

Perspective

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Soft devices empowered by mechanoluminescent materials

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Abstract

Mechanoluminescence is the phenomenon in which certain materials emit light when subjected to mechanical stimuli, such as bending, stretching, or compression. Soft devices containing embedded mechanoluminescent materials are capable of responding to mechanical deformation by emitting light, which can be utilized for various applications, including sensing, display, communication, and visual feedback. In this Perspective, we discuss recent advancements and emerging applications of mechanoluminescent materials for soft devices, with a focus on the remaining challenges in mechanoluminescent materials, such as performance, mechanism, synthesis, and device fabrication, that need to be addressed for developing advanced soft devices, and propose the potential solutions.

Keywords: Mechanoluminescence, mechano-to-light conversion, soft devices, wearable electronics, internet of things



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INTRODUCTION

Soft devices, including flexible/stretchable sensors, processors, actuators, and displays, have gained substantial interest due to their remarkable potential to revolutionize sectors such as precision healthcare, robotics, personal electronics, energy, and human-machine interaction. Their ability to conform to irregular shapes and endure deformation without sustaining damage paves the way for new capabilities and improved user experiences^[1,2]. Integrating luminescence into soft devices offers an additional dimension to their applications, promising exciting advances in areas such as camouflage, communication, visual feedback, and improved visibility in low-light conditions such as *in vivo* manipulation and therapy^[3,4]. However, the luminescence in current soft devices primarily relies on triggers such as electricity or light, and thereby, their applications were restricted due to the requirement for auxiliary electrical or optical components and power supplies, adding complexity to device fabrication and system integration.

Mechanoluminescence (ML) refers to the emission of light from materials in response to external mechanical stimuli, such as stretching, rubbing, compressing, scratching, shaking, grinding, cleaving, and impacting^[5]. This intriguing phenomenon is commonly observed in both natural and synthetic materials^[6,7], such as quartz, diamond, aluminates, silicates, oxysulfides, alkali halides, and AIEgens, tracing back to its first scientific record in 1605 when Francis Bacon noticed it upon scraping hard sugar with a knife. It is estimated that about one-third of organic molecule solids and half of inorganic salts exhibit ML, covering conductors, semiconductors, and insulators^[8]. The mechano-to-light conversion of ML materials is straightforward and energy-autonomous without any assistance from other forms of energy, such as electron or photon excitation. This unique characteristic makes ML materials highly suitable for diverse areas. [Figure 1](#) highlights the development of ML materials and their representative applications.

In the realm of soft devices, a variety of cutting-edge functional technologies have emerged. These innovations encompass tactile sensors^[9,10], displays^[11-13], actuators^[14], transducers^[15], and wearable components^[16,17], all powered by ML materials. This transformative field has witnessed significant developments ever since Jeong *et al.* introduced a flexible ML composite film with a brightness of ~120 cd/m² and a remarkable durability of over 100,000 mechanical cycles^[18]. For instance, Qian *et al.* reported a photonic skin composed of ZnS:Mn/Cu@Al₂O₃ particles embedded within a nano-doped poly(dimethylsiloxane) (PDMS) matrix. This creation has the capability to display captivating augmented animations and expressions^[19]. Wang *et al.* demonstrated a flexible ML-based device for monitoring heartbeats constructed using pure organic luminogens. This innovation holds promise for applications in communication, information storage, and healthcare^[17]. Li *et al.* presented a mechanoluminescent robot infused with bioluminescent algae, enabling optical signaling and illumination^[14]. Hou *et al.* showcased an interactive mouthguard equipped with an array of ML-powered distributed-optical-fiber (mp-DOF) sensors that can recognize the patterns of dental occlusion and sequentially control devices such as wheelchairs and smartphones^[20]. Furthermore, we recently unveiled an untethered tactile sensing glove that leverages a combination of heterogeneous mechanoluminescent materials. This innovation enables the discrimination of multiple tactile stimuli and facilitates wireless human-machine interactions^[21]. Despite these significant progresses, several issues related to ML materials, including performance, underlying mechanisms, synthesis, and device fabrication, still need to be addressed to fully exploit their potential in engineering innovative soft devices. In this Perspective, we delve into recent advancements and emerging facets of these aforementioned challenges concerning ML materials while also proposing potential solutions.

