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Molecular-Scale Carrier Localization Boosts High-Temperature Energy Storage and High-Entropy Energy Harvesting

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High-temperature insulation/energy storage applications boost the rapid development of polymer dielectrics such as polyimide (PI) with excellent thermal stability. However, PI exhibits significant leakage current due to intra- and inter-chain charge transfer complexes (CTCs), which seriously increases the risk of thermal runaway. Although single charge transfer inhibition strategies have been developed, the results remain unsatisfying due to the "short-board effect in carrier suppression." Herein, a "carrier localization" approach is introduced that synergistically regulates both intramolecular and intermolecular CT suppression. Thanks to molecular engineering and a directional intercalation structure, the all-organic polyimide dielectric materials with multidimensional carrier migration suppression achieve an exceptional breakdown strength of 878.9 kV mm^{-1} and U_d of 8.93 J cm^{-3} at 150°C and energy density of 5.64 J cm^{-3} at 200°C ($\eta > 90\%$), outperforming many reported systems. The all-organic polyimide dielectric materials also enable to possess high charge density ($215 \mu\text{C m}^{-2}$) when integrated into a self-excited triboelectric nanogenerator, which is highly desirable for the harvesting of low-frequency, irregular mechanical energy from a high-entropy environment. Collectively, this work provides a solid foundation for advanced energy storage and conversion applications.

1. Introduction

As the advanced energy systems and electrical equipment evolve toward high capacity/power and miniaturization, high-performance polymer dielectric-based capacitors are increasingly and urgently needed.^[1–4] Besides, the sustainable energy supplies through energy harvesters such as Triboelectric nanogenerator (TENG) also become increasingly significant to address the distributed micro-nano energy issues, where tribo-dielectric materials are the fundamental and core component.^[5] Normally, the film capacitor and TENG possess similar physical models and operating conditions. Specifically, under high-temperature and high-electric-field conditions, the free charge density in dielectric materials dramatically increases and gives an unavoidable rise to conduction loss, therefore seriously degrading the charge storage performance of dielectrics.^[6,7] Moreover, the carrier accumulation caused by localized electric field concentration augments the risk of breakdown and catastrophic failure of the device.^[8–11] Therefore, we are facing a scientific challenge to

resolve the issues on how to modulate the key parameters, such as conductive/dielectric loss, breakdown strength, as well as high energy density in film dielectrics toward high temperature energy storage and energy harvesting applications.

Polymers with high glass transition temperatures (T_g), including polyetheretherketone (PEEK), polycarbonate (PC), and polyimide (PI) have been considered as promising candidates for high-temperature dielectric materials.^[12–14] Particularly, the PI is featured with a highly customizable structural design, which has attracted extensive research attention both from academia and industry.^[15] Although the conjugated aromatic structure within the molecular backbone of traditional PI imparts its excellent thermal stability, this conjugation in return reduces the bandgap ($<3.2 \text{ eV}$), bringing about the formation of charge transfer complexes (CTCs) under high-temperature and high-electric-field conditions.^[16–20] These CTCs further induce intramolecular and intermolecular charge transfer (CT) effects, thereby enhancing the excitation and transport of thermal charge carriers.^[21–28]

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