “transformation of materials within charged or confined liquids by coupled electrical, mechanical and chemical effects”. The curved surfaces of droplets experience strong electric fields, chemical gradients arising from reactive species such as protons and intense transient mechanical stresses such as those arising from Coulomb fission, inducing lattice reconstruction, fracture and associated effects in the materials. Unlike conventional mechanochemistry driven by external stimuli, MDMC occurs due to the energy inherent in the system and occurs under ambient conditions. The process represents a unique electro-

mechanochemical regime where solvent—most efficiently water itself—acts as an agent of materials synthesis and transformation.

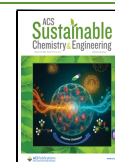
Discovery and Implications. When rocks break in milliseconds: In our initial work, quartz, ruby and synthetic alumina particles of 5–10 μm were dispersed in water sprayed to make charged droplets.³ The products were collected a few cm away from the spray showed uniform nanocrystalline particles of ~5–10 nm in diameter exhibiting specific crystallographic planes. From the distance traveled and estimated number of charges, for accelerating droplets of finite size, we estimate the time taken for spray deposition as ~30 ms. As the particles disintegrate, transient structures such as “flower-like” intermediates were seen. High resolution transmission electron microscopy (TEM) coupled with powder X-ray diffraction (XRD) confirmed that the minerals disintegrated to form nanoparticles. Computations suggested that the intense electric fields and the chemical species, especially protons are involved in the process. The electric fields polarize the bonds and the protons help in the breakage of the particles. Other chemical

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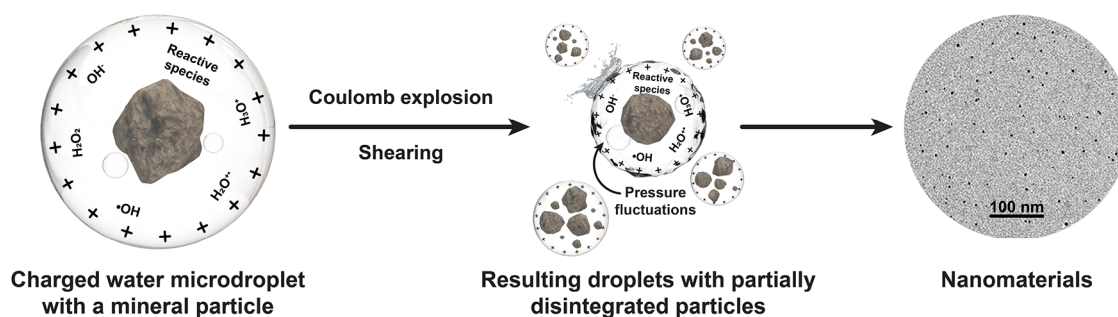


Figure 1. A schematic representation of one form of *Microdroplet Mechanochemistry* where materials transform within charged or confined aqueous microdroplets through coupled electrical, mechanical, and chemical effects. The charges shown may be due to different species. Species other than OH^+ , H_2O^{+*} , H_3O^+ , OH^- , etc., can also be present depending on the ingredients in the droplet. The case shown is that of water. The TEM image shown was adapted from ref 3, Spoorthi et al., *Science*, DOI: 10.1126/science.adl3364 [2024], AAAS.

species such as H_2O^{+*} and OH^+ might also be involved. These species reduce the barrier for slip or fracture. The transient mechanical stresses produced during the Coulomb fission of the droplets might be the trigger to cause the disintegration and further facilitated by the reactive chemical species. The observed phenomenon for fracture is labeled as “proton-induced slip”. On the whole, these processes constitute a field-driven, proton-assisted lattice breakdown—a unique coupling of electrostatic and mechanical forces in the confinement of droplets. Microdroplet Mechanochemistry leading to the disintegration of minerals is represented schematically in Figure 1.

