

Supplementary Information

Superior Electrostrictive Strain Achieved Under Low Electric Field in Relaxor Ferroelectric Polymers

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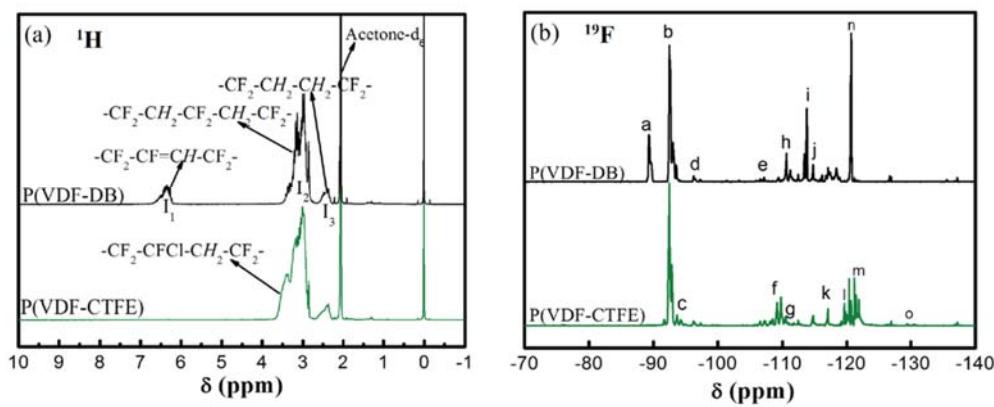


Fig. S1. a) ^1H NMR of P(VDF-CTFE) bearing 20 mol% CTFE and P(VDF-DB) synthesized from the fully dehydrochlorination of P(VDF-CTFE). b) ^{19}F NMR spectra of P(VDF-CTFE) with 20 mol% CTFE and P(VDF-DB) synthesized from the fully dehydrochlorination of P(VDF-CTFE).

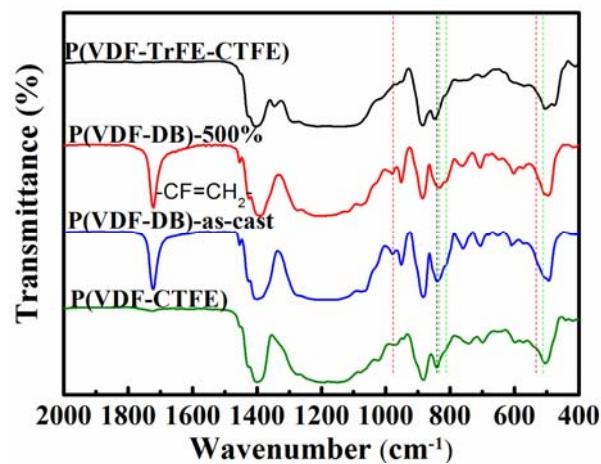


Fig. S2. FTIR spectra of P(VDF-TrFE-CTFE), P(VDF-DB)-500, P(VDF-DB)-as-cast and P(VDF-CTFE).

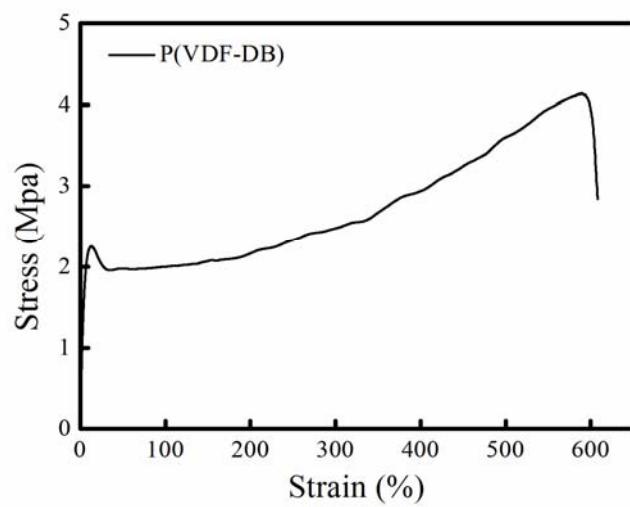


Fig. S3. Stress-strain curve of P(VDF-DB).

