Irregular structure breakthrough thermoelectric ceiling in organic materials

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Waste heat, ubiquitous in industrial processes and daily life, holds significant potential for energy harvesting and carbon reduction. Thermoelectric (TE) materials have been justified as an effective solution for direct heat-electricity conversion. However, state-of-the-art TE performance is still dominated by inorganic semiconductors, whose brittleness, high density, potential toxicity, and processing complexity severely limit their application in flexible and wearable devices. In contrast, organic thermoelectric (OTE) materials offer advantages of lightweight, flexibility, environmental friendliness, and solution processability, providing a promising alternative for next-generation wearable power suppliers and personal temperature regulators.

The performance of TE materials is commonly evaluated using the figure of merit (zT), which is defined as:

$$zT = \frac{S^2\sigma}{\kappa}T = \frac{S^2\sigma}{\kappa_e + \kappa_l}T$$

where S, σ, κ, and T represent the Seebeck coefficient, electrical conductivity, thermal conductivity, and absolute temperature, respectively. κ consists of two contributions: the electronic (charge carrier) component (κ_e) and the lattice (phonon) component (κ_l). The term $S^2\sigma$ is defined as the power factor (PF), which quantifies the electrical power generation capability.

Research on OTE materials dates back to the late 20th century and has experienced rapid development over the past two decades, driven by

