Supplementary Information (SI) for Journal of Materials Chemistry C.

SUPPORTING INFORMATION

Enhanced piezoelectric output performance in flexible polyvinylidene fluoride foam with a hierarchical dual-pore structure

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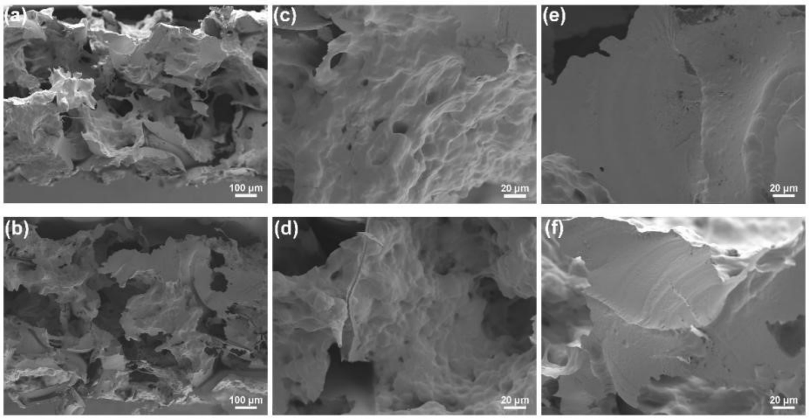

Fig. S1 SEM images of the cross section of the Foam-pf sample.

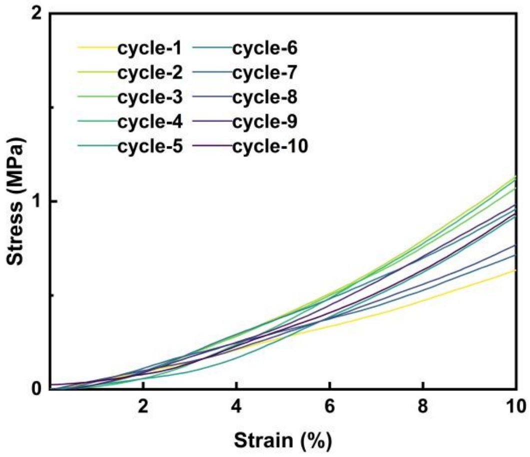

Fig. S2 10-cycle compression curves of the Foam-pf sample.

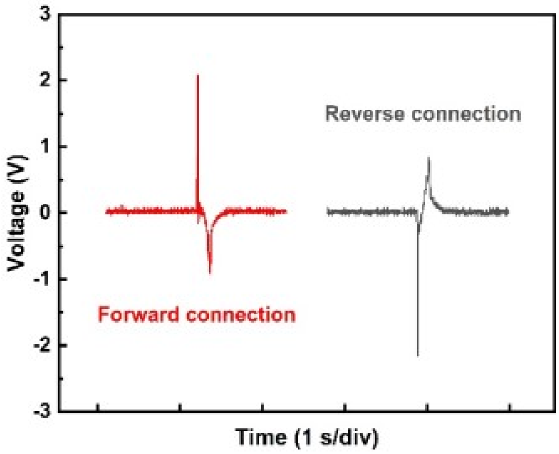

Fig. S3 The voltage output of Foam-pf under pressurized load, the red and black curves were tested under forward and reverse connections.

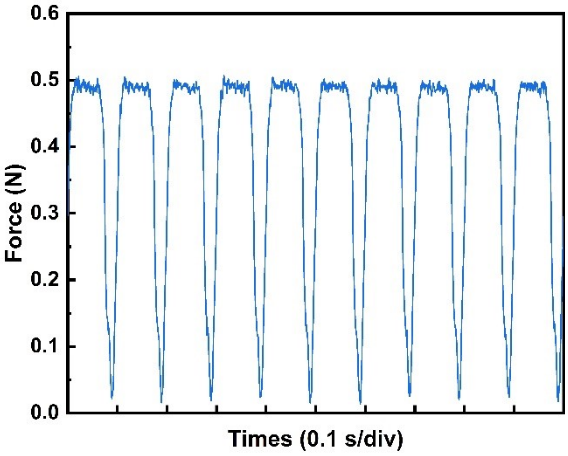

Fig. S4 Cyclical compressive pressure load-release curves of the exciter at 10 Hz.