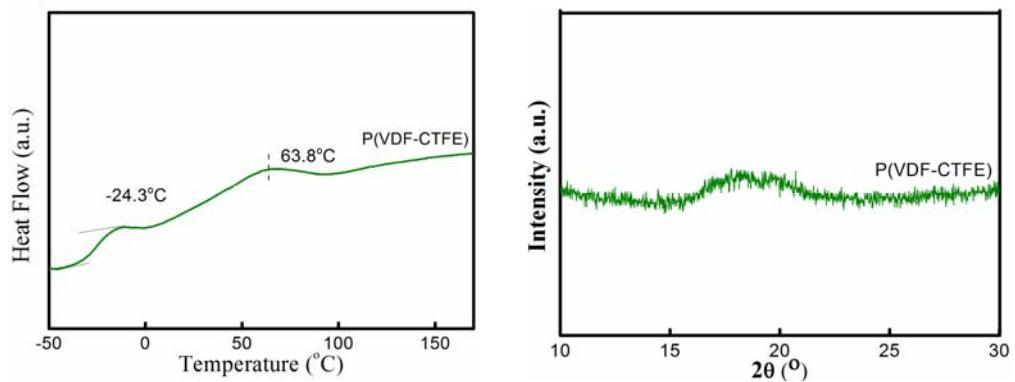


Fig. S4. DSC and XRD of P(VDF-CTFE).

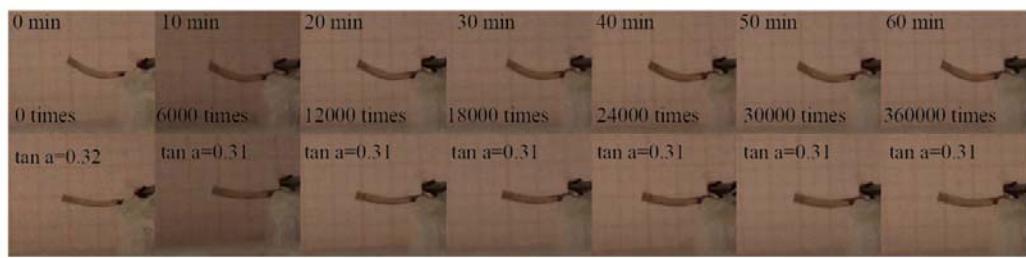


Fig. S5. The fatigue behavior of electrostrictive strain of P(VDF-DB)-500 with field.

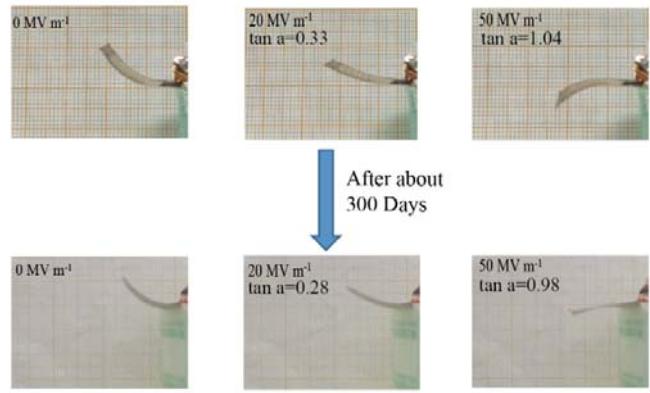


Fig. S6 The fatigue behavior of electrostrictive strain of P(VDF-DB)-500 with time.

