

FERROELECTRICS

Intrinsically elastic polymer ferroelectric by precise slight cross-linking

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Ferroelectrics are an integral component of the modern world and are of importance in electronics, electronics, and biomedicine. However, their usage in emerging wearable electronics is limited by inelastic deformation. We developed intrinsically elastic ferroelectrics by combining ferroelectric response and elastic resilience into one material by slight cross-linking of plastic ferroelectric polymers. The precise slight cross-linking can realize the complex balance between crystallinity and resilience. Thus, we obtained an elastic ferroelectric with a stable ferroelectric response under mechanical deformation up to 70% strain. This elastic ferroelectric exerts potentials in applications related to wearable electronics, such as elastic ferroelectric sensors, information storage, and energy transduction.

Wearable electronics with stretchability should be sufficiently elastic to conform to biological tissues and accommodate body movements upon large (up to 50 to 80%) and frequent strains (1, 2). These requirements have gradually become fundamental features of materials for skin-like elastic electronics (3, 4). Recently, prototypes of wearable sensors and circuits based on intrinsically elastic conductors or semiconductors have been proposed (2, 5, 6). However, the elastic ferroelectric resulting from the elastification of ferroelectrics (FEs), which are crucial and promising basic materials for modern electronics (7–11), is still lagging behind that of their intrinsically elastic counterparts, impeding their application in emerging wearable devices.

FEs feature reversible spontaneous polarization and distinctive properties (FE response) that make them suitable for a wide range of applications, ranging from electronic to electric power and biomedical applications (12–16). However, the dominant polymer FEs that are based on poly(vinylidene difluoride) (PVDF) failed to recover after stress relieving though enabling large plastic deformation (17, 18), let alone the traditional oxide FEs that even experience brittle failures at the very beginning of stretching (19, 20). Consequently, developing a new FE for wearable electronics that overcomes this FE response–elastic recovery dilemma is critical but challenging.

Typically, the elastification of electronic materials can be realized by structural engineering of rigid materials with mechanical and geometrical designs (4), blending of functional materials into elastomers (2, 3), or intrinsic elastification of functional materials (5, 6). Samples fabricated by structural engineering only enable a limited stretching range (less than the prestrained value) (21); they often require complicated fabrication techniques and show reduced device densities within an array (4, 21). The composites obtained by blending oxide FE particles into elastomers encounter extremely challenging poling problems (22–25). Indeed, in these composites, the FE domains of each oxide particle are orientated randomly, which requires effective poling (22, 23). However, as the electric field mainly partitions in the low-conductive elastomer phase, a very high poling field for the oxide FE particles is needed at the high risk of dielectric and electromechanical breakdown of the composites (24, 25). Therefore, intrinsic elastification may be the only possible route to impart elastic resilience to FEs effectively. This process could enable the development of materials characterized concurrently with scalable production by solution, high device densities, and excellent strain endurance (5, 6).

In the past few years, chemical cross-linking has witnessed substantial development in the intrinsic elastification of semiconductors and conductors (5, 6). Nevertheless, from the perspective of the elastification of polymer FEs, the FE response–elastic recovery dilemma may still occur through conventional chemical cross-linking. In more detail, the ferroelectricity of PVDF-based polymer FEs originates from the orientation and polarization of FE domains in the crystalline region (8, 20, 26), which implies that an excellent FE response requires a high crystallinity (27–29). On the contrary, a medium or low crystallinity would be essential for outstanding elastic recovery of elastomers. Therefore, addressing the FE response–elastic

recovery dilemma of excellent elasticity high crystallinity in the elastification of FEs is a challenging (30, 31). By using poly(vinylidene fluoride–trifluoroethylene) [P(VDF-TrFE)] with a soft- and long-chain cross-linker, we successfully developed an intrinsic elastification strategy for establishing a stable network in elastic polymer FEs by precise slight cross-linking.