Mechanistic Framework. Microdroplet mechanochemistry integrates three otherwise independent stimuli for transformation: (i) electrostatic fields concentrated by curvature, (ii) proton mediated processes at hydrated interfaces, and (iii) mechanical impulses from Coulomb fission, or physical events. These act cooperatively, enabling reactions inaccessible in bulk liquids or classical mills. The interface clearly represents a distinct phase of reactivity⁷—a transient “electro-mechanochemical state” of matter. Subsequently, it was shown that graphene and MoS_2 also disintegrate in charged water microdroplets making corresponding nanoparticles.⁸ Disintegration occurs as the droplets fly through air and the process could be visualized systematically.⁹

Connecting to Sustainability and Green Chemistry. Microdroplet mechanochemistry aligns naturally with the core principles of green chemistry;¹⁰ (i) waste prevention (no auxiliary reagents), (ii) safer solvents (water), (iii) energy efficiency (ambient conditions), and (iv) inherently benign design (minimal waste). It enables (v) rapid, high-yield transformations (atom economy), using (vi) abundant minerals (renewable raw materials), without external energy inputs and facilitates (vii) circularity (recycling) of the medium. Other aspects of green chemistry may also be considered. The process parallels events in nature—such as those in aerosols and clouds—and therefore offers a direct connection between the planet’s material cycling and manufacturing processes. For industrial adoption of this technology, cost of manufacture by microdroplet mechanochemistry must be comparable to conventional processes. In view of the benefits outlined above, the cost of manufacture is expected to be lower although production at scale is needed to make realistic cost calculations.

Natural Nanofactory. Aerosols are everywhere in Nature. Such particles carry charges¹¹ due to triboelectricity due to atmospheric friction as well as dissolved ions and they have been long implicated in interface chemistry. Droplets are known to have potential differences. Due to the diversity of processes by

which such droplets can be produced such as sea spray, fog, and rain, events such as the ones demonstrated in our work may have been important in early-Earth chemistry such as weathering of rocks, formation of colloidal minerals and associated chemistry making early molecules.⁷ From this perspective, every raindrop can be viewed as a nanofactory of minerals.

Interfacial Chemistry. Microdroplets can contain nanobubbles composed of dissolved gases, which possess reactive interfaces. Changing solubility of gases in charged confined media may lead to such phenomena. These bubbles can be visualized exhibiting unusual phenomena.¹² Electric fields at such interfaces due to the presence of surfactants and physical effects of shape and concentration gradients can induce reactivity, leading to transient chemical species. The observed MDMC may be initiated by a combination of reactive species (originating from micro/nano bubbles or otherwise), just as radical initiators start certain polymerization reactions. Electric fields of $(5-9) \times 10^7 \text{ V cm}^{-1}$ have been observed at the interface oil droplets of $250 \mu\text{m}$ diameter in water.¹³

Origins. Right after the discovery of molecular reactions in organic and aqueous environments,⁴⁻⁶ nanoparticles of gold were synthesized using microdroplets.¹⁴ Controlled deposition of such nanoparticles gave functional and patterned Raman active surfaces.¹⁵ Controlling electric fields on depositing particles created nanowires from nanoparticles.¹⁶ Such nanowire structures, created over cm^2 areas, harvest water efficiently.¹⁷ Zare and colleagues too made gold nanoparticles.¹⁸ Later, it was found that large, polydisperse, monolayer-protected silver nanoparticles anneal to form smaller, monodisperse analogues within ambient microdroplets.¹⁹ This demonstrates the capacity of droplets to reorganize matter, breaking and making hundreds of chemical bonds. Together, these findings established a continuum—from droplet-assisted synthesis to restructuring. This prompted us to explore the possibility of transforming macroscopic materials with extended chemical bonds, as in a piece of rock, by electro-mechanochemical transformation in a charged droplet. That culminated in positioning microdroplet mechanochemistry as a sustainable route to the creation of materials. While many of the observed processes still need deeper investigations to understand better, it is beyond doubt that droplets can break and make chemical bonds from molecular to macroscopic length scales.

Green Nanomanufacturing. MDMC can be harnessed for green nanomanufacturing. The only consumables needed are solvents—and water is preferred—which can be recycled. The power requirement is minimum as processes occur under ambient conditions. The applied electrical potentials are in the

range of kV, but currents are low. Types of products and their properties, such as size and shape, can be controlled by the experimental parameters such as potential, spray distance, concentration of the reagents and composition of solvents. Conducting experiments with multinozzle electrospray and continuous spray with repeated cycling can increase conversion yields.²⁰ Because droplets disintegrate in air and deposit nanoparticles directly onto substrates, the process functions as a room-temperature nanoparticle sprayer—an ambient, scalable route to catalysts, coatings, and sensors. Renewable sources may be used for electrical power.

