



Novel high-entropy polymers with superior capacitive energy storage

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High-entropy materials are highly attractive due to their unique structural characteristics and exceptional properties which find various promising applications such as energy technology, electronics, catalysis, and biomedicine [1–3]. Current high-entropy materials essentially involve alloys and ceramics, while “high-entropy polymers have been rarely reported” [4], as commented by Prof. Yeh JW who coined the term of high-entropy alloys in 2004 [1]. Initial efforts to induce entropy stabilization by blending multiple polymers were unsuccessful, primarily due to the lack of molecular engineering of the chain structure in these blends [4]. Moreover, none of previous works explicitly defined the high-entropy state with the proof showing

that configurational entropy ΔS_{conf} exceeds $1.5R$ (R is the gas constant) like alloys and ceramics [1–3]. Therefore, entropy-stabilized polymers remain largely unexplored, despite the inherent disorder offering vast potential for regulation at the molecular level.

Recently, a joint research team led by Prof. Huamin Zhou and Prof. Yang Liu from Huazhong University of Science and Technology and Prof. Qing Wang from Penn State University [5] launched a pioneer study in *Nature Materials*, in which high-entropy polymers are demonstrated via low-dose proton irradiation for the first time which overcomes the long-standing challenge in relaxor ferroelectric polymers for capacitive appli-