Design and synthesis of elastic polymer FEs

PVDF-based FEs are semi-crystalline polymers and ductile under strain with characteristic yielding (8). The relative slippage among molecular chains induced the necking deformation of polymers under strain (Fig. 1A, top panel). This behavior is an unrecoverable deformation as the stress is removed, with the stress–strain curve of a semi-crystalline polymer with a characteristic yielding point (8, 31). Thus, PVDF-based FEs exhibit good plasticity but poor elasticity (32, 33). To combine ferroelectricity and elasticity in one material, we used chemical cross-linking to convert a plastic deformation into an elastic one. To avoid the high Young's modulus resulting from general cross-linking, we developed a cross-linking strategy with low cross-linking densities (1 to 2% in this work), named the “slight cross-linking” methodology; a long- and soft-chain in the cross-linker acts as a plasticizer for a low modulus. Low cross-linking density (see table S1 for more details about the definition and fig. S1 for the effect of cross-linking density on the mechanical recovery) can impart elastic resilience (recoverable up to 125% strain) to the linear polymer FEs while maintaining high crystallinity for a good FE response. As a result, an elastic FE network obtained by slight cross-linking of linear-polymer PVDF-based FEs with long and soft chains was established for the simultaneous realization of good crystallinity and elastification (Fig. 1A, bottom panel). The elastic FEs that we developed have a FE response and elastic resilience, with an acceptable crystallinity decrease. Overall, this “slight cross-linking” methodology should be an effective solution to the FE response–elastic recovery dilemma.

Experimentally (Fig. 1B), P(VDF-TrFE) is liable to form the β crystalline phase by a solution process and, therefore, we used it as the linear FE polymer to obtain the slight cross-linking (8, 16, 28, 33). Compared with other compositions, we selected P(VDF-TrFE) 55/45 mol % for its low modulus and large elongation (34) (fig. S2). This enables elastification for wearable electronics while preserving the good crystallinity of P(VDF-TrFE). The block of polyethylene glycol (PEG) was selected as the soft chain because its immiscibility with PVDF enables cross-linking to occur mainly in the amorphous region, as a result of which the high β -phase content in P(VDF-TrFE) crystalline regions was preserved for excellent FE



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properties (35). Then, the vulcanization of fluororubbers was realized by cross-linking P(VDF-TrFE) and PEG diamine through imine bonds to prepare the elastic FE (36). Compared with other cross-linking methods, the use of diamines as cross-linkers has a high reactivity (37), which is ideally suited for the “slight cross-linking” methodology with the reactive group at a low concentration and can be completed only by heating (36, 38). Moreover, its mechanism is clear with a simple addition reaction to the polymer backbone (8, 39). By designing this method to develop an elastic polymer FE, we expected that the high β -phase content and the stable network would ensure both a FE response and elastic recovery.

Cross-linking and characterization of elastic polymer FEs

We dissolved P(VDF-TrFE) and PEG diamine in cyclohexanone. We then cast the solution into thick films or spin-coated it into thin films. After removing the solvent under vacuum, we obtained cross-linked P(VDF-TrFE) films through thermal cross-linking. As shown by the differential scanning calorimetry (DSC) curve of the P(VDF-TrFE) and PEG diamine blended film (Fig. 2A), the cross-linking reaction commences at 188°C and ends at 254°C. We found the highest crystallinity in the sample cross-linked at 240°C under vacuum by the DSC peaks with different thermal cross-linking temperatures (fig. S3); hence, we chose this as the cross-linking condition to finalize the transition from plastic to elastic FEs.