Comparison with Conventional Mechanochemistry. Conventional mechanochemistry uses macroscopic grinding, friction, or compression to drive reactions in the solid state, generally at significant energy cost.²¹ In contrast, microdroplet mechanochemistry operates through intrinsic electro-mechanochemical coupling at ambient conditions, performing reactions without external mechanical energy input. Besides, unlike conventional mechanochemistry where forces operate from outside, MDMC operates from within as electro-mechanochemical coupling occurs at this length scale, within the droplet.

Next Steps. The materials formed have active surfaces and naturally, they are sites for new catalysis. The droplets can contain reagents which can be transformed at these surfaces using the transient ingredients in the droplets. Stimuli such as photons can provide additional incentives for such processes. As chemical reactions can also occur in microdroplets, MDMC can also make new solids like solvothermal synthesis, although at unprecedentedly shorter time scales.

Impact on Other Disciplines. Potential impacts of MDMC span various fields, including: atmospheric chemistry (aerosol reactivity and cloud nucleation), environmental remediation (defect-rich, in situ-born nanoparticles for catalysis), planetary science (charged ice–regolith interactions), and sustainability education (a new paradigm of water as an architect of matter). Many of these implications are presented in Figure 2. In the following, we dwell on some of these aspects.

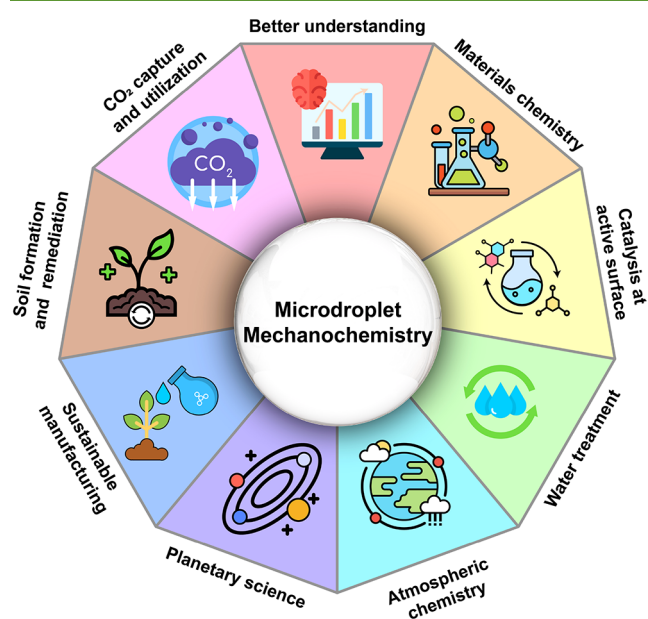


Figure 2. Expansion of the phenomenon of MDMC and implications to other areas.

Atmospheric Chemistry and Cloud Nucleation. Charged water microdroplets are ubiquitous in sea spray and clouds, yet their application as active chemical reactors remains underexplored. Laboratory observations³ of spontaneous mineral disintegration in such environments suggest similar pathways for making silicates, metal oxides, clays, etc., analogous to those detected in the atmosphere. MDMC therefore provides a unified framework to examine chemo-mechano-electro-driven processes that may underlie atmospheric processes, complementing studies of interfacial oxidation in atmospheric aerosols.

Environmental Remediation and Catalysis. Surfaces of nanoparticles formed by MDMC can possess high catalytic activity. Spraying contaminated water through charged nozzles could generate active nanocatalysts for in situ degradation of dyes, nitrates, or emerging pollutants such as perfluorinated alkyl substances (PFAS). Analogous field-driven reduction of CO₂ to formate and nitrogen fixation in aqueous microdroplets illustrate the feasibility of such reagent-free remediation processes through interfacial electrochemistry.^{22,23} Triboelectric degradation of poly(trifluoroethylene) (PTFE) in water occurs when PTFE-covered magnetic pellets are stirred in bulk water.²⁴ PFAS degradation has been demonstrated by microdroplet chemistry.²⁵ Active radicals and other species in conjunction with nanomaterials could offer a method for water treatment. Such processes along with traditional water treatment materials such as aluminates, silicates and phosphates (e.g., NaAlO₂, CaSiO₃) may be advantageous as no unknown substance is then introduced in the treatment process. Besides MDMC, microdroplet chemistry in other forms can also help in environmental remediation. For example, Mo-rich holes can be created in MoS₂ nanosheets by electrospray, which can make H₂O₂ from water upon light exposure, leading to water disinfection.²⁶ Nanowires formed by microdroplet chemistry can be used for water harvesting, as noted before.¹⁷

Planetary and Geochemical Contexts. Evolution of the planet, soil formation, dissolution of minerals, chemical synthesis or relevance to life, etc., are important to consider in the context of MDMC. Triboelectricity of droplets can lead to MDMC of entrapped particles and molecules. Field-assisted mineral transformation or redox chemistry may occur at ice surfaces and at cryogenic temperatures of relevance to other planets or outer space, paralleling liquid droplet-driven fragmentation in the laboratory. These considerations extend MDMC to space, although in the solid state, where similar forces may sculpt surfaces and prebiotic materials.