PERFORMANCES OF ML MATERIALS

ML can be divided into triboluminescence and deformation luminescence, each distinguished by the method of luminescence induction. Triboluminescence is usually attributed to a triboelectric field, a

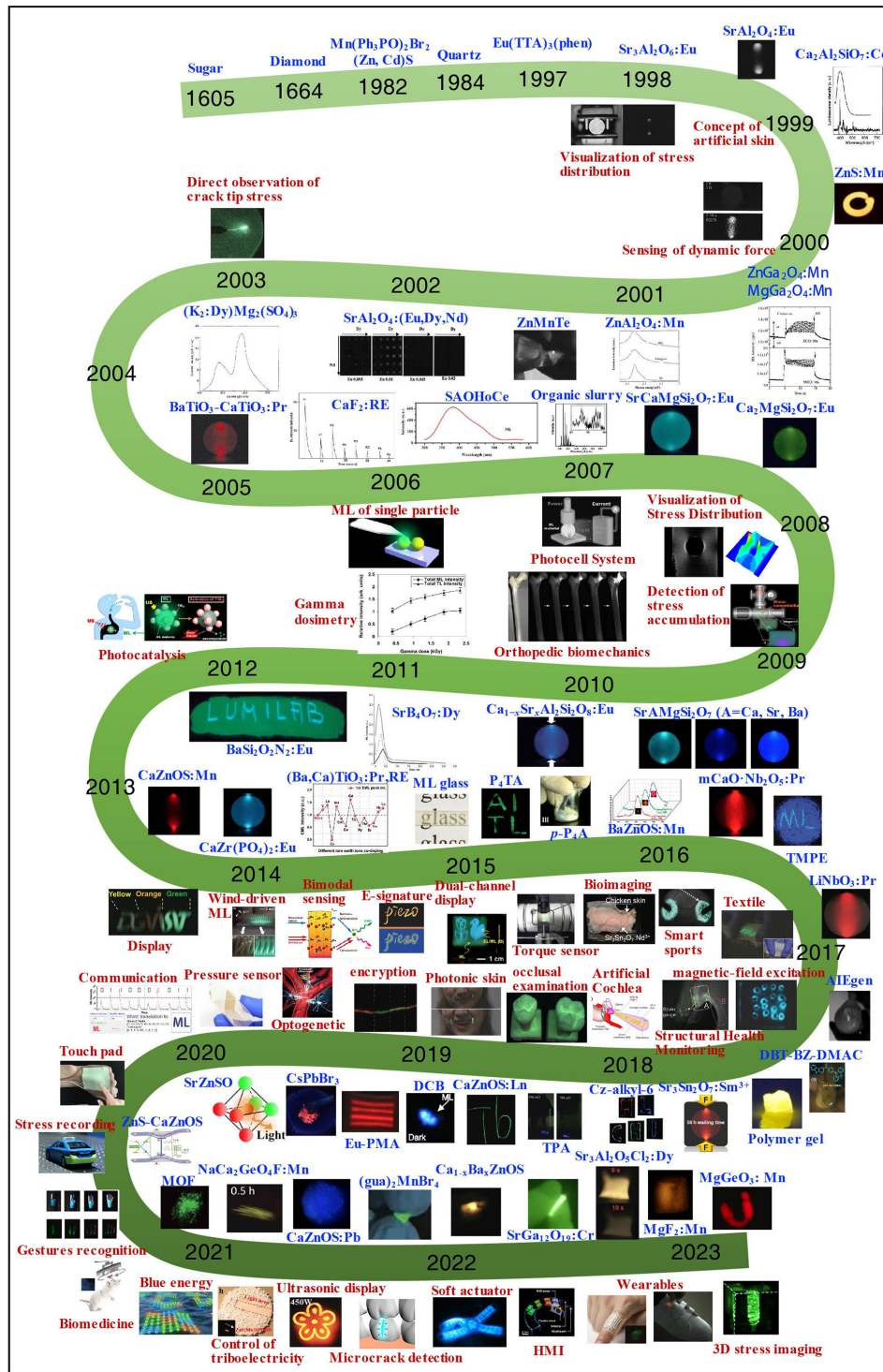


Figure 1. The development of ML materials and their representative applications. ML: Mechanoluminescence.