We confirmed the formation of the cross-linked structure by the broad peak at 1647 cm^{-1} (attributed to the imine bond C=N) in the Fourier-transform infrared (FT-IR) results (Fig. 2B) (36). The chemical valance of F and C elements remained almost unchanged because of the low cross-linking density, which was confirmed by x-ray absorption near-edge structure (XANES, fig. S4) results. All the amino groups in the cross-linker (PEG diamine) were reacted into imine groups, which we confirmed by the disappearance of the amino peak spectra and the existence of only the imine peak in the cross-linked P(VDF-TrFE) of XANES (fig. S5) and x-ray photoelectron spectroscopy (XPS, fig. S6) spectra. Therefore, we could calculate the cross-linking density from the amount of PEG diamine used in the preparation stage. In addition, the decrease in the intensity of the peaks around 1120 and 1174 cm^{-1} , which represented the TrFE unit marker (40) and the CF_2 antisymmetric stretch, respectively, suggests that the cross-linking dominantly occurred on the TrFE units (Fig. 2B, inset). We confirmed the good thermal stability of the cross-linked P(VDF-TrFE) by the onset degradation temperature above 300°C (as seen in the thermogravimetric analysis curve, fig. S7). The cross-linked P(VDF-TrFE) films cannot be dissolved in common organic solvents, such as dimethylformamide, acetone, isophorone, and cyclohexanone; however, they possessed a swelling behavior with a gel content of ~60% when exposed to these solvents (fig. S8 and table S2). The thermal and solvent resistanc-

es of the cross-linked P(VDF-TrFE) ensure its feasibility for standard electronic industry processing methods.

As expected, the crystalline phase in cross-linked P(VDF-TrFE) mostly retained the β -phase morphology, which was confirmed by the β -phase marker peaks in the FT-IR spectra at 840, 1091, and 1284 cm^{-1} (28, 40). We also confirmed the presence of the β phase by the 19.2° peak in the x-ray diffraction (XRD) pattern (fig. S9). We verified the presence of the 3/1-helical phase using the peaks at 507 cm^{-1} and 18.9° in the FT-IR and XRD spectra, respectively (28). The polar crystalline phase that we observed in the cross-linked P(VDF-TrFE) films ensures a large remanent polarization under the applied external electrical fields. However, compared with the pristine P(VDF-TrFE) film, the crystallinity of the cross-linked P(VDF-TrFE) films decreased as the cross-linking density increased. We confirmed this result by calculating the latent heat and melting enthalpy from DSC curves (figs. S10 and S11 and table S3) (28). Consequently, we achieved both low modulus and elasticity through decreased crystallinity and increased cross-linking density.

The pristine P(VDF-TrFE) film annealed at 135°C for 4 hours showed typical semi-crystalline polymer mechanical properties with a yield point at a strain of 11% and an elongation at break above 800% (Fig. 2C). However, after cross-linking, the elongation at break of films rapidly decreased, and the stress-strain curves gradually changed from showing plastic to elastic