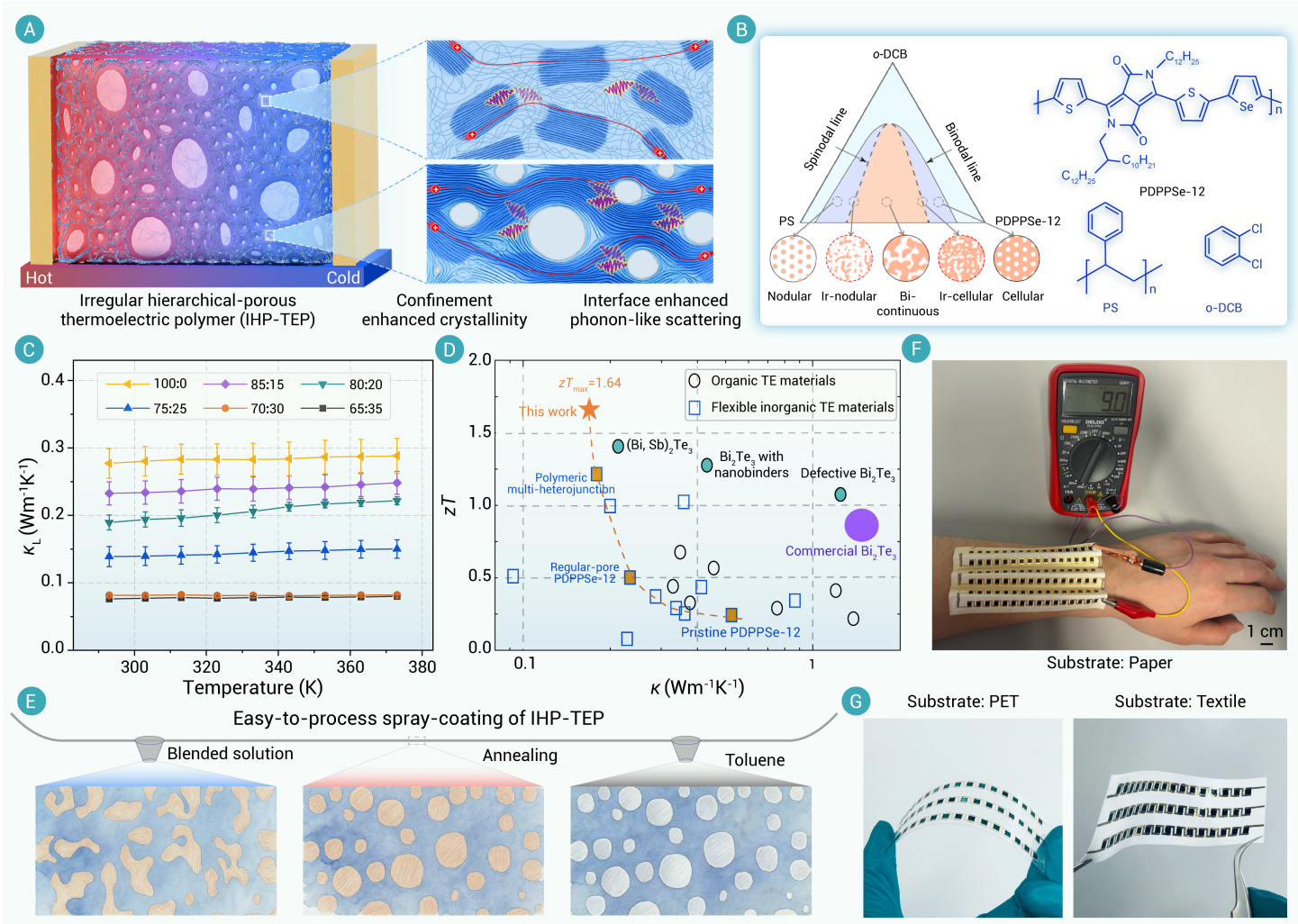

Figure 1. Schematic and TE Performance of IHP-TEP Films and Devices (A) Illustration of IHP-TEP films for TE energy conversion. Polymer confinement within pores enhances film crystallinity and charge transport, while the multiscale porous architecture increases phonon scattering. (B) (Top) Scanning electron microscopy (SEM) and (bottom) atomic force microscopy (AFM) images of the PDPPSe-12:PS blend (70:30, w/w). Insets illustrate magnified pore and throat features. Scale bars: 50 nm (SEM) and 2 μm (AFM). (C) Temperature-dependent lattice thermal conductivity (κ_l) of both pure and IHP-TEP undoped films. (D) Comparison of zT and κ of IHP-TEP films with representative OTE materials, flexible inorganic TE materials, and commercial bulk Bi_2Te_3 materials near room temperature. (E) Schematic of the spray-coating process for large-area device fabrication. (F) Photograph of a large-area IHP-TEP generator operating on an adult's arm. Scale bar: 1 cm. (G) Photographs of IHP-TEP thermoelectric generators (TEGs) on different flexible substrates.

advances in molecular design, doping strategies, and morphology control.[1] However, their performance still lags behind inorganic systems, with zT often below 0.5.[2] A long-standing challenge is the intrinsic coupling between electrical and thermal transport, which complicates the simultaneous achievement of high PF and low κ. Recent interfacial engineering approaches, including multilayer and heterojunction architectures, have further enhanced phonon scattering through interfacial molecular interactioins and pushed zT values to 1.28, approaching inorganic levels at near room temperature range.[3] However, these approaches largely rely on intrinsic molecular-level electronic structure tuning or introducing discrete planar interfaces, suffering from restricted tunability of the coupled transport parameters and increased synthesis or fabrication complexity. Last year, thermoelectric elastomers that combine high thermoelectric performance with excellent mechanical resilience have been developed,[4] enabling new features and opportunities for conformally wearable electronics. Despite these advances, challenges related to scalability and further decoupling of these parameters remain.

To address these issues, Di et al.[5] report an unconventional yet highly effective strategy by designing irregular hierarchical-porous TE polymer (IHP-TEP) films with fine-tuned phase separation and confinement-enhanced crystallization (Figure 1A). By employing a solution-processable phase separation method based on the Flory-Huggins theory, they combine selenium-substituted diketopyrrolo[3,4-c]pyrrole polymer (PDPPSe-12) as the matrix and polystyrene (PS) as a sacrificial phase-separating reagent. This simple process involves blending, phase separation, and selective removal of PS. As a result, the final PDPPSe-12 film features a multiscale, irregular pore network spanning from sub-10 nm to micrometer scales (Figure 1B), enabling decoupled thermal and electrical transport. This strategy establishes a new performance benchmark with a record zT of 1.64 at 343 K. The central advance of this work lies on the introduction of an extrinsic structural engineering that operates across multiple scales, rather than relying on discrete or isolated interfaces. Notably, this method demonstrates strong generality across various conjugated polymer systems, including PDPPSe-12, PBTTT, and PDPP3T, all of which achieve a reduction in k of 55% ~ 72% and significant enhancements in zT values of over 80%. The IHP-TEP architecture exhibits a three-dimensional, omnidirectional network of curved interfaces and nanoscale throats, enabling broadband phonon scattering. Such a high degree of freedom in transport modulation is not accessible in conventional planar heterostructures or molecular-level tuning strategies.