chemical reaction, or heat generated in the contact area between two different materials during friction. In contrast, deformation luminescence arises from the mechanical deformation of the material itself and is independent of the medium causing the deformation. This type of luminescence can be further categorized into elástico ML (EML), plástico ML (PML), and fracto ML (FML) according to the deformation threshold

triggering the luminescence. Among these, EML materials are particularly sought after due to their ability to perform reproducible and durable emission within the reversible deformation while avoiding the restrictions of tribological materials or conditions that are inherent to triboluminescence. Therefore, much effort has been denoted to develop EML materials over the past two decades, particularly after Xu *et al.* first reported two EML materials of $\text{Sr}_3\text{Al}_2\text{O}_6:\text{Eu,Dy}$ and ZnS:Mn at the end of the last century^[22,23]. A large assortment of EML materials, such as sulfides, oxides, oxysulfides, silicates, aluminates, and fluorides, have been reported. These materials can emit intense brightness of $> 100 \text{ cd/m}^2$ and tunable spectrum ranging from ultraviolet to near-infrared wavelength, enabled by element doping or substitution, defect control, heterojunction construction, and anion mixture, among others^[24-27]. Additionally, innovative supportive strategies, including ion irradiation^[28], modification of matrix^[19], integration with FET^[29], and combination with fluorescent additive^[30,31], have been proposed to augment the ML intensity, boost the force-to-light conversion, lower the detection limitation, and manipulate the ML color, respectively. Meanwhile, the EML intensity generally exhibits a linear relationship with the magnitude of the applied force, which is ideal for force-sensing calibration.

However, the linear relationship between intensity and force usually deviates in the initial and final phases of force application. This initial deviation can be attributed to the slow rates of carrier de-trapping, while the final deviation may be due to a limited number of residual trapped carriers^[6]. This issue has been tentatively tackled by regulating the trap distribution through doping engineering, which allows for a higher saturation threshold^[32,33]. Another issue is the sensitivity of the intensity-based signal of EML materials to the acquisition apparatus and environment, causing inaccuracy or even severe errors. As a remedy, a ratiometric detection scheme using the intensity ratio instead of the absolute intensity has recently been proposed to circumvent this problem^[34-36]. Most notably, most EML materials require pre-irradiation with ultraviolet light, which provides charge carriers to be stored in host traps. The trapped carriers are then released to excite ML when the material undergoes mechanical deformation. Once the trapped carriers are exhausted, these EML materials need to be re-irradiated for ML generation, posing a huge hurdle for their practical applications. In addition, these EML materials usually exhibit a long persistent afterglow, hindering the high-contrast recognition of ML signals. A select few EML materials, such as ZnS:Cu/Mn , ZnS-CaZnOS:Mn/Ln , and $\text{MgF}_2:\text{Mn}$, can emit light without any pre-irradiation. These are referred to as self-recovery EML materials^[37], and their afterglow lifetime is typically short, endowing the unambiguous recognition of ML signals. Although the afterglow can also be correlated to the trap distribution and has been addressed by doping or defect engineering^[32,38], the self-recovery mechanism of EML materials remains elusive. This lack of understanding contributes to the absence of a clear guideline for designing EML materials in a self-recovery manner.

ML MECHANISMS

The comprehension of the physics underlying the mechano-to-light conversion of ML materials is crucial for material innovations and practical applications. However, a universal explanation of this process is missing due to the complex interplay between mechanical actions and multiple light emissions when ML occurs. The ML mechanism has been phenomenologically or empirically attributed to dislocation motion, piezoelectric effect, or triboelectric effect^[39,40]. The dislocation model was first proposed by Chandra *et al.* in 1982. This model posits that various dislocation activities, such as unpinning, interaction, stripping, bending, and annihilation, may occur during the dislocation movement to release energy or ionize the color centers for the excitation of ML^[41,42]. Subsequently, it was observed that ML usually manifests in piezoelectric materials, whereas non-piezoelectric materials rarely exhibit ML. This led to the formulation of the piezoelectrification model and the piezoelectric-induced carrier de-trapping model. The former is applicable to the FML of piezoelectric crystals and organic compounds, wherein a strong electric field is produced at the newly fractured interfaces facilitated by the piezoelectrification effect. This results in the