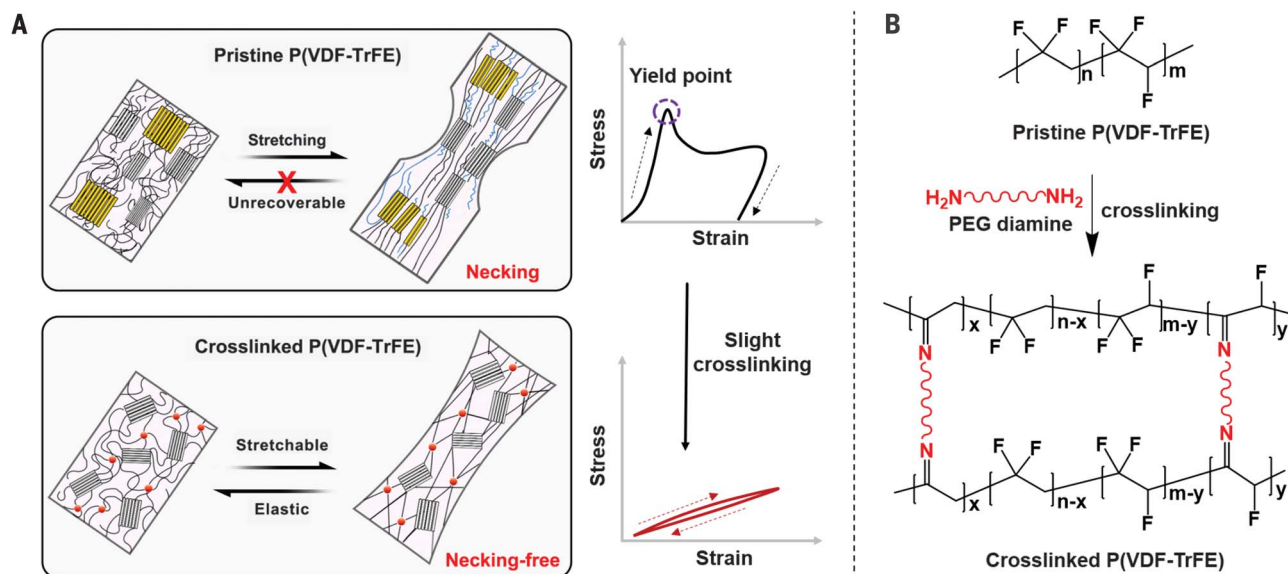


Fig. 1. Conception and synthesis of elastic FEs. (A) Schematics of the macro and molecular size changes and stress-strain curves of plastic (relative slippage among molecular chains leading to necking and unrecovered, top panel) and elastic (chemical cross-linking blocking the relative slippage among molecular chains and imparting good elastic resilience, bottom panel) deformations of polymer FEs under strain. (B) Synthesis of an elastic polymer FE by slight cross-linking.

deformations (Fig. 2C, inset). We used cross-linking densities ranging from 0.43 to 2.16% to evaluate the mechanical characteristics of cross-linked P(VDF-TrFE). Films with cross-linking densities above 1.44% exhibited the characteristics of elastomer stress-strain curves, i.e., low modulus without yielding (Fig. 2D). In the yielding-free samples, the cross-linked P(VDF-TrFE) with a cross-linking density of 1.44% possessed the highest degree of crystallinity and a suitable modulus; therefore, we selected this cross-linking density for all the

subsequent experiments. We observed the elastic recovery, mainly due to the change in entropy rather than energy (fig. S12), in the cyclic stress-strain curves of the cross-linked film under different strains ranging from 25 to 125% (10 cycles in Fig. 2E and >3000 cycles at a 40% strain level in fig. S13). Compared with the low resilience of the pristine P(VDF-TrFE) films (Fig. 2E, inset), these results confirm that the intrinsic elasticity of the polymer FEs was successfully realized by precise slight cross-linking of the linear polymer FEs.

Ferroelectricity of cross-linked P(VDF-TrFE)

We confirmed the ferroelectricity of the cross-linked P(VDF-TrFE) films by analysis of the temperature dependence of the dielectric constant ($\epsilon-T$ curve), polarization-electric field (P-E) loops, and piezoresponse force microscopy (PFM) results. In Fig. 3A, we observed a peak at $T_c = \sim 65^\circ\text{C}$ in the $\epsilon-T$ curve of cross-linked P(VDF-TrFE), as a feature of the FE-paraelectric phase transition ("Curie transition"), indicating that a β -like phase similar to that in pristine P(VDF-TrFE) is preserved (28).

