

Supplementary Materials

Hierarchical porous scaling-up TENGs with enhanced charge density via conductive-ferroelectric hybrid fillers for intelligent home ecosystems

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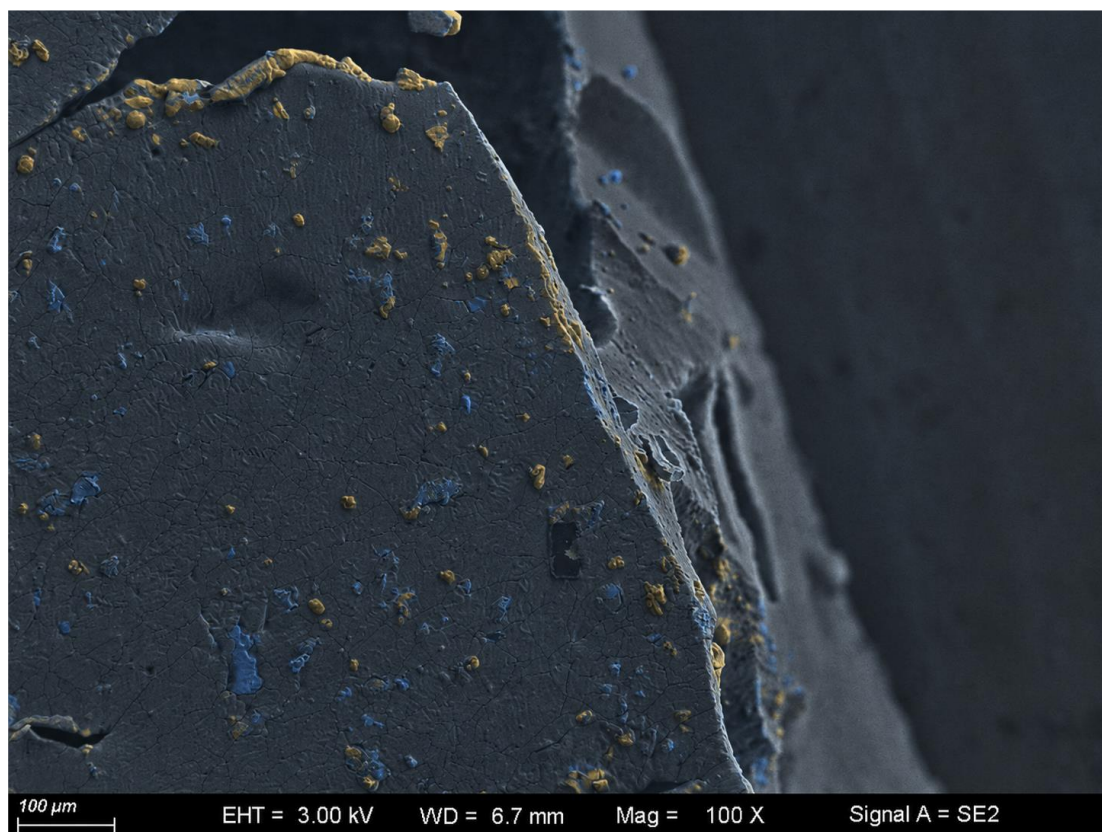


Figure S1. The cross-sectional SEM image of BT/Ecoflex/rGO-Fe composite sponge. Note: False colors were added solely to improve visual contrast and do not represent quantitative elemental identification.