dielectric breakdown of surrounding gases or direct excitation of the solids for ML^[43,44]. The piezoelectric-induced carrier de-trapping model is applicable to EML of trap-controlled materials, in which the piezoelectric potential generated upon elastic deformation decreases the trap depth, leading to the release of trapped carriers. Then, the nonradiative recombination of de-trapped carriers transfers the energy to the doped ions to facilitate ML^[45,46]. The ML of trap-controlled materials with a centrosymmetric structure has been related to the piezoelectric models as well, assuming a local dissymmetry in structure due to the doped ions or defects^[47,48]. However, this model encounters difficulty in explaining the dependence of ML of such materials on the friction materials or matrix. As such, a triboelectricity-induced carrier de-trapping model is proposed for ML of trap-controlled materials with a centrosymmetric structure^[49]. For the trap-independent ML materials with a centrosymmetric structure, especially for those that emit only when blended with a polymer matrix, models such as the triboelectricity-induced electron bombardment model^[50] and the contact electrification electron-cloud model^[51] have recently been established. In these models, triboelectrification permits a direct excitation-emission process within the luminescence centers.

All the aforementioned models have made significant contributions towards understanding the inherent mechanism of ML. However, none of them can completely explain all ML phenomena and definitively describe self-recovery behavior. Therefore, continued efforts, including both experimental and theoretical, are required for a deeper insight into ML. Recently, an experimental approach to the simultaneous detection of imposed pressure and spatiotemporal ML patterns within one single ZnS:Mn microparticle has been reported. This approach unveils the microscopic mechanism of ML in conjunction with nanoscale structural characterization^[52]. However, the ML pattern is limited to an optical resolution, and the simultaneous observation of optical and structural images is not achievable. This limitation could lead to confusion regarding critical information. A promising method for illuminating the physical foundation of ML could be the integration of an imager and spectrophotometer with high spatiotemporal resolution, such as transmission electron microscopy and cathodoluminescence detector. This would enable the *in-situ* recording of both optical and structural information simultaneously.

SYNTHESIS OF ML MATERIALS

The synthesis of ML materials is currently predominantly conducted through high-temperature solid-state reactions due to the high brightness and yield. This process involves calcining a mixture of raw materials at an elevated temperature of over 1,000 °C, resulting in a hard block that needs to be milled into ML particles for applications. However, these prepared ML particles exhibit irregular morphology, random size (ranging from sub-micrometer to tens of micrometer), and poor homogeneity. These attributes hinder their practical applications, such as pixelated sensing and display. Therefore, alternative strategies, including physical vapor deposition^[23,53,54], ultrasonic spray pyrolysis^[55], combustion synthesis^[56,57], melt-quenching approach^[58], solution synthesis^[26,59,60], suppressed dissolution approach^[61], and molten salt shielded sintering^[62,63], have been developed. For example, biphasic ZnS ML microparticles have been synthesized using an in-air molten salt shielded sintering method, eliminating the need for grinding [Figure 2A]^[63]. The stable colloidal solutions composed of ML nanoparticles as small as 20 nm in diameter have been prepared through a biomineral-inspired suppressed dissolution approach [Figure 2B]^[61]. The monodispersed ML nanocrystals of NaLuF₄:Tb³⁺ with a uniform size (~25 nm) and morphology have been demonstrated via the solution coprecipitation method [Figure 2C]^[60]. Despite these advancements, a comprehensive approach to synthesizing ML materials with controlled multidimensionality in terms of morphology, dimension, components, and structures has yet to be achieved. This is expected to be realized by a combination of suitable synthesis methods and post-treatment technologies such as annealing, irradiation, ion exchange, surface modification, acoustic cavitation, and molecular packing. For example, a series of metal sulfide nanorods with controlled morphology, components, and heterostructures has been demonstrated by the combination of solvothermal synthesis and cation-exchange reaction [Figure 2D]^[64].