We measured the P-E loops using a capacitor-type device with a Pt/cross-linked P(VDF-TrFE) (250 nm)/Pt/Si structure (fig. S14). The P-E loop expands gradually when the electric field increases above 120 MV/m and then saturates after the electric field exceeds 200 MV/m (Fig. 3B). The coercive field (E_c) of the elastic FE film is 60 MV/m, similar to that of the pristine P(VDF-TrFE) (55/45) film ($E_c \sim 59$ MV/m, fig. S15). The maximum (P_{max}) and remanent (P_r) polarization of the elastic ferroelectric film are 9.1 and 4.70 $\mu\text{C}/\text{cm}^2$, respectively, whereas those for the pristine P(VDF-TrFE) (55/45) film are 9.5 and 5.31 $\mu\text{C}/\text{cm}^2$, respectively. The polarization switching of cross-linked P(VDF-TrFE) can be cycled more than 10^6 times (fig. S16). We also measured the P-E loops of the cross-linked P(VDF-TrFE) at different frequencies (Fig. 3C). As the frequency decreases from 10 kHz to 500 Hz, P_r increases from 3.02 to 5.87 $\mu\text{C}/\text{cm}^2$, and the shape of the P-E loops becomes increasingly rectangular, indicating that the switching of the FE domain in the cross-linked P(VDF-TrFE) requires a longer time and more energy than that in the pristine material (fig. S17). The difference in the P-E loops of the cross-linked P(VDF-TrFE) films is attributed to the decreasing crystallinity and size of the crystals with the increasing cross-linking densities, compared with those in the pristine P(VDF-TrFE) films (fig. S18) (41). Indeed, the crystallinity of the cross-linked P(VDF-TrFE) films decreased to $38.36 \pm 1.76\%$; the FE phases also changed from the pure β phase to a mixture of β and 3/1-helical phases. Although the latter is considered a weak polar phase, it can be transformed into the former by poling under a high electric field at room temperature and, thus, contribute to overall polarization (42).

PFM is a powerful tool that can verify the ferroelectricity of a material by exploiting the existence of domains with different polarization orientations and local hysteretic domain switching triggered by electric fields (43–45). The ferroelectricity test of the elastic polymer FE was conducted by PFM using conductive tips on the cross-linked P(VDF-TrFE) film with a thickness of 250 nm deposited on a Pt/Si substrate. Box-in-box phase and amplitude patterns are obtained by applying a -30 V bias on a $12 \mu\text{m}$ by $12 \mu\text{m}$ zone and then a $+30$ V bias in the $8 \mu\text{m}$ by $8 \mu\text{m}$ -centered one (Fig. 3,