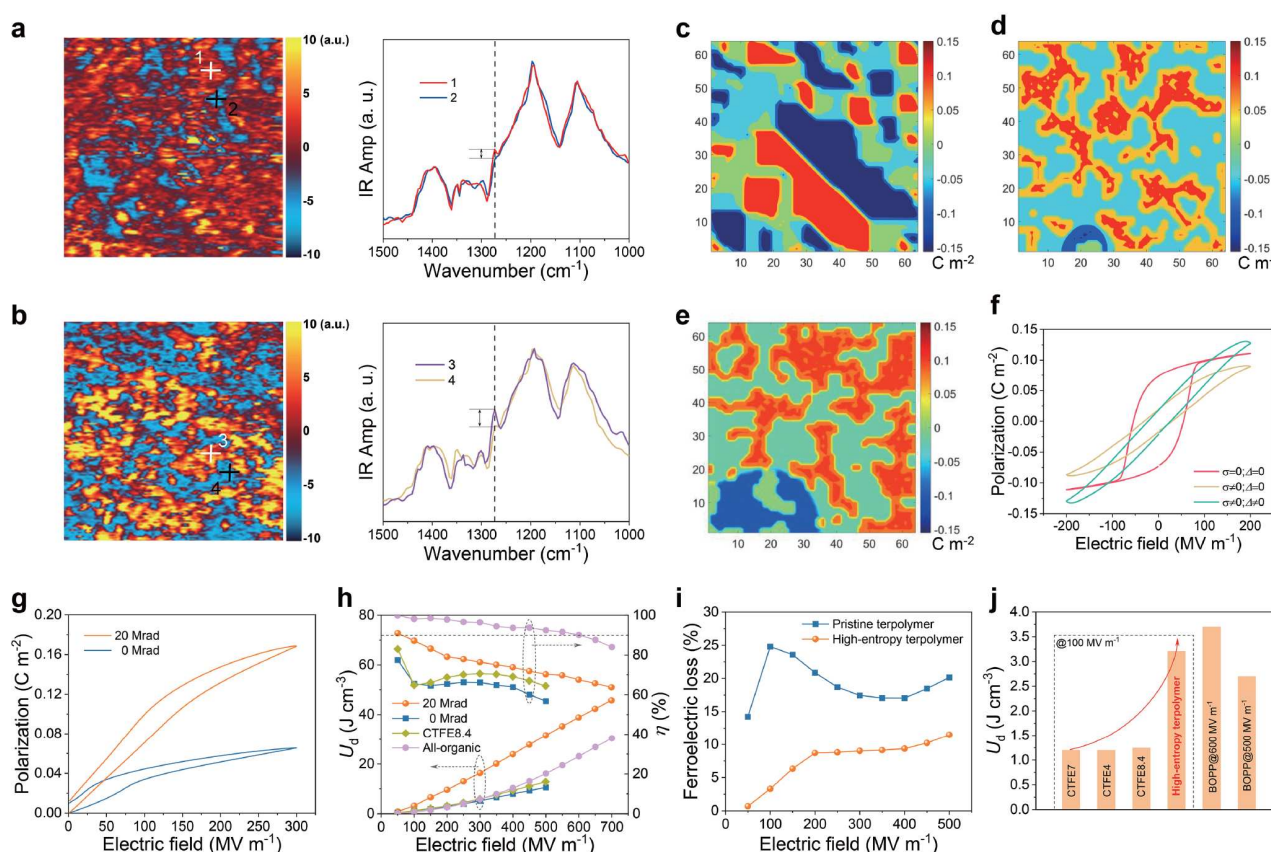


Figure 1 (a) Atomic force microscopy-infrared (AFM-IR) spectroscopy on pristine polymers. (b) AFM-IR characterization of high-entropy polymers. The size of the images is $1\ \mu\text{m} \times 1\ \mu\text{m}$. The local infrared data from spots 1–4 (right panels), as indicated in the chemical maps. (c–e) Phase-field simulations results considering different variances of random field (σ) and diffused Curie temperature (Δ): $\sigma = 0$; $\Delta = 0$ (c); $\sigma = 88\ \text{MV m}^{-1}$; $\Delta = 0$ (d); $\sigma = 88\ \text{MV m}^{-1}$; $\Delta = 3.6\ \text{K}$ (e). (f) Polarization-electric field loop results. (g) Unipolar polarization-electric field loops. (h) Comparison of ferroelectric loss between pristine and high-entropy polymers. (i) Discharged energy density U_d and charge/discharge efficiency η . (j) Comparison of U_d .

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cations. Li *et al.* [5] proposed a new approach to define the high-entropy state in polymers through the relation $\Delta S_{\text{conf}} = -R \sum_{i=1}^n c_i \ln c_i$, where c_i is the molar fraction of one chemical bond in different comonomers and n is the species of chemical bonds. The high entropy state can be achieved in polymers if $\Delta S_{\text{conf}} \geq 1.5R$, just like alloys and ceramics. To realize the high-entropy state in polymers, it generally requires both more than five types of chemical bonds with appropriate molar fractions of each bond. By incorporating a single chemical bond such as C=C double bond, ΔS_{conf} is well below $1.5R$, indicating the absence of a high-entropy state. Therefore, the definition of high-entropy state in polymers is provided for the first time which represents a milestone in the field of high-entropy materials.

Based on this definition, Li *et al.* further show high-entropy design through proton irradiation, bringing various chemical bonds by irradiation-induced chemical reactions as evidenced from combined structural characterization (Fig. 1a, b) and phase-field simulations (Fig. 1c–e). They further use the high-entropy strategy to overcome the longstanding challenge in relaxor ferroelectric polymers for dielectric energy storage applications [5]. Despite high dielectric constant, relaxor ferroelectric polymers have long been hindered for capacitive energy storage due to the presence of large ferroelectric loss, early polarization saturation and reduced polarization [6]. In relaxor ferroelectric polymers, the ferroelectric loss arises mainly due to intercoupling between different local polar states which results in nonzero local ferroelectric-switching barriers. High-entropy design not only significantly smears the barriers leading to considerably reduced ferroelectric losses [7,8] but also delays the polarization saturation (Fig. 1g, h). Moreover, as irradiation introduces polar bonds with increased local dipole moments (Fig. 1a–f), the dielectric constant and polarization are found to be markedly enhanced, which is highly desired for developing high-energy density capacitors.

High-entropy state is achieved based on morphotropic phase with small energetic barriers [9] whereas low-dose irradiation triggers the phase transition into high-entropy phase. This is one main reason to retain high polarization in high-entropy state. As a result, through the high-entropy design, irradiated polymers exhibit an ultrahigh discharged energy density of 45.7 J cm^{-3} at a

high field of 700 MV m^{-1} , exceeding the previous results under the same electric field (Fig. 1i). Meanwhile, a markedly enhanced discharged energy density of 3.2 J cm^{-3} with a charge/discharge efficiency of 87% at a low field of 100 MV m^{-1} which is comparable to benchmark biaxially oriented polypropylene (BOPP) obtained under a much higher field of $500\text{--}600 \text{ MV m}^{-1}$ close to its dielectric breakdown field (Fig. 1j). In addition, to mitigate the conductive loss, a scalable sandwich structure was used to enable a high discharged energy density of 23.1 J cm^{-3} with an efficiency of over 90% at 600 MV m^{-1} (Fig. 1i).

In short, the authors propose a new definition of high-entropy polymer and utilize the high-entropy strategy to address the key limitations of relaxor ferroelectric polymers, including ferroelectric loss and early polarization saturation, while taking advantage of their high dielectric constants, for dielectric energy storage applications. Their work may lay the foundation for future explorations of high-entropy polymers with striking functional properties such as the electrocaloric effect.

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