From the thermal transport perspective, the randomly distributed, multiscale interfaces create diverse phonon scattering pathways, reducing κ_l by over 72% (0.08 $W{\cdot}m^{-1}{\cdot}K^{-1}$) with a minimum total κ of 0.16 $W{\cdot}m^{-1}{\cdot}K^{-1}$ (Figures 1C-D), thereby entering a low thermal conductivity region that traditional high efficiency inorganic TE materials (such as Bi_2Te_3) cannot reach. This suppression arises from enhanced boundary scattering, defect-induced scattering, and enhanced phonon-phonon scattering within nanoconfined domains. This demonstrates that disorder structures, when properly engineered, can outperform structural regularity in blocking heat transport.

More strikingly, unlike the conventional expectation that porosity degrades electrical transport, the porous IHP-TEP system instead promotes electrical conduction. During phase separation, nanoconfinement not only improves polymer crystallization but also fundamentally modifies charge transport pathways. The reduced π-π stacking distance and increased coherence length both enhance intermolecular orbital overlap, facilitating more efficient carrier delocalization. As a result, carrier mobility increases by ~52%. Together with the contribution of interfacial energy filtering at pore boundaries, the S is largely preserved, leading to a high PF of 772 $\mu W{\cdot}m^{-1}{\cdot}K^{-2}$. These results demonstrate that IHP-TEP films have broken the conventional trade-off between S and σ ($S \propto \ln(\sigma)$). A similar effect was also observed in our TE elastomer work.[4] Therefore, this work has found a more optimal balance along the S-σ trade-off, whilst simultaneously incorporating a sharp reduction in κ, ultimately achieving a leap in the zT value.

Equally important is the scalability of the structural design. The IHP-TEP fabrication is fully compatible with large-scale spray-coating techniques (Figure 1E), avoiding complex nanofabrication or multilayer assembly. This simple solution-processable strategy enables large-area film fabrication on flexible substrates (e.g., PET, paper, textiles) (Figures 1F-G). A paper-based TE generator (TEG) containing 108 TE pairs was fabricated and delivered an open-circuit voltage of 185 mV and a normalized power density of 1.28 $\mu W{\cdot}cm^{-2}{\cdot}K^{-2}$ at a hot plate temperature of 413 K, comparable to flexible inorganic counterparts. When attached to a human arm, the device generated a voltage of approximately 9.0 mV by harvesting body heat (Figure 1F). Moreover, the device maintains stable performance under mechanical deformation, such as bending and stretching, retaining 93% conductivity after 10^4 bending cycles, highlighting its potential to meet the practical requirements for wearable devices.

Despite the remarkable achievements, several potential issues warrant further investigation. First, the long-term structural stability of hierarchical pores remains unclear, particularly under thermal cycling or mechanical deformation, where pore coalescence or collapse may occur. Second, the high surface area of porous structures may increase susceptibility to environmental degradation, such as oxidation or moisture uptake, potentially affecting both electrical and thermal transport. Third, device-level reliability under prolonged operation, especially at elevated temperatures, remains to be systematically evaluated. In addition, although the strategy shows promising generality across different material systems, the extent of performance improvement may still be system-dependent, and further validation across a broader range of materials is necessary to fully establish its universality.

Overall, this work has pushed the decoupling of electrical and thermal transport to a new level. The IHP-TEP films have achieved a unique combination—ultralow k and enhanced electrical transport properties far exceeding those of conventional polymers. This multiscale structural engineering strategy sets a new pathway for OTE materials and narrows the long-standing performance gap between organic and inorganic systems.

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AUTHOR CONTRIBUTIONS

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DECLARATION OF INTERESTS

The authors declare no competing interests.