Educational Perspective. MDMC represents sustainability in action—chemistry without external energy or waste. Demonstrations of mineral-to-nanoparticle conversion under ambient conditions provide a compelling tool for teaching interfacial phenomena, green chemistry, and materials transformation. An MDMC experiment in the classroom may be an important way to demonstrate sustainable synthesis of nanomaterials.

Multi-Scale Modeling Opportunities. A challenge or an opportunity in MDMC is that the relevant physics spans multiple orders of magnitude, from Å-scale solvation structures to tens-of-micrometer hydrodynamic deformations and from femtosecond bond dynamics to millisecond droplet lifetimes. It is clear that no single modeling approach can capture this range currently. At the molecular level, reactive MD, QM/MM, and machine-learned potentials can reveal how constituent species experience partial desolvation, interfacial orientation, field fluctuations, evaporation-driven compression within charged

droplets as well as their Coulombic fission. Such simulations are ideally suited to quantify the relative contributions of electric fields, confinement, ion partitioning, and mechanical stresses to bond activation. However, such simulations will require realistic inputs such as evolving local concentrations, which in turn demand microkinetic descriptions and field-dependent elementary reaction rates that must be determined self-consistently.

At mesoscopic and continuum scales, coarse-grained MD, dissipative particle dynamics (DPD), and electrohydrodynamic models can describe evaporation kinetics, hydrodynamic shear, droplet oscillations, charge redistribution, and Rayleigh instabilities—the boundary conditions actually experienced by molecules. A long-term goal is the development of a coupled multiscale framework in which continuum electromechanics provides time-resolved surface charge, curvature, evaporation rate, and stress fields, while atomistic simulations model chemistry under these evolving conditions, allowing feedback between chemical events and droplet mechanics. Such an integrated approach would enable predictive modeling of reaction acceleration, selective bond activation, nanoparticle nucleation, and even triboelectrically charged nebulized droplets, which may operate under distinct mechanochemical regimes.

Developing this hierarchy of models is central to establishing microdroplet mechanochemistry as a quantitative, designable platform for chemical and materials discovery. Such models are especially valuable in regimes that are experimentally inaccessible—allowing one to predict, for example, whether ultrafast field spikes during droplet deformation can transiently lower activation barriers, how concentration and desolvation profiles evolve in droplets below optical diffraction limit, or whether triboelectrically charged nanodroplets reach charge–density or curvature thresholds capable of initiating chemistry. Ultimately, such predictions must help explain experimental realities.

Although classical electrohydrodynamics of droplets such as Taylor cones, the leaky-dielectric model, charge relaxation, and Rayleigh instabilities, is well developed in certain parameter regimes, these frameworks describe systems that are closer to equilibrium than those encountered in MDMC. In microdroplet regime, several interfacial processes occur simultaneously, creating a regime where electric fields, hydrodynamic flow, reactive transport, and mechanical stresses are coupled and evolving dynamically. The resulting electro-reacto-hydrodynamics of curved, deforming, and chemically active droplet interfaces is largely unexplored, and defining its governing principles represents a major open challenge - and a rewarding opportunity - for the field.

Critical Overview. Although the observed phenomena have been reproduced across laboratories, there have not been consensus on the origin of reaction acceleration in microdroplets. In particular, questions have been raised about the extent to which electric fields contribute relative to other factors such as concentration, phase behavior, and confinement. Early interpretations of microdroplet reactivity often invoked extraordinarily strong interfacial electric fields,¹ estimated at 10^7 – 10^8 V·cm⁻¹ (equivalent to roughly 0.01–0.1 V·Å⁻¹), as inferred from Stark-shift measurements.