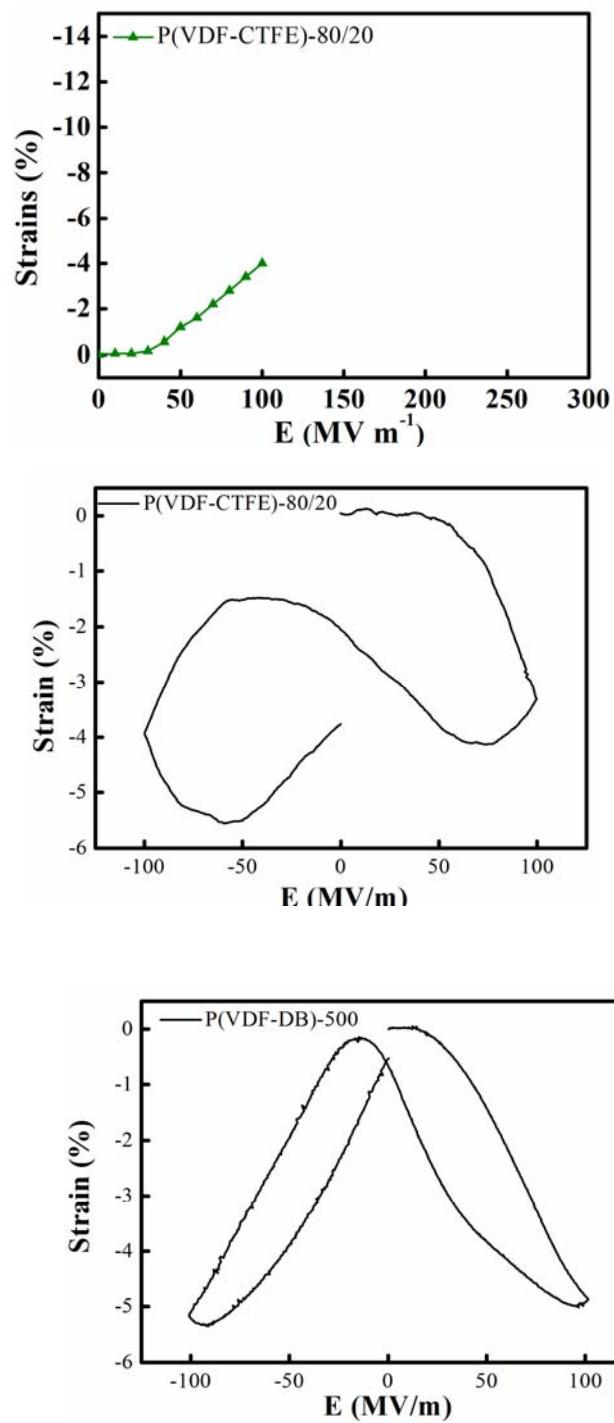


Fig. S7. Strain of P(VDF-CTFE) and P(VDF-DB)-500 under the applied field

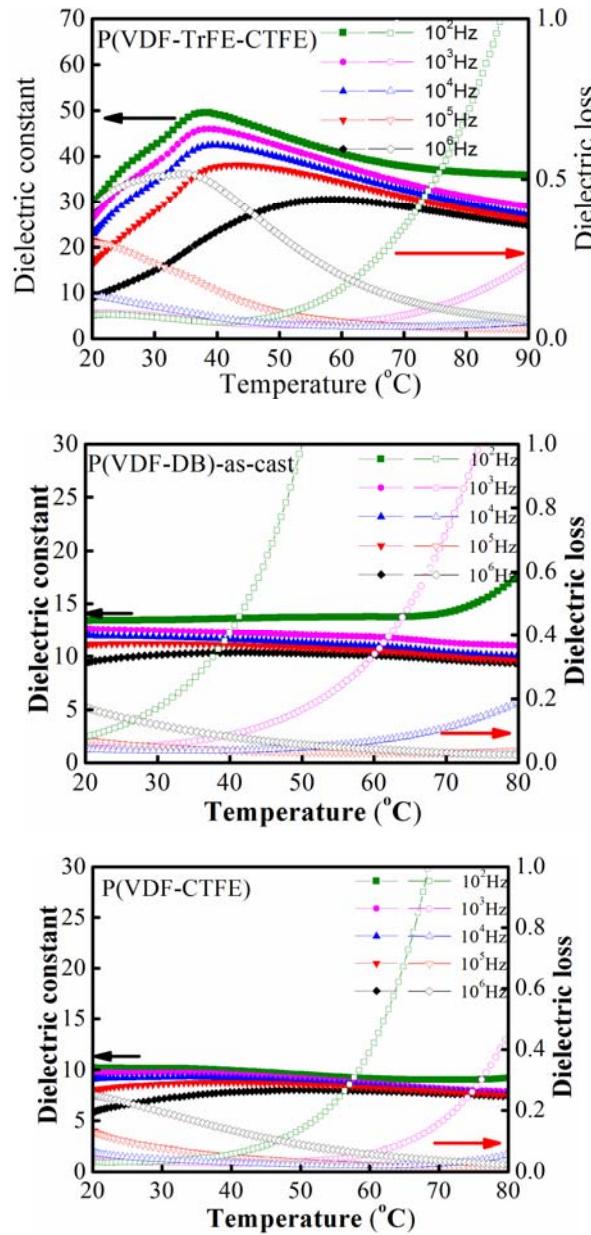


Fig. S8. Dielectric spectra of P(VDF-TrFE-CTFE), P(VDF-DB)-as-cast and P(VDF-CTFE).