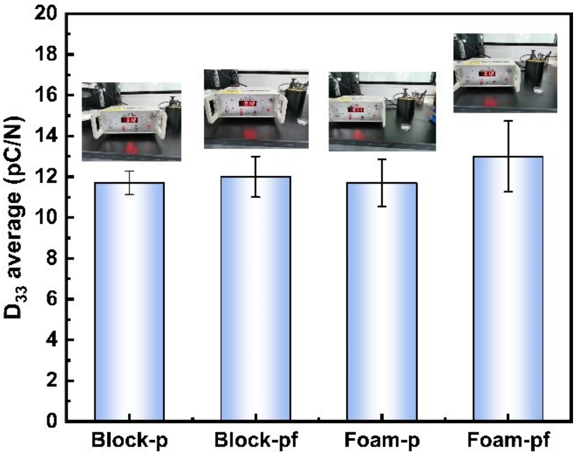

Fig. S5 Average piezoelectric coefficient (d_{33}) of different samples.

In the test, random points on the samples were selected for testing, and the piezoelectric coefficients of various samples fluctuated in the range of 11~15 pC/N, and there was no obvious difference between these samples.

Table S1. The dielectric constant setting of the simulated material.

Sample	Block-p	Block-pf	Foam-p	Foam-pf
Porosity (%)	9.43	18.11	71.41	71.54
ε_{11}	6.80	6.24	7.40	6.24
ε_{22}	8.52	7.80	9.30	7.80
ε_{33}	6.98	6.40	7.60	6.40

Since it has been reflected in the modeling of network pore, the influence of macropores on the dielectric constant is not considered, for example, the dielectric constant used in Foam-p is the dielectric constant of dense PVDF, and the dielectric constant of Foam-pf refers to the Block-pf prepared with the same composition.

Table S2. The values of sensitivity of piezoelectric materials under low loads

Materials	Applied load (N)	Sensitivity (V/N)	Refs
PVDF/MWCNT	2	3.25	[58]
P(VDF-TrFE) nanofiber	0.05	0.0605	[59]
P(VDF-TrFE) film	1	0.01	[60]
PVDF/$BaTiO_3$/graphene	2	5	[61]
PVDF foam	2	5.92	[17]
PVDF film	1	0.24	[62]
PVDF/PDA/$BaTiO_3$	3	4.67	[63]
PVDF/$BaTiO_3$	2	9.65	[64]
PVDF/GP	2	8.49	[65]
PVDF film	0.09	4.961	[66]
PVDF-HFP/ZnO	0.5	4.625	[67]
PVDF film	1~11	0.0356	[68]
Hierarchical dual-pore PVDF	0.5	11.597	**This work**

The phase content calculation methods

$$F_{(\beta)} = \frac{A^{841}_{\beta}}{\left(\frac{K^{841}_{\beta}}{K^{764}_{\alpha}}\right) A^{764}_{\alpha} + A^{841}_{\beta}} \times 100\%\#\ (1)$$

$$A_{762} = K^{762}_{\alpha} \cdot X_{\alpha} \cdot t\#\ (2)$$

$$A_{1275} = K^{1275}_{\beta} \cdot X_{\beta} \cdot t\#\ (3)$$

$$A_{835} = \left(K^{835}_{\beta} \cdot X_{\beta} + K^{835}_{\gamma} \cdot X_{\gamma} + K^{835}_{am}(1 - X_{total})\right)\ t\#\ (4)$$

$$F_{(\gamma)} = \frac{X_{\gamma}}{X_{\alpha} + X_{\beta} + X_{\gamma}}\#(5)$$

where A^{j}_{i} is the baseline-corrected absorbance of i-phase at j cm^{-1}, and K^{j}_{i} is the absorption coefficient, $\frac{K^{841}_{\beta}}{K^{764}_{\alpha}}$ is determined at 1.26, A_{i} is the baseline-corrected absorbance at i cm^{-1}, X_{i} represents mole fraction of i-phase, and t is the thickness calculated from the IR absorption band at 1070 cm^{-1} which corresponds to $A_{1070} = 0.095t + 0.07$. X_{total} is the total crystallinity of the sample, calculated by the DSC from Equation 2. For K^{762}_{α}, K^{1275}_{β}, K^{835}_{β}, K^{835}_{γ}, and K^{835}_{am}, the values of absorption coefficients obtained were 0.365, 0.140, 0.132, 0.150, and 0.0259 μm^{-1}, respectively.

The content of the *β*-phase ($F_{(\beta)}$) could be calculated with Equation 3. The characteristic infrared peaks of the γ-phase were partially overlapped with other phases and its absorption coefficient at the individual characteristic peaks was relatively small, which was not conducive to the accuracy of the quantitative analysis. So the content of the γ-phase could be calculated in the combination with the *α*-phase and *β*-phase, as shown in Equation 1 to Equation 5.