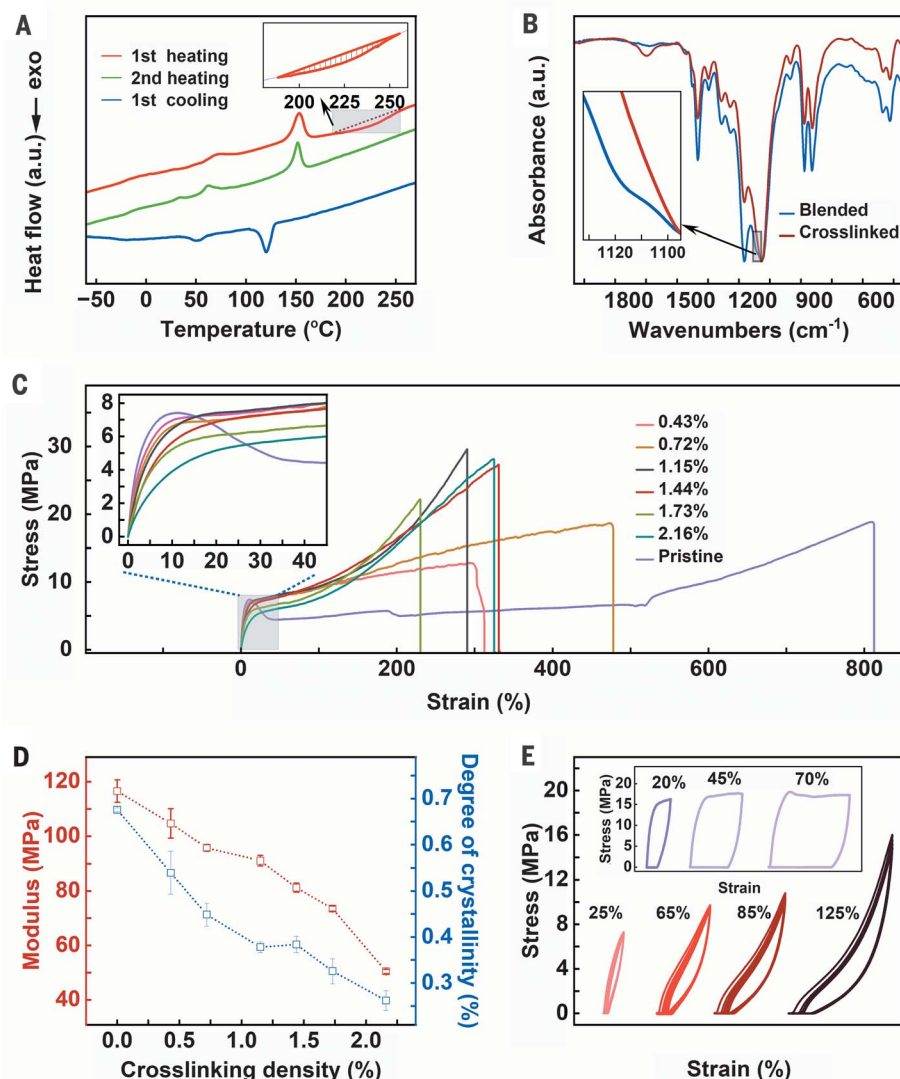


Fig. 2. Cross-linking characterization and mechanical properties of elastic FE films. (A) DSC curves of the P(VDF-TrFE)/PEG diamine blend indicating in situ cross-linking. (B) FT-IR spectra of blended and cross-linked P(VDF-TrFE)/PEG diamine (the peak at 1647 cm^{-1} from C=N bond confirms the occurrence of cross-linking). (C) Stress-strain curves of pristine and cross-linked P(VDF-TrFE) thick films at various cross-linking densities represented by different colors. (Inset) Transition from plasticity to elasticity is shown by the stress-strain curves below 45% strain presented in (C). (D) Modulus and degree of crystallinity of cross-linked P(VDF-TrFE) at various cross-linking densities ($n = 4$, where n is the number of samples that are used to generate statistical data). (E) Cyclic stress-strain curves of cross-linked P(VDF-TrFE) films with a cross-linking density of 1.44% and pristine P(VDF-TrFE) films (inset) under different strains (the first cycle was omitted owing to clamp sliding), indicating the excellent resilience of the cross-linked P(VDF-TrFE) films compared with the unrecoverable deformation of the pristine P(VDF-TrFE) films.