Reactive-force-field simulations of curved interfaces predicted average surface field strengths of ~ 16 MV/cm, which when projected onto free surface O–H bonds can reach an order of magnitude larger.²⁷ Measured fields of $(5$ – $9) \times 10^7$ V cm⁻¹ at oil–water interfaces,¹³ mentioned earlier in the text may arise from anisotropic hydrogen bonding and charge asymmetry. At

the same time, quantum many-body analyses report that interfacial electric fields at the pristine air–water interface are systematically $\sim 10\%$ weaker²⁸ than in bulk, reflecting density depletion rather than amplification, and *ab initio* molecular dynamics simulations on a model Diels–Alder (DA) reaction show that interfacial electric fields fluctuate between -0.1 and $+0.1$ V·Å⁻¹ but average to only ~ 0.007 V·Å⁻¹, a magnitude far too small to meaningfully influence the reaction barrier.²⁹ Even a perfectly oriented external field of 0.1 V·Å⁻¹ lowers this barrier by only ~ 2 – 3 kcal·mol⁻¹ (ref 29), understandable for a nonpolar/nonionic intermediate of a DA reaction. Complementary SFG studies³⁰ report no persistent macroscopic fields at pristine air–water interfaces. Xie and colleagues suggest that many earlier “giant-field” interpretations conflate rapidly fluctuating microscopic fields—present in all hydrogen-bonded liquids—with sustained macroscopic electrostatic forces capable of catalysis.²⁹ These findings, together with the broader framework of electric-field-mediated catalysis,³¹ indicate that although strong fields may occur at certain chemically structured interfaces, pristine air–water and neutral microdroplet interfaces do not typically exhibit anomalously large, catalytically dominant fields.

In the present Perspective, however, *Microdroplet Mechanochemistry* is introduced based on observations of materials transformations occurring within charged microdroplets generated by electrospray, an environment different from that of neutral interfaces.^{32–34} Electrosprayed droplets inherently possess net charge, exhibit surface charge segregation, and undergo evaporation, shape oscillation, Coulombic deformation, and Rayleigh fission, which are well-documented.^{32–36} These processes generate intrinsic interfacial electric fields, steep charge-density gradients, and intense pressure fluctuations absent in macroscopic liquids. It is within this highly nonequilibrium environment, characterized by partial desolvation, transient confinement, hydrodynamic shear, evaporation-driven compression, and charge-induced interfacial restructuring, that chemical reactions and materials transformations occur.^{37,3} Thus, the “mechanochemistry” in Microdroplet Mechanochemistry arises not from macroscopic grinding but from the synergistic action of confinement, charge, interfacial fields, and dynamic mechanical stresses unique to charged electrospray droplets.^{33–37} Moreover, at sufficiently high liquid flow rates, nebulization without electrospray can itself generate substantial triboelectric charging, creating transient interfacial fields and stresses that may also contribute to Microdroplet Mechanochemistry.¹¹ Overall neutral droplets too may possess transient fields due to dynamic fluctuations at the nanometer length enhancing chemical reactions.

Currently, the most important limitation of the area is processing at scale. The synthesis of nanoscale materials using MDMC is limited to milligrams per hour in published literature (2024) and processing at tens of grams per day is happening now. Scale-up to kilograms is essential for detailed investigations and adoption of the method to other materials of industrial value. Processing in water suggests potential applications of such materials in agriculture, water treatment, construction industry, CO₂ sequestration, cosmetics, personal care products, paints, detergents and many others, although capacity to make materials at hundreds of tons at affordable cost is essential for industrial adoption. At that scale, the green chemistry advantages will begin to drive many industries to adopt MDMC in the production workflow, with consequent benefits to Sustainable Development Goals. The science of MDMC will be understood

better with a combined electro-mechanochemical approach with inputs from time-resolved examination of droplets with nanosecond or better resolution. The phenomena occurring in the droplets such as luminescence, formation of reactive species, propagation of shock waves during droplet fission, etc., need examination. These provide crucial insights leading to accurate modeling of the process.

Future Directions. For a mature discipline of microdroplet chemistry in general, and MDMC in particular, in which processes can occur with predictive outcomes so that engineering and technology development become possible, we need to (i) establish international partnerships and shared resources; (ii) develop in situ diagnostics for droplets in flight with good time resolution; (iii) test scaling laws linking droplet size, charge and experimental parameters with material hardness, and nanomaterial yield; (iv) Advance modeling spanning multiple scales, including (a) electronic and atomistic methods to capture field–reactivity coupling at curved, charged interfaces, (b) mesoscale electrohydrodynamics to describe evaporation, deformation, and reactive flow, and (c) upscaling frameworks that connect MDMC principles to atmospheric, environmental, and geological processes; (v) quantify the relevance of observed phenomena to atmospheric processes using experiments in controlled environments; and finally (vi) engineer continuous droplet reactors at scale for circular materials chemistry so that industrial applications become possible.