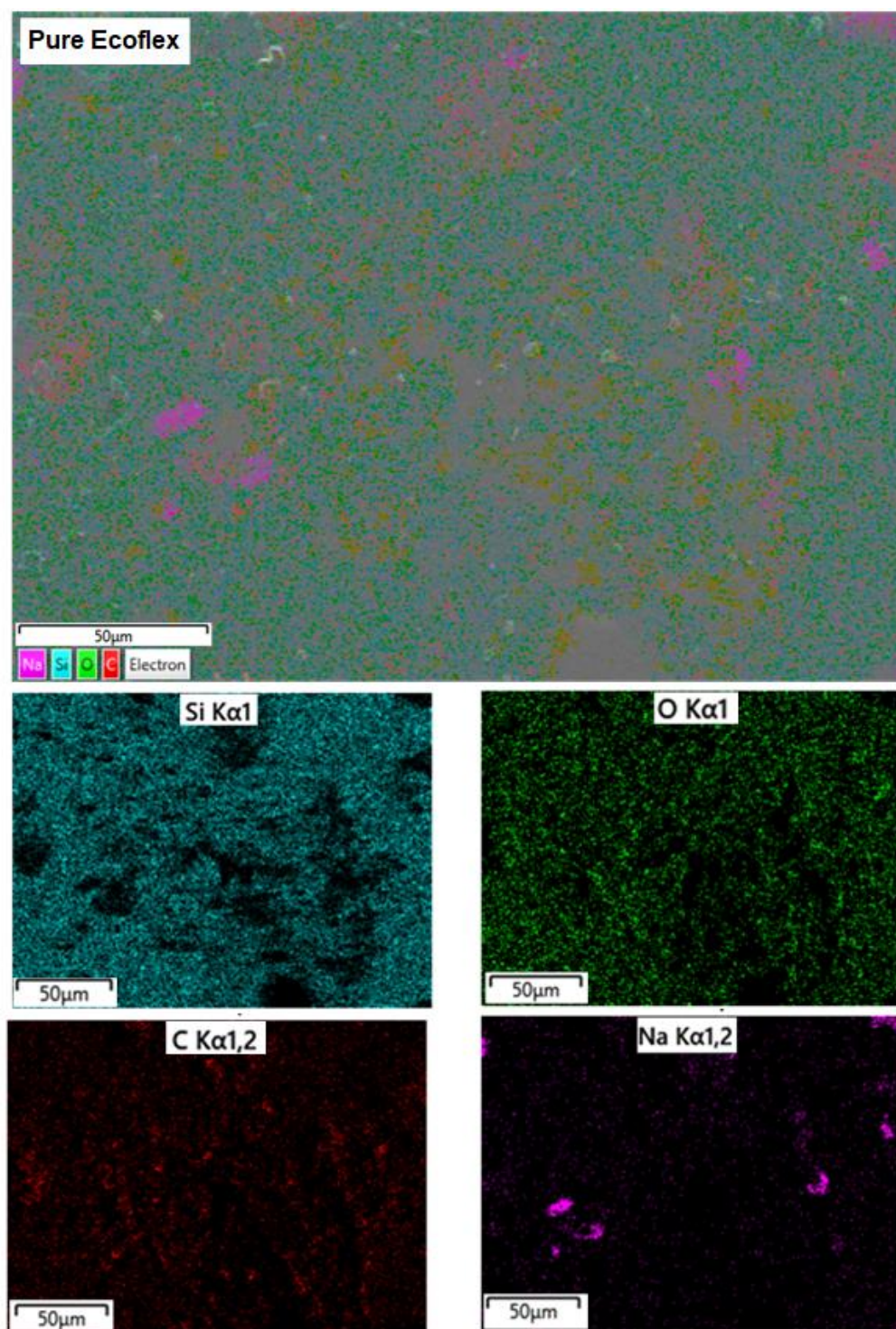


Figure S2. Energy Dispersive Spectroscopy of pure Ecoflex.