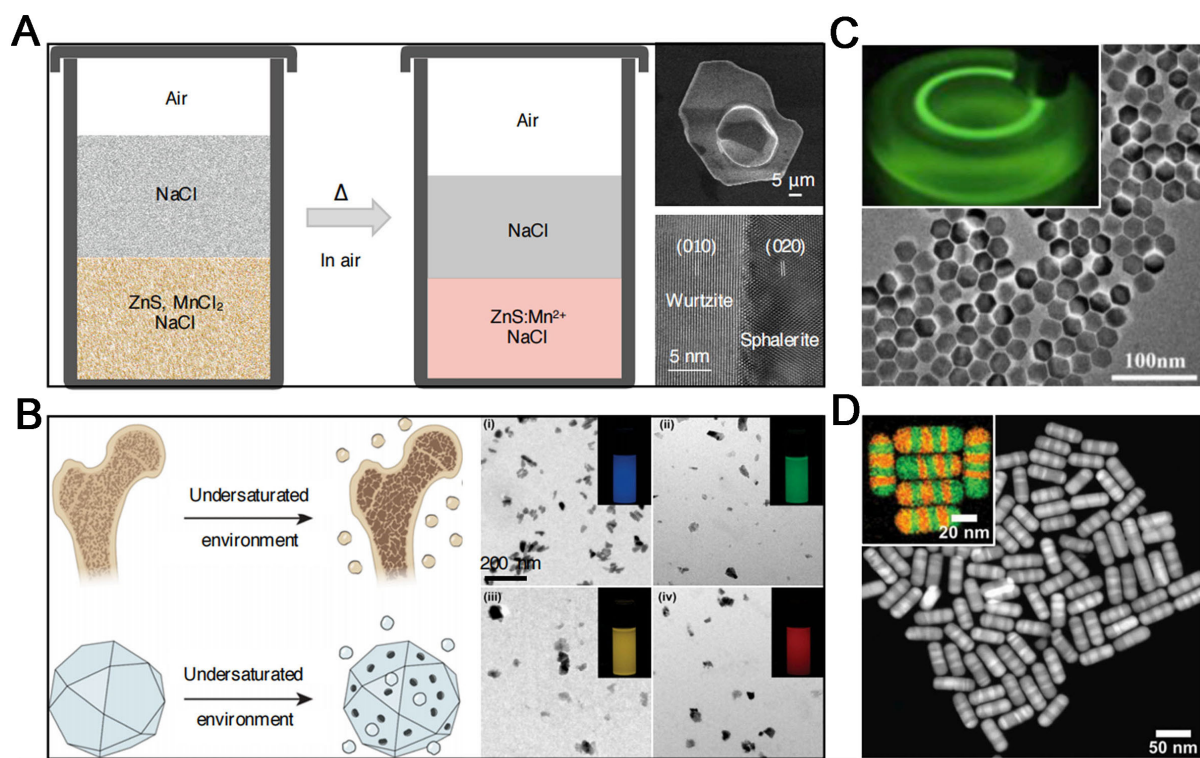


Figure 2. (A) ZnS ML microparticles synthesized utilizing the in-air molten salt shielded sintering method. Reprinted with permission from Ref.^[63]. Copyright 2022, Springer Nature; (B) Mechano-luminescent fluids produced by a biomimetic-inspired suppressed dissolution approach. Reprinted with permission from Ref.^[61]. Copyright 2022, American Chemical Society; (C) The monodispersed ML nanocrystals of NaLuF₄:Tb³⁺ prepared via a solution coprecipitation method. Reprinted with permission from Ref.^[60]. Copyright 2023, American Chemical Society; (D) The multicomponent metal sulfide nanorods synthesized by a combined strategy of solvothermal approach and cation-exchange reaction. Reprinted with permission from Ref.^[64]. Copyright 2020, AAAS. ML: Mechano-luminescence.

FABRICATION OF ML DEVICES

ML devices composed of ML particles and polymeric matrices (PDMS, acrylic resin, *etc.*) are generally fabricated by mold casting or coating. These conventional fabrication methods constrain the ML devices to simple geometries, which restricts their application scenarios and decreases their mechano-to-light efficiency. Recent advancements in 3D printing technologies provide a paradigm-shifting scheme for the construction of ML devices with intricate design and complex architecture^[19,60,65-67]. For example, Patel *et al.* reported an anisotropic ML device fabricated by one-step direct write 3D printing, showing different ML patterns in response to stresses from different directions [Figure 3A]^[65]. Zhao *et al.* fabricated a hierarchical ML device with programmed luminescent behaviors using extrusion-based 3D printing and envisioned that more colors and functions could be integrated into one device by expanding this strategy to a multi-nozzle 3D printing technique [Figure 3B]^[66]. In addition, vat photopolymerization 3D printing has been used to manufacture ML structures, enabling a more intricate and customizable geometry [Figure 3C]^[68]. However, these ML devices suffer from insufficient spatial resolution, exceeding 100 μm, which is far from the resolution limit of the used instrument. This issue is probably caused by the large dimension of the used ML particles, with a size distribution of several to tens of micrometers. The use of ML nanomaterials could solve this problem. For example, the 3D nanoprinting of a ML structure with a spatial resolution of ~2 μm has been realized by utilizing the NaLuF₄:Tb³⁺ ML nanocrystals [Figure 3D]^[60].