The subject of MDMC presents a new possibility of conducting chemistry without external energy. The process unites chemical, electrical and mechanical forces to transform or make materials fast with reduced impact on the environment. An expansion of the area will lead to greater collaborations between chemistry, physics, biology, geoscience, and sustainability science. In the years ahead, I see the establishment of factories where materials are made in droplets sustainably with new application areas being evolved.

While the discussion has focused on water as a Newtonian fluid, the principles of MDMC extend naturally to complex fluids. Polymer and biopolymer solutions, surfactant-rich mixtures, ionic liquids, and colloidal or nanoparticle suspensions are important to consider. In them, viscoelasticity, microphase separation, interfacial structuring, and long relaxation times may couple nonlinearly with charge transport, evaporation, and interfacial fields, creating mechanochemical pathways distinct from those in simple aqueous droplets. For example, understanding how chain entanglement, micellar dynamics, or colloidal assembly respond to droplet deformation, fission, and interfacial charging represents an unexplored frontier. Obviously, MDMC in complex fluids is a rich direction for discovering new reactivity, emergent materials, and non-equilibrium structures that are inaccessible in bulk phases. Although some experiments have been performed, work in this area is at infancy.

CONCLUSIONS

Microdroplet chemistry in general and microdroplet mechanochemistry in particular, transform our understanding of both water and matter, blurring boundaries between electrical, mechanical, and chemical forces at small scales. In fact, the emergence of the area is a unification of these disciplines to address sustainability. In Green Chemistry terms, it is a zero-carbon, zero-waste pathway to materials chemistry. Every droplet becomes a reminder that this chemistry is already

happening around us—in clouds, in rain, and now, in the laboratory.

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Author Contributions

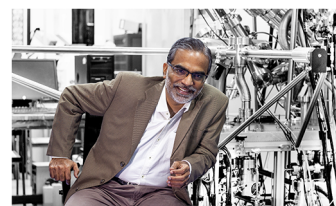
T.P. conceived the idea and wrote the manuscript.

Notes

We emphasize that the process described is not mechanochemistry in the conventional sense of stress- or strain-driven chemistry in the solid state, but rather an electric-field-dominated regime in which mechanical effects are themselves field-induced and the mechanical component is not externally applied but emerges from the electrostatic forces and Coulomb instabilities in charged droplets. As microdroplets are often formed by electrospray, electrical charges are already part of the droplets. Even for microdroplets formed by other means such as nebulization or cavitation, charges exist on their surfaces. The reactive chemical species in droplets exist largely due to the large electric field. Although chemo-electro-mechano effects are acting in unison at this length scale, we propose to call the area as Microdroplet Mechanochemistry. Adding prefixes (such as electro-, chemo-, physico- to mechanochemistry) can dilute the conceptual beauty and may risk the growth of the field to be inclusive with other stimuli.

The author declares no competing financial interest.

Biography



Thalappil Pradeep is an Institute Professor and Deepak Parekh Institute Chair Professor at the Indian Institute of Technology Madras (IITM), Chennai, India. His research interests are in molecular and nanoscale materials. He has authored 600 scientific papers, 10 books, 100+ Indian and 29 US/PCT patents, cofounded 7 start-ups and founded the International Centre for Clean Water (<https://icw.world>). He is involved in the development of affordable technologies for drinking water purification. His pesticide removal technology has reached >10 million people. His arsenic removal technology, approved for national implementation, is delivering arsenic free water to >1.4 million people every day. He is a recipient of recognitions including the Shanti Swaroop Bhatnagar Prize, The World Academy of Sciences (TWAS) prize, Padma Shri, Vigyan Shri, Nikkei Asia Prize, Prince Sultan Bin Abdulaziz International Prize for Water, VinFuture Prize and ENI award. He is a Fellow of all the science and engineering academies of India, TWAS, American Association for the Advancement of Science, African Academy of Sciences, US National Academy of Engineering and Academia Europaea. He has received the *Lifetime Achievement Research Award* of IITM and *Distinguished Alumnus Award* of Indian

Institute of Science. As part of philanthropy, he supports a school in his village where 500 students are on rolls.

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