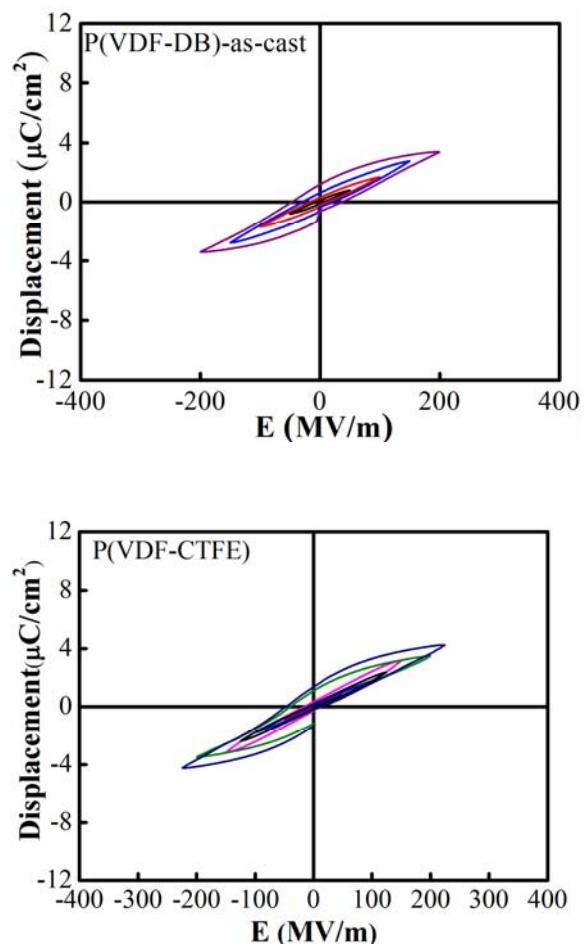


Fig. S9. D-E loops of P(VDF-DB)-as-cast and P(VDF-CTFE)

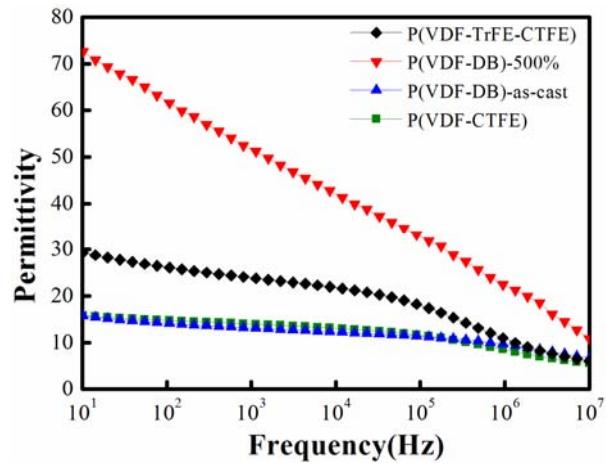


Fig. S10 The frequency dependent permittivity of P(VDF-DB)-as-cast, P(VDF-CTFE), P(VDF-TrFE-CTFE) and P(VDF-DB)-500.