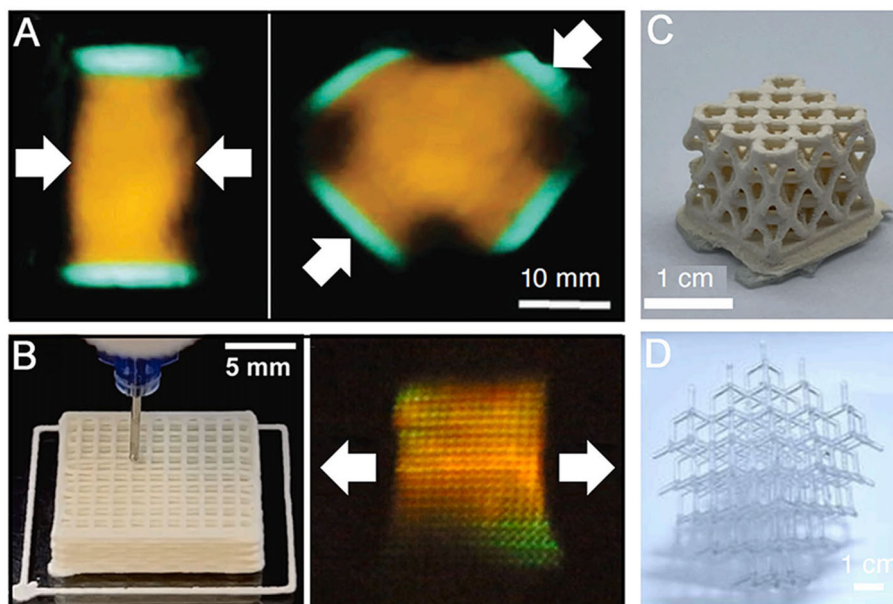


Figure 3. (A) An anisotropic ML device fabricated by one-step direct write 3D printing. Reprinted with permission from Ref.^[65]. Copyright 2018, Royal Society of Chemistry; (B) A hierarchical ML device with programmed luminescent behaviors prepared via extrusion-based 3D printing. Reprinted with permission from Ref.^[66]. Copyright 2022, Elsevier; (C) The ML structure fabricated via vat photopolymerization 3D printing. Reprinted with permission from Ref.^[68]. Copyright 2023, Elsevier; (D) The ML structure prepared via 3D nanoprinting. Reprinted with permission from Ref.^[60]. Copyright 2023, American Chemical Society. ML: Mechanoluminescence.

CONCLUSION

In summary, ML materials represent a captivating frontier with the potential to revolutionize the landscape of soft devices by enabling them to convert mechanical deformation into a captivating visual response. This transformative capability not only unlocks new realms of scientific exploration but also propels us towards pioneering applications across a wide spectrum of fields, including sensing, displays, and human-machine interactions. Nonetheless, it is crucial to acknowledge that this cutting-edge research area remains in its infancy, with a myriad of challenges awaiting diligent exploration. These challenges span a broad spectrum, from the intricacies of material synthesis to the mastery of material properties and the comprehensive understanding of the mechanisms underpinning mechanoluminescence. Moreover, the art of device fabrication is yet to be perfected, standing as a hurdle on the path forward as well.

In addition, the potential synergy between ML materials and other emerging soft materials, such as hydrogels, dielectric elastomers, and functional polymers, remains largely unexplored. This interplay between different soft materials promises a harmonious fusion of capabilities, opening up new vistas of possibilities in diverse applications. For instance, soft actuators with visual sensory feedback enabled by the conjunction of ML materials with dielectric elastomers or programmed devices that seamlessly combine ML materials with shape-memory polymers.

These aforementioned issues call for interdisciplinary collaboration. Scientists, engineers, chemists, physicists, and material scientists must unite their efforts to surmount these obstacles and usher ML materials into their full potential.

DECLARATIONS

Authors' contributions

Wrote the original draft: Wang C, Zhu D

Supervised, reviewed, and revised the manuscript: Wang C, Hu H, Peng D, Dong L, Zhu D

Availability of data and materials

Not applicable.

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Conflicts of interest

All authors declared that there are no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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