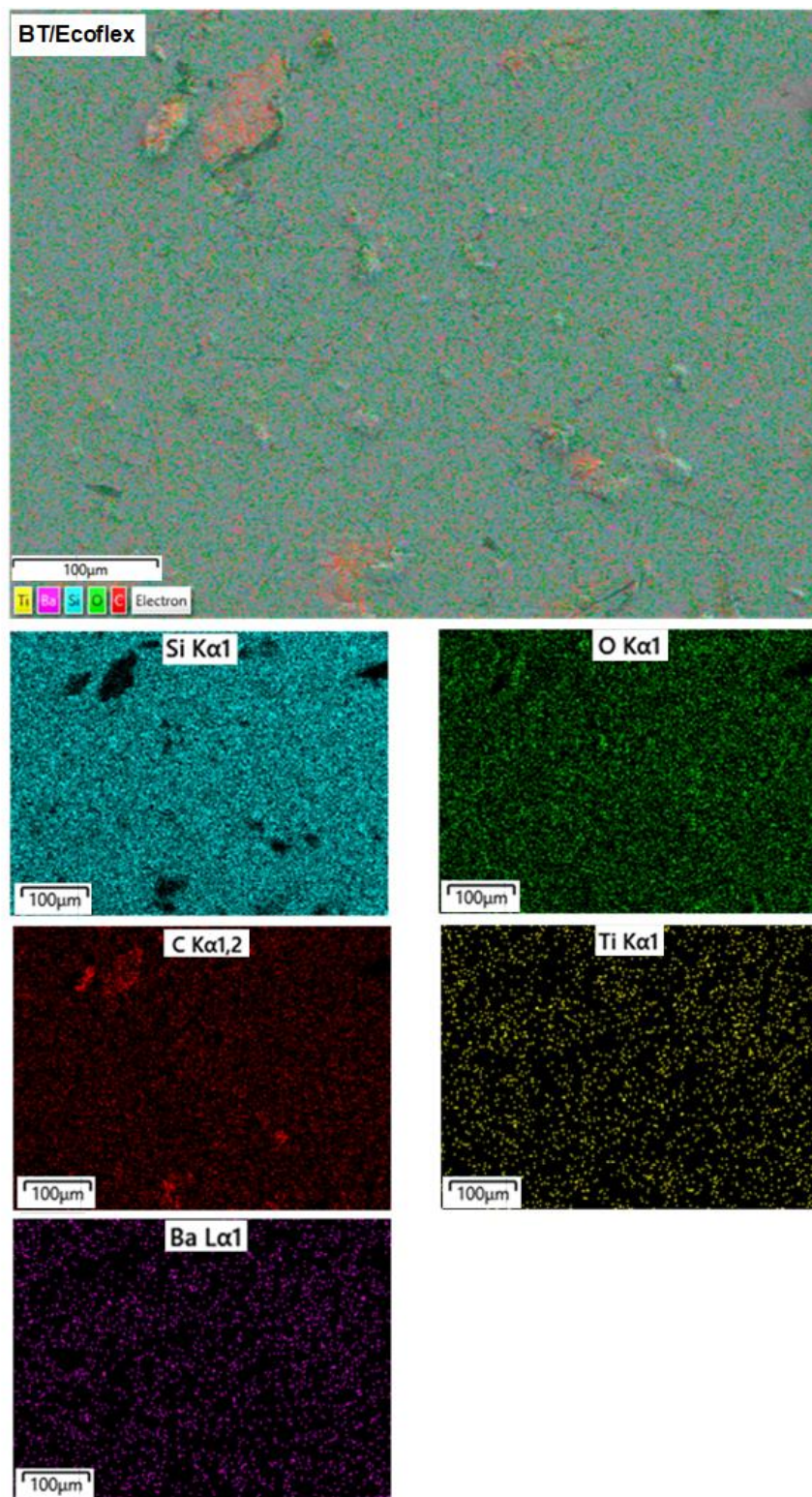


Figure S3. Energy Dispersive Spectroscopy of BT/Ecoflex.

