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Composite Materials COMMENTARY

Flexible 2D-2D composite film for magnetoelectric sensing

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Nanocomposites have long been celebrated for surpassing the intrinsic limitations of polymers. They achieve dramatic improvements in mechanical, electrical, thermal, and optical properties, even with low filler loadings. The key to these enhancements lies in dimensionality (denoted as D). As Osami Kamigaito defined in 1994, when one or more dimensions of a filler are confined below 100 nm, the resulting composite exhibits properties vastly different from those of either constituent.[1] Decades of research have shown that 0D nanoparticles, 1D nanowires, and 2D nanosheets each offer distinct advantages. Among these, 2D fillers stand out for their exceptional surface area, atomic-scale thickness (Figure 1A), and ability to form strong interfacial interactions, enabling performance gains that bulk or microscale fillers cannot match.

WHY 2D–2D LAMINATE CONFIGURATIONS ARE SPECIAL

Random dispersions of nanofillers in polymers often lead to interfacial disorder, high energy losses, and poor structural control. In contrast, 2D-2D laminate configurations—where 2D inorganic nanosheets are aligned within laminated 2D polymer matrices—offer an elegant strategy to maximize interfacial coupling. Such arrangements create submolecularly (and even atomically) flat interfaces with vast contact areas. This allows efficient transfer of strain, charge, and polarization across phases. In the context of magnetoelectric (ME) composites, where performance depends on the interaction between magnetic order (spin alignment) and electric polarization (charge displacement), the laminate structure is particularly advantageous. This architecture effectively addresses the interfacial mismatches and rough boundaries that plague traditional particle-based composites, unlocking more reliable and stronger coupling. However, the construction of ME composites with highly ordered 2D-2D architectures faces two critical challenges: perfectly aligning nanosheets to prevent random stacking, and establishing robust interfacial interactions for efficient transfer of functional responses in high-performance flexible ME devices.