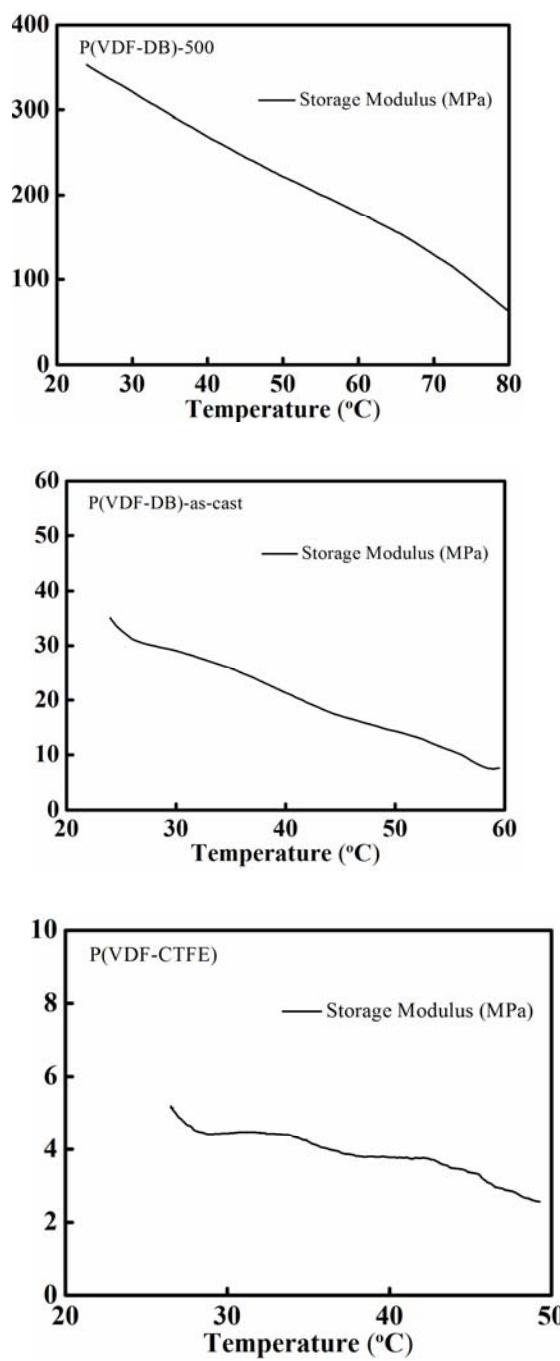


Fig. S11. DMA of P(VDF-CTFE)-500, P(VDF-DB)-as-cast and P(VDF-CTFE).

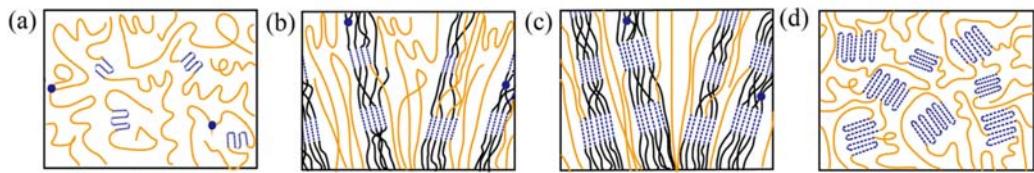


Fig. S12. Schematic of lamellae in crystalline and chains in a) P(VDF-DB)-as-cast. b) P(VDF-DB)-300. c) P(VDF-DB)-500 and d) P(VDF-TrFE-CTFE).

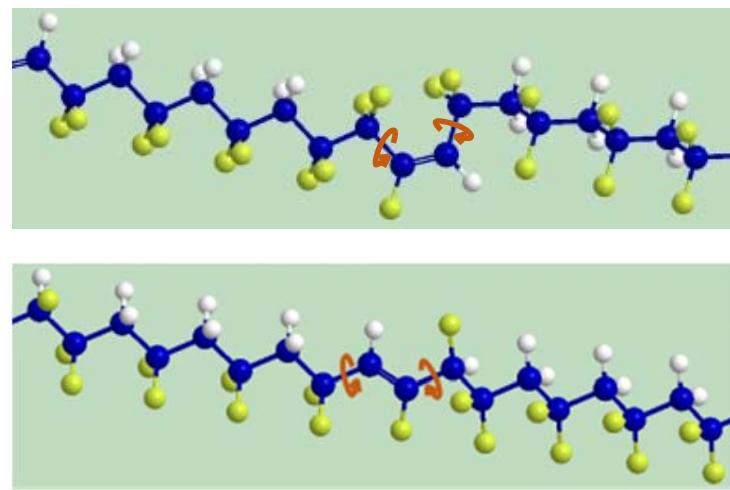


Fig. S13. Easily rotated σ -bonds in chains of *trans* formation of P(VDF-DB)-500 and *gauche* formation of P(VDF-DB)-500.

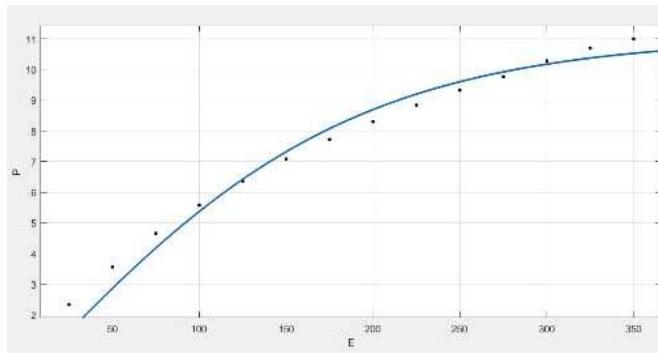


Fig. S14. The P_s and k are calculated from $|P_E| = P_s \tanh(k|E|)$ by Matlab; $f(x) = a * (\exp(2 * b * x) - 1) / (\exp(2 * b * x) + 1)$, $a = 11.05 \mu\text{C cm}^{-2}$, $b = 0.5313 * 10^{-8} \text{ m/V}$ (P_s is a and k is b).