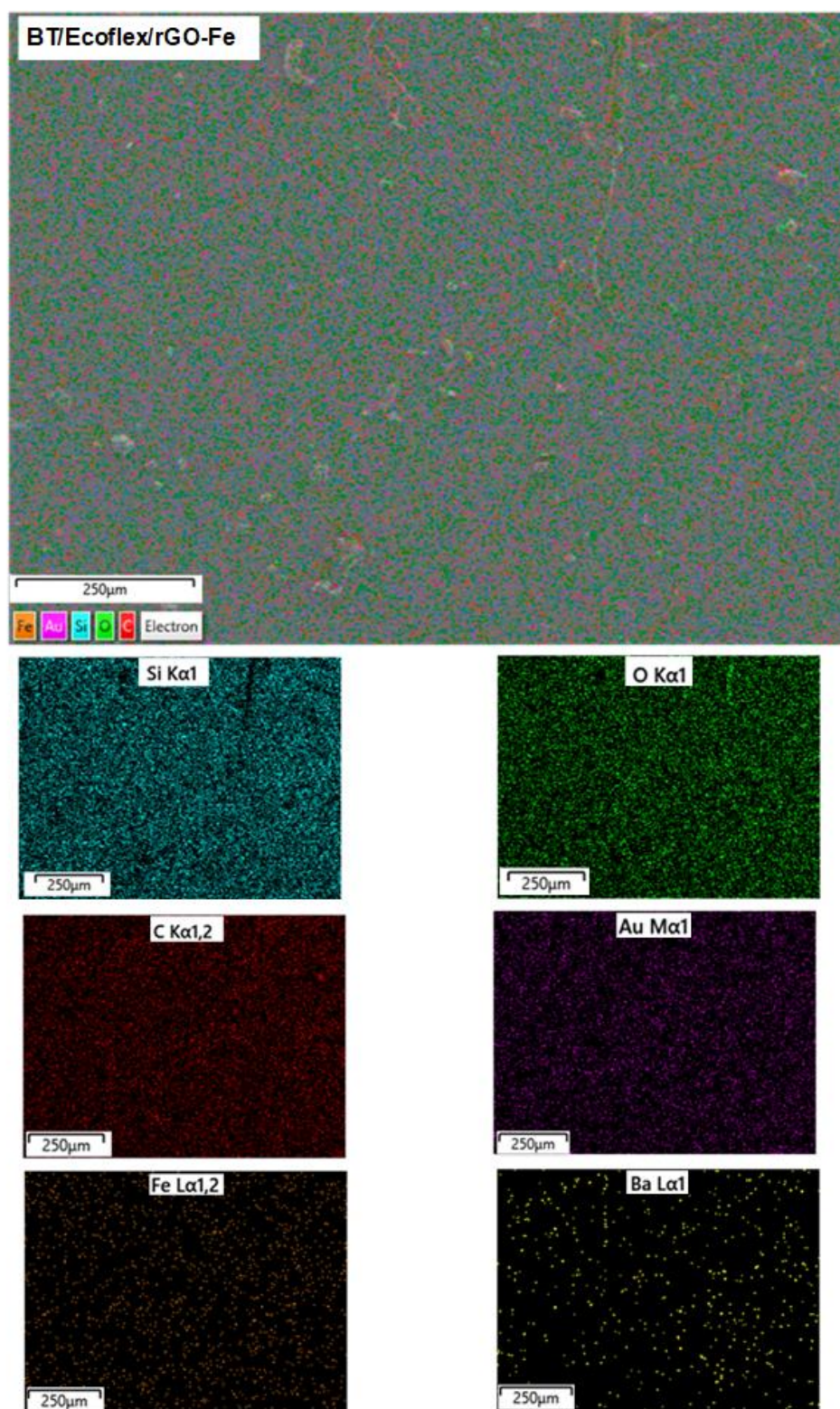


Figure S4. Energy Dispersive Spectroscopy of BT/Ecoflex/rGO-Fe.

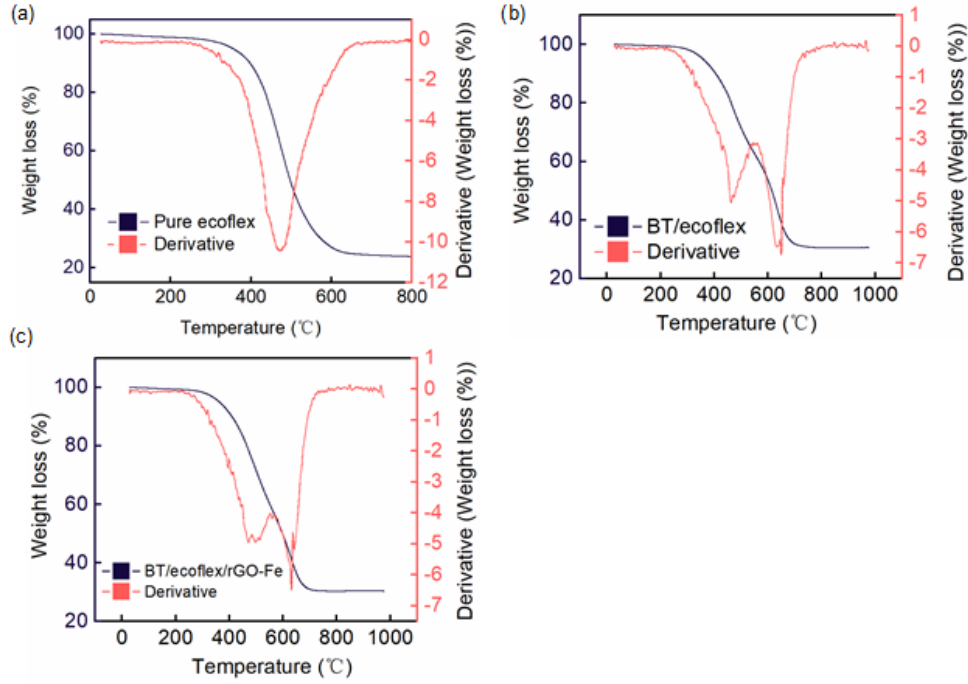


Figure S5. Thermogravimetric analysis of composite materials. (a) The thermogravimetric analysis of pure Ecoflex; (b) The thermogravimetric analysis of BT/Ecoflex; (c) The thermogravimetric analysis of BT/Ecoflex/rGO-Fe.