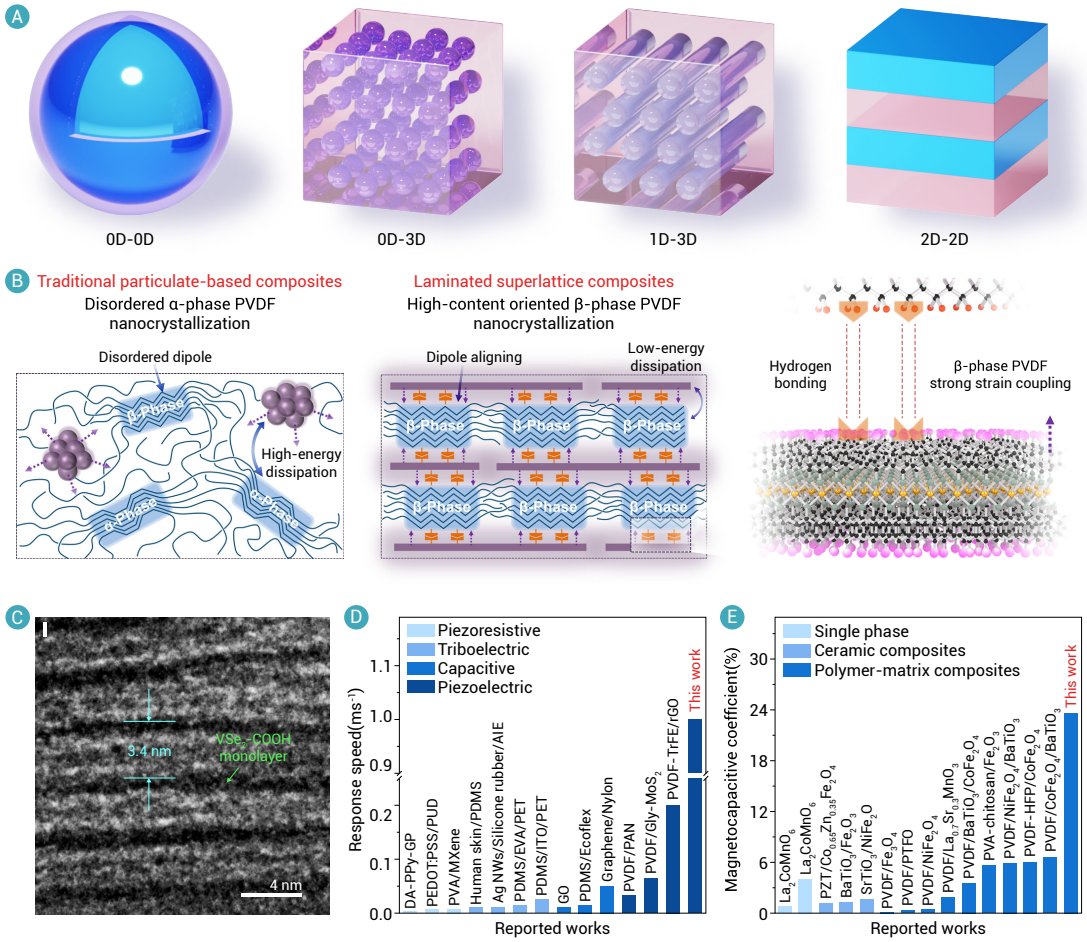

Figure 1. Interfacial cocrystallization strategy to fabricate PVDF-VSe_2-COOH magnetoelectric (ME) composite and its ME properties (A) Schematic diagram of composite dimensional architectures. (B) Schematic diagram of traditional particulate ME composites versus laminated superlattice composites. The distinction lies in the disordered matrix (α-phase) and random filler distribution of traditional composites, in contrast to the ordered/ferroelectric matrix (β-phase) and the aligned filler arrangement that forms a superlattice in laminated superlattice composites. (C) High-resolution cross-sectional transmission electron microscopy image of PVDF-VSe_2-COOH (25 wt%) film. (D) Comparison of the piezoelectric response speed of prepared PVDF-VSe_2-COOH (25 wt%) film with other flexible force sensors, where response time was converted to response speed for intuitive demonstration of piezoelectric efficiency. (E) Comparison of the ME coupling coefficient of PVDF-VSe_2-COOH film with other reported materials.

THE PROMISE OF MONOLAYER VSe_2 AS A MAGNETIC 2D FILLER

The discovery of intrinsic ferromagnetism in 2D materials has opened new possibilities for composite design.[2] Vanadium diselenide (VSe_2), a transition metal dichalcogenide, is especially intriguing: when thinned down to the monolayer limit, it exhibits robust room-temperature ferromagnetism despite its van der Waals layered nature.[3] Unlike many other 2D magnets (e.g., CrI_3), VSe_2 offers the crucial advantages of solution processability, environmental stability, and a functionalizable surface. This rare combination of ferromagnetism, chemical tunability, and ultrathin morphology makes monolayer VSe_2 an ideal building block for ME composites. Its atomically flat, dangling-bond-free surfaces provide pristine interfaces. While surface functionalization strategies—such as diazonium modification—can tailor interfacial chemistry to form ordered nanoconfined structures, and establish a hydrogen bonding network with polymer chains, ultimately promoting crystallization and enhancing ferroic coupling. Integrating such magnetic 2D nanosheets with ferroelectric polymers offers a pathway toward flexible, high-performance ME devices that traditional ceramic composites cannot achieve due to brittleness and poor compatibility with soft substrates.