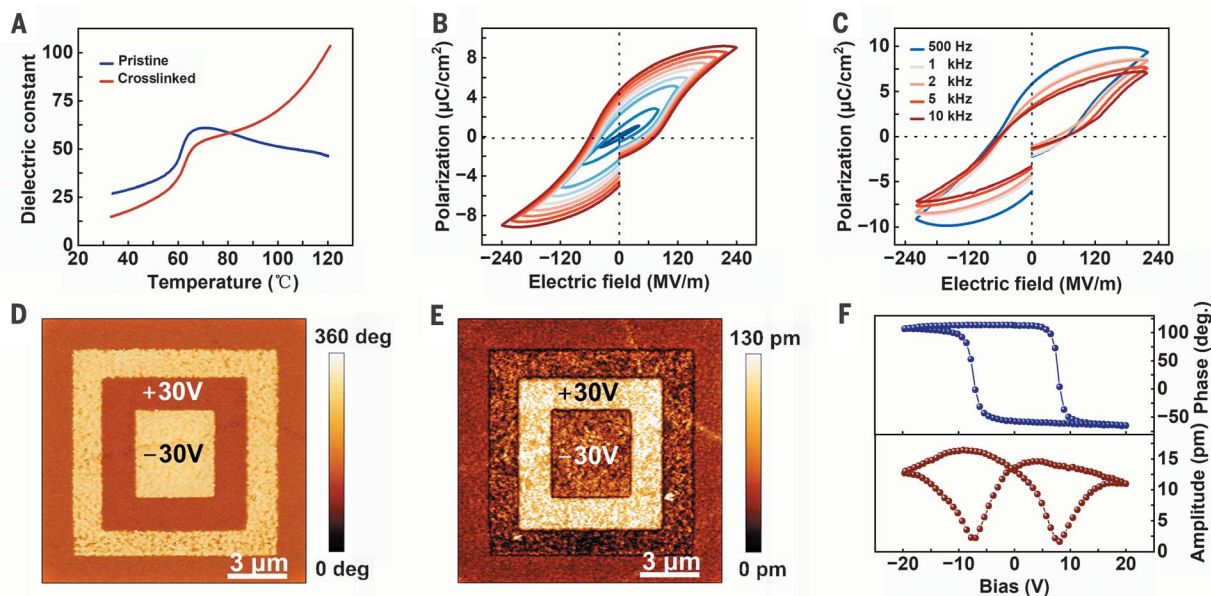


Fig. 3. FE response of cross-linked P(VDF-TrFE) thin films. (A) ϵ -T curve at 1 kHz, confirming that the cross-linked P(VDF-TrFE) maintained FE polymer behaviors. (B and C) P-E loops of Pt/cross-linked P(VDF-TrFE)/Pt under different electric fields at 1 kHz (B) and at different frequencies (C). (D and E) Phase (D) and amplitude (E) mapping of PFM. (F) Phase-voltage hysteresis and amplitude-voltage butterfly loop.

D and E). Our results show that the polarity of the FE domains in the crystalline region can be reversibly switched by applying voltage rather than the injected charges, as indicated by scanning Kelvin probe microscopy (SKPM) (fig. S19). Furthermore, a piezoelectric coefficient (d_{33}) of 11.3 pm/V was revealed by PFM (fig. S20). As exhibited by the hysteresis and butterfly loops (Fig. 3F), the local piezoresponse indicates a complete FE switching.

FE response of cross-linked P(VDF-TrFE) under strain

We prepared fully elastic capacitors with a liquid metal (gallium, Ga) as elastic electrodes using a sacrificial-layer microfabrication method (6) (fig. S21) to test the FE response of the elastic polymer FE under different applied strains and frequencies. We present the full elastic Ga/cross-linked P(VDF-TrFE)/Ga/PDMS device in Fig. 4A. Compared to a rigid device with Pt as electrodes, the P-E loops of our fully elastic device, without an applied strain and at different voltages, show more rectangular P-E hysteresis (fig. S22) with larger P_r and P_{max} values. Nevertheless, we observed higher E_c values, which may be attributed to the different thicknesses of the transition layer formed by different electrode materials. We observed the frequency-dependent behavior for the P-E response loops of the fully elastic device at different frequencies (fig. S23) that is similar to that of the Pt-electrode device (Fig. 3B). We concluded that the cross-linked P(VDF-TrFE) films in the fully elastic device also provide an equivalent FE response compared with those in the rigid device. We have therefore devel-