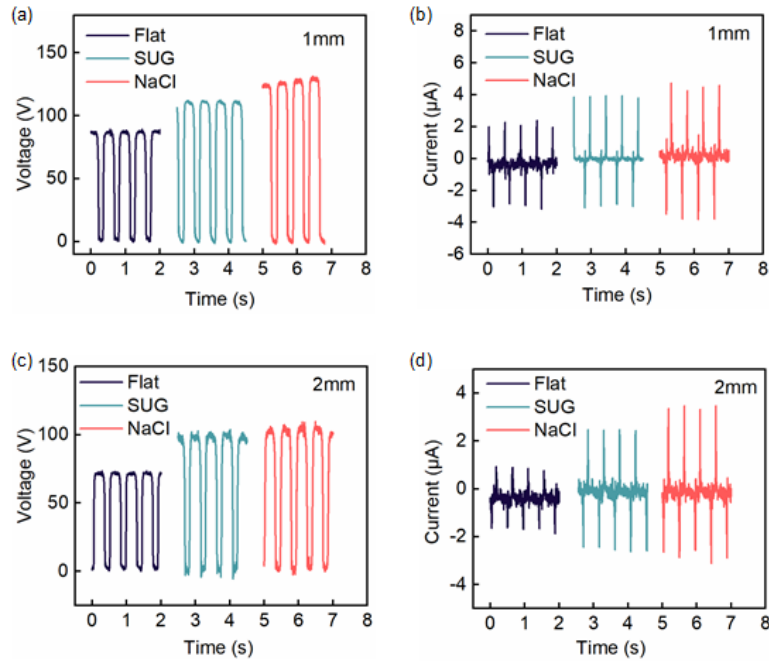


Figure S6. Electrical output performance of pure Ecoflex-based TENG with different particle sizes. (a) voltage of TENG when Ecoflex thickness is 1 mm; (b) current of TENG when Ecoflex thickness is 1 mm.; (c) voltage of TENG when Ecoflex thickness is 2 mm; (d) current of TENG when Ecoflex thickness is 2 mm;