1 **Table S1.** ^{19}F NMR assignment of pristine P(VDF-CTFE) and resultant P(VDF-DB)

Peak	Sequence	Chemical Shift (ppm)
a	-CF=CHCF ₂ CH ₂ CF ₂ -	-89.2
b	-CF ₂ CH ₂ CF ₂ CH ₂ CF ₂ -	-92.3
c	-CFCICH ₂ CF ₂ CH ₂ CF ₂ -	-93.16 to -93.67
d	-CH ₂ CH ₂ CF ₂ CH ₂ CF ₂ -	-94.26 to -97.31
e	-CF ₂ CFCICF ₂ CFCICF ₂ -	-106.5 to -107.82
f	-CF ₂ CH ₂ CF ₂ CF ₂ CFCl-	-108.0 to -109.34
g	-CF ₂ CFCICF ₂ CFCICH ₂ -	-109.34 to -113.10
h	-CF ₂ CFCICF ₂ CF=CH-	-110.1 to -111.6
i	-CF ₂ CF ₂ CF=CHCF ₂ -, CF ₂ CH ₂ CF ₂ CF ₂ CF=CH-	-113.4 to -113.7
j	-CF ₂ CH ₂ CF ₂ CF ₂ CH ₂ -	-114.7
k	-CH ₂ CF ₂ CF ₂ CH ₂ CH ₂ -	-116.6 to -118.8
l	-CH ₂ CF ₂ CF ₂ CFCICH ₂ -	-118.25 to -119.98
m	-CF ₂ CF ₂ CFCICH ₂ CF ₂ -	-120.35 to -123.32
n	-CH ₂ CF ₂ CF ₂ CF=CH-	-120.0 to -120.8
o	-CF ₂ CH ₂ CFCICF ₂ CH ₂ -	-129.46 to -130.53

Table S2. The permittivity of the polymers measured at 20 °C

Polymer	ϵ_r (0.01 HZ)	ϵ_r (0.1 HZ)	ϵ_r (1 Hz)	ϵ_r (10 HZ)	ϵ_r (10 ² HZ)	ϵ_r (10 ³ HZ)	ϵ_r (10 ⁴ HZ)	ϵ_r (10 ⁵ HZ)	ϵ_r (10 ⁶ HZ)
P(VDF-TrFE-CTFE)	68.35	44.60	34.38	29.21	25.95	24.15	21.79	17.86	10.89
P(VDF-DB)-500	656.4	107.13	81.23	72.41	61.95	51.61	42.06	32.73	21.81
P(VDF-DB)-as-cast	473.6	33.82	19.89	15.73	14.15	13.03	12.36	11.46	9.55
P(VDF-CTFE)	231.8	18.86	13.14	11.35	10.78	9.94	9.33	8.09	6.51

Table S3. The electromechanical property of typical relaxor ferroelectric polymers

polymer[†]	S_{30}^{\dagger} (%)	S_{50}^{\dagger} (%)	S_M(%)	W_m (J cm⁻³)	k_{33}	Ref
Irradiated-P(VDF-TrFE) ^t 68/32	-0.6	-1.5	-5	0.5	0.3	32
P(VDF-TrFE-CTFE) [†] 65/35/10	-0.6	-1.6	-4	0.32	0.28	25
P(VDF-TrFE-CFE) [†] 68/32/4	0	-0.5	-4.5	1.1	0.55	17
P(VDF-TrFE-CFE) [†] 68/32/9	-0.3	-1.2	-7	0.73	-	12
P(VDF-DB)-500 [†]	-1.3	-2.5	-13.4	3.1	0.5	This work

[†] S_{30} is the strain measured at 30 MV m⁻¹. S_{50} is the strain measured at 50 MV m⁻¹. Irradiated-P(VDF-TrFE), P(VDF-TrFE-CTFE) and P(VDF-TrFE-CFE) are measured at 1 Hz. P(VDF-DB)-500 is measured at 10 Hz.