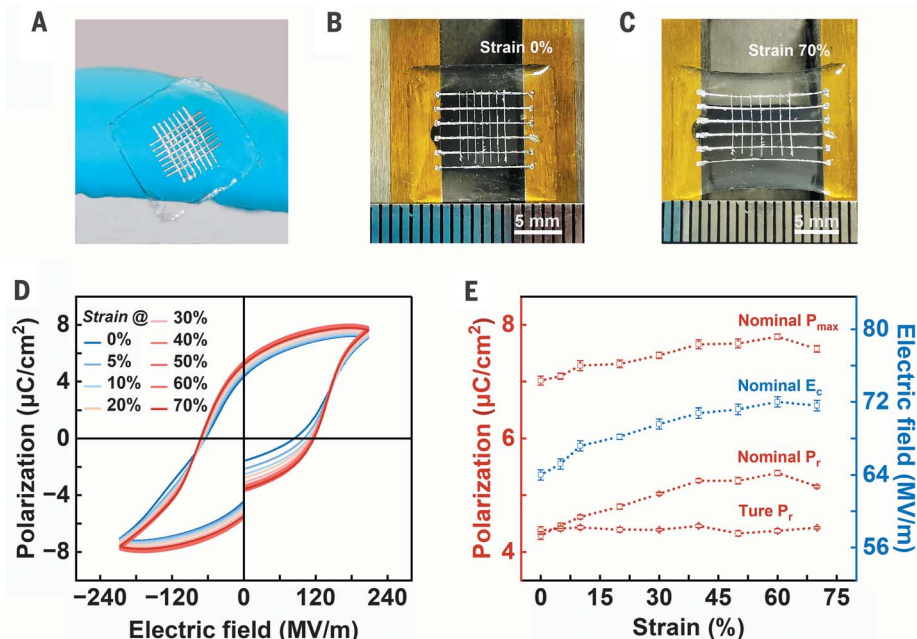


Fig. 4. FE response of elastic FEs under strain. (A to C) Full elastic device (A) in the stretching set-up under 0 (B) and 70% strain (C). (D) P-E loops at 1 kHz under strain from 0 to 70%. (E) Nominal and true P_r , P_{max} , and E_c under different strains ($n = 5$, where n is the number of P-E loops that are used to generate statistical data), suggesting that the cross-linked P(VDF-TrFE) possessed a stable FE response.

oped an effective method to prepare a fully elastic FE device that can be applied to other elastic FE materials.

We developed the stretching set-up to apply the strain for the FE response under strain (fig. S24). After uniaxial stretching, the full elastic device presented a rubber-like behavior with a Poisson's ratio of ~ 0.45 (e.g., 70% axial

strain and less than 35% transverse strain in our case) (Fig. 4, B and C). The electric fields and device areas without strain were taken as nominal values. We show P-E loops under strains ranging from 5 to 70% (figs. S25 to S32). As the strain increased, the degree of rectangularity of the P-E loops of the elastic FEs (Fig. 4D) gradually improved. At the same time, the values

of P_D , P_{\max} and E_c extracted from the P–E loops under different strains showed the same trend (Fig. 4E). To evaluate the effect of strain on the FE response, we performed a simulation to calculate the true values of the area (figs. S33 to S35), and the results were in accord with Eshelby's inclusion (46). The true values of P_D , deduced by changing strain values, during stretching from 0 to 70% remained almost constant, which implies that the FE response of our elastic ferroelectrics was not affected by the applied strain. After the stress was removed, the FE response remained nearly invariable (fig. S36).

We proposed a “slight cross-linking” methodology to develop elastic FEs. We successfully prepared an elastic FE by slightly cross-linking plastic polymeric FEs with soft chains into a stable FE network, which can be produced by solution processing and standard electronic industry methods. Our elastic polymer FE simultaneously showed a FE response and elastic recovery, even under strains up to 70%. The slight cross-linking is an effective way out of the FE response–elastic recovery dilemma for the elastification of polymer FEs. We believe that the realization of intrinsically elastic FEs can bridge the gap between FE materials and emerging wearable electronics, enabling broad potential applications such as wearable sensing, information storage, and the transduction and storage of energy.

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SUPPLEMENTARY MATERIALS

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Materials and Methods
Supplementary Text
Figs. S1 to S36
Tables S1 to S3

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Editor's summary

Ferroelectric materials change electrical properties in response to an electric field, which is a useful property for a range of applications. However, even polymer-based ferroelectrics tend to lack flexibility. Gao *et al.* added a small amount of cross linking to a ferroelectric poly(vinylidene difluoride) terpolymer, which dramatically improved its elasticity (see the Perspective by Zhang and Xiong). The material can be stretched repeatedly without degrading its ferroelectric properties, making it a potentially attractive strategy for developing wearable devices or for use in other applications. —Brent Grocholski

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