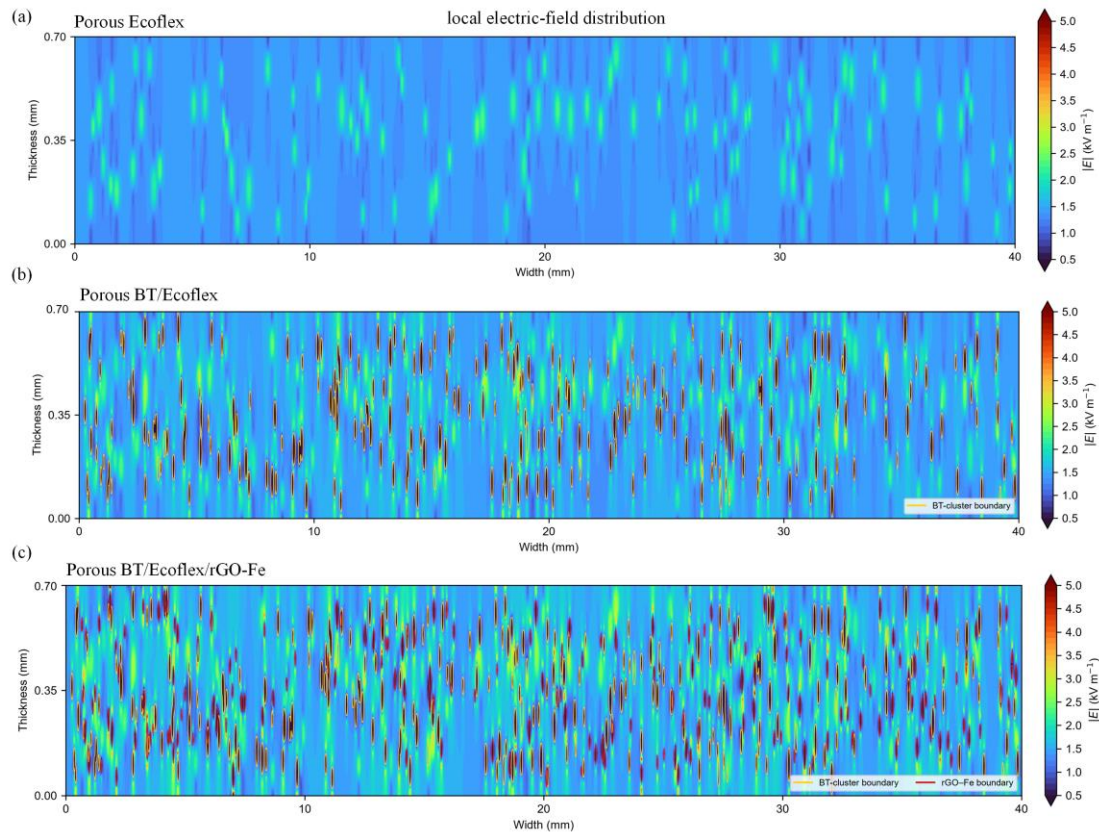


Figure S7. Finite-element analysis of the porous dielectric composites under compression. (a) Local electric-field distribution in porous Ecoflex. (b) Electric-field distributions in porous BT/Ecoflex. (c) Electric-field in porous BT/Ecoflex/rGO-Fe.

A two-dimensional finite element model was established in COMSOL Multiphysics to study the electric field and polarization distribution of porous Ecoflex, porous BT/Ecoflex and porous BT/Ecoflex/rGO-Fe under compression. The representative cross section has a width of 40 mm and a thickness of 1 mm. The compression deformation is set along the thickness direction, and 30% compression strain is selected as the representative state. Pores and packing clusters are represented by nearly circular regions which are randomly and uniformly distributed. In order to isolate the influence of material composition, a statistically comparable pore structure and the same boundary conditions are adopted. The upper boundary is given a standardized potential of 1 volt, while the lower boundary is grounded, and the calculation frequency of electrical response is 3 Hz. The compressed geometry is transferred to electrostatic calculation to obtain local electric field intensity, polarization intensity and electrostatic energy density. By using standardized electrical excitation, the relative field response and polarization response among the three material models can be compared, instead of being used to reproduce the absolute output voltage of the device.

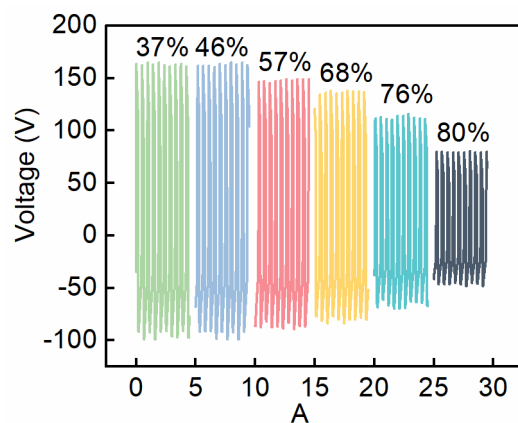


Figure S8. Effect of humidity on output voltage of composite sponge-based TENG.

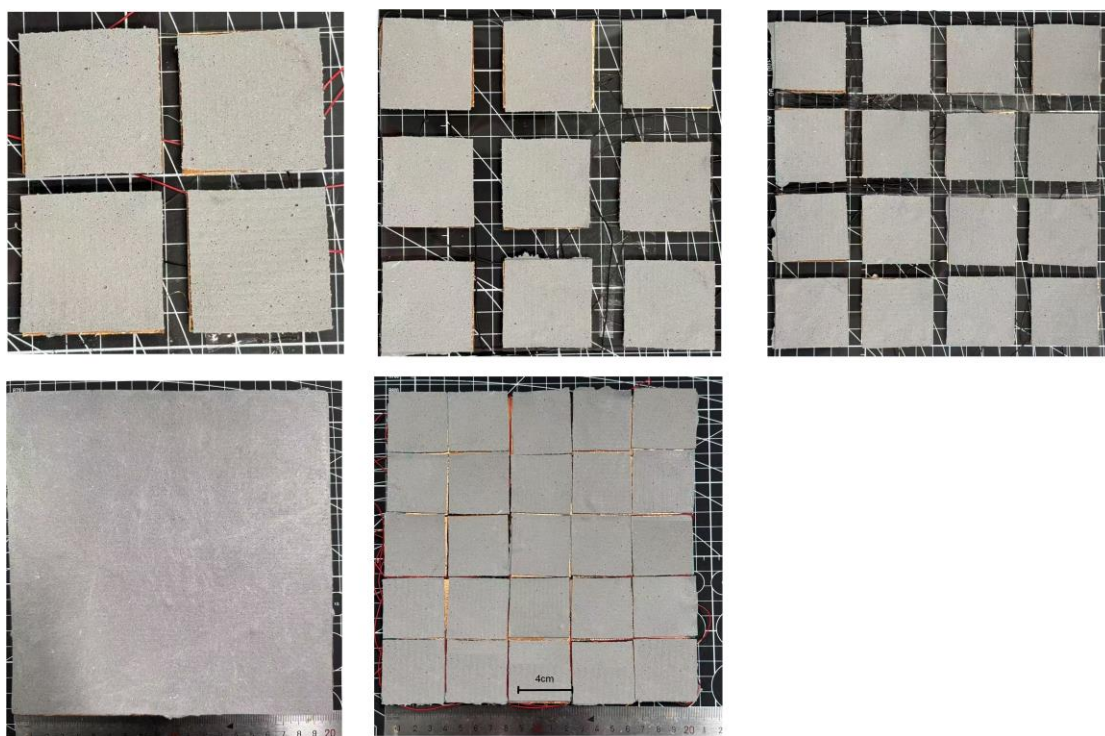


Figure S9. Physical photos of TENGs with different areas. (The photographs were taken by the authors.)

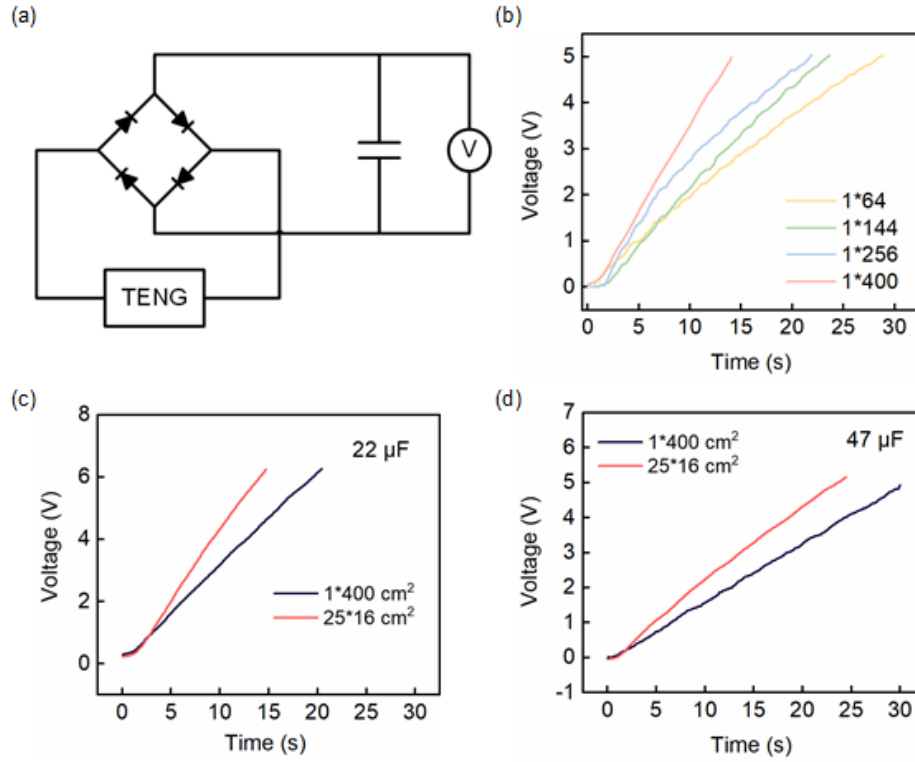


Figure S10. Capacitance charging experiments of the M-TENG and S-TENG. (a) Schematic diagram of charging circuit; (b) Charging 33 μ F capacitor with S-TENG; (c) Charging 22 μ F capacitor with large area TENGs; (d) Charging 47 μ F capacitor with large area TENGs.

Supplementary Note 1. Effective dielectric response of the porous composites

The porous composite sponge constitutes a multiphase dielectric system consisting of the Ecoflex-based solid skeleton, BT, rGO-Fe, and air-filled pores. Therefore, the measured relative permittivity represents the effective dielectric response of the entire porous sample, rather than the intrinsic relative permittivity of a single component.

Under the influence of an alternating electric field, the dielectric response can usually be expressed by the complex relative permittivity:

$$\varepsilon^* = \varepsilon' - j\varepsilon'' \quad (1)$$

where ε' represents the real part related to polarization and capacitive energy storage, while ε'' represents the imaginary part related to dielectric loss.

The dielectric loss tangent is defined as

$$\tan \delta = \frac{\varepsilon''}{\varepsilon'} \quad (2)$$

For a sinusoidal electric field, the average dielectric-loss power density can be expressed as:

$$P_{\text{loss}} = \frac{1}{2} \omega \varepsilon_0 \varepsilon'' E_0^2 = \frac{1}{2} \omega \varepsilon_0 \varepsilon' \tan \delta E_0^2 \quad (3)$$

where ω is the angular frequency, ε_0 is the vacuum permittivity, and E_0 is the electric field amplitude. Therefore, ε' and $\tan \delta$ characterize two complementary aspects of the dielectric response: polarization/capacitance storage and dielectric dissipation, respectively.

For isotropic multiphase dielectric systems, the effective dielectric can be approximated in complex form [S1]:

$$\sum_i \phi_i \frac{\varepsilon_i^* - \varepsilon_{\text{eff}}^*}{\varepsilon_i^* + 2\varepsilon_{\text{eff}}^*} = 0 \quad (4)$$

where ϕ_i is the volume fraction of component i , ε_i^* is the complex relative permittivity of component i , and $\varepsilon_{\text{eff}}^*$ is the effective complex relative permittivity of the porous composite material.

To illustrate the effect of porosity, porous composite materials can be simplified as a two-phase system consisting of an effective solid composite skeleton and air-filled pores:

$$(1-P) \frac{\varepsilon_s^* - \varepsilon_{\text{eff}}^*}{\varepsilon_s^* + 2\varepsilon_{\text{eff}}^*} + P \frac{\varepsilon_{\text{air}}^* - \varepsilon_{\text{eff}}^*}{\varepsilon_{\text{air}}^* + 2\varepsilon_{\text{eff}}^*} = 0 \quad (5)$$

where P is the porosity, and ε_s^* represents the effective complex relative permittivity of the solid composite framework.

For the air phase,

$$\varepsilon'_{\text{air}} \approx 1, \varepsilon''_{\text{air}} \approx 0 \quad (6)$$

Therefore, increasing the volume fraction of air-filled pores dilutes the effective dielectric contribution of the solid composite skeleton. In this composite system, Ecoflex, BT, and rGO-Fe together constitute the effective solid skeleton. Although the BT content is 10 wt%, its corresponding volume fraction is relatively low due to the much higher density of BT compared to Ecoflex. Furthermore, the BT particles are embedded in the low-dielectric-constant Ecoflex matrix and spatially separated, thus preventing the formation of a continuous high-dielectric-constant ceramic phase.

These combined effects explain the experimentally observed moderate increase in the effective relative permittivity from approximately 2.3 to 2.8 after BT doping. Within the studied load range, the addition of rGO-Fe did not lead to a significant further increase in the effective relative permittivity.

Therefore, the numerical values shown in Figure 3(d) characterize the capacitance-dependent effective relative permittivity of the porous samples and are used to compare their polarization/capacitance responses. The complex permittivity form described above further distinguishes this contribution from the dielectric loss component and provides a consistent theoretical framework for explaining the role of porous structures in the dielectric response of composite sponges.

References

- [S1] Bruggeman, D. A. G. Berechnung verschiedener physikalischer Konstanten von heterogenen Substanzen. I. Dielektrizitätskonstanten und Leitfähigkeiten der Mischkörper aus isotropen Substanzen. *Annalen der Physik* 1935, 416, 636-664. [DOI:10.1002/andp.19354160705], URL: <https://onlinelibrary.wiley.com/doi/10.1002/andp.19354160705>.

Supplementary Note 2: Parameters of capacitors and rectifiers

The parameters of capacitors and rectifiers in this paper:

1. The saturation voltage of the capacitors is 50 V.
2. Specifications of the rectifier bridge are:
 - (1) Model: 2W10
 - (2) Maximum Repetitive Peak Reverse Voltage: 1000 V;
 - (3) Maximum RMS Voltage: 700 V;
 - (4) Maximum DC Blocking Voltage: 1000 V;
 - (5) Maximum Average Forward Rectified Output Current: 2.0 A;
 - (6) Peak Forward Surge Current: 50A;
 - (7) Maximum Instantaneous Forward Voltage Drop Per Leg @2.0A: 1.1V;
 - (8) Operating Temperature Range -55°C to +125°C;
 - (9) Storage Temperature Range -55°C to +150°C;

Supplementary Note 3: The equivalent circuit model of Composite sponge-based TENG

Each TENG unit is represented by a displacement-related current source i_T , $i(t)$ in parallel with its internal capacitance $C_{T,i}[x_i(t)]$ and charge leakage resistance $R_{\text{leak},i}$, followed by an equivalent series resistance, a bridge rectifier and an energy storage capacitor C_L . The basic voltage-charge relationship of each contact out cell is expressed as

$$V_{