INTERFACIAL CO-CRYSTALLIZATION UNLOCKS RECORD MAGNETOELECTRIC PERFORMANCE

He et al. proposed a landmark demonstration of the construction of 2D-2D ME nanocomposites with highly ordered alignment of 2D nanosheets and strong interfacial interactions, which were enabled by an interfacial cocrystallization strategy.[4] They designed a flexible polymer-inorganic composite film by embedding diazonium-functionalized VSe_2 monolayers (VSe_2-COOH)[5] into a polyvinylidene fluoride (PVDF) matrix. The carboxyl groups grafted onto VSe_2 surfaces served dual roles, namely preventing nanosheet aggregation and providing a strong hydrogen bonding network (O-H···F) that directed PVDF chains into the electroactive β-phase (Figure 1B). Evidence from FTIR and XPS spectroscopy demonstrates the predominant presence of the β-crystalline phase and confirms the formation of interfacial hydrogen bonding, respectively. Furthermore, molecular dynamics simulations reveal that the electric field generated by these interfacial hydrogen bonds guides the directional alignment of PVDF chains, thereby promoting preferential crystallization into the β-phase. This interfacial co-crystallization resulted in an ultraflat hybrid structure with an exceptional orientation order (approximately 0.71, as measured via small-angle X-ray scattering), enabling the highly efficient transfer of strain energy and polarization across the two phases.

The results were striking. The composite (Figure 1C) achieved a β-phase content of up to 86%, with VSe_2 nanosheets retaining ferromagnetism and PVDF exhibiting robust ferroelectricity. The response speed dramatically decreased from over 10 ms to within 1 ms (Figure 1D), rivaling or even surpassing those of rigid ceramic-based ME materials. This ultrafast response is enabled by the continuous and well-aligned interfaces that facilitate rapid polarization switching under external stimuli. The resulting synergistic coupling yielded a record-breaking ME coefficient of 23.6% (Figure 1E) for flexible systems. The significant enhancement, alongside the well-preserved ferromagnetism of the VSe_2 nanosheets, stems from the interfacially directed crystallization prompted by the hydrogen-bonding network, which effectively guides the PVDF chains into the electroactive polar phase. In addition, the strong interfacial interaction and high alignment in the 2D-2D laminate structure endow the composite film with excellent flexibility and mechanical robustness, where the tensile strength and fracture toughness were improved six-fold and eight-fold compared to pristine PVDF, respectively.

TOWARD NEXT-GENERATION FLEXIBLE MAGNETOELECTRIC DEVICES

By pioneering an interface-driven design strategy that transcends the traditional "mix-and-composite" paradigm, the research team demonstrated how precise sub-molecular engineering of 2D-2D interfaces can simultaneously tailor lattice alignment and interfacial interaction force, the universal principle lies in replacing random physical adsorption with designed chemical recognition. Such control enables the realization of strong ME coupling, exceptional mechanical flexibility, and scalable fabrication through a simple yet efficient solution-based approach. Importantly, the concept of interfacial co-crystallization proposed here extends far beyond ME materials. It provides an inspiring framework for addressing long-standing challenges in triboelectric, thermoelectric, and other flexible energy-harvesting devices, where the interplay between structure and function remains a key bottleneck.

Looking ahead, this research sets a blueprint for miniaturized, multifunctional, and self-powered devices, ranging from wearable health monitors to implantable neural interfaces, where strong ferric coupling and mechanical compliance are expected to coexist. Future efforts shall aim to achieve atomic-level control of such 2D-2D configuration. Given the intrinsic advantages of van der Waals monolayer surface, further refinement at this scale is not only feasible but also promises to reveal emergent interfacial phenomena and nanoscale confinement effects beyond ME behavior. Additionally, precise intercalant manipulation-for example, inserting polymerizable monomers of functional molecules between 2D inorganic nanosheets-could open pathways to customized superlattice architectures and next generation functional devices. Finally, such well-controlled interfacial configurations offer a powerful platform for fundamental studies, enabling deeper insight into energy transfer mechanisms across interfaces. Understanding these processes will be crucial for advancing a wide spectrum of technologies, including flexible sensors, energy-harvesting and storage systems, and low-dimensional quantum devices.

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AUTHOR CONTRIBUTIONS

M. Wei, Q. Zheng and M. Wu co-wrote the manuscript.

DECLARATION OF INTERESTS

The authors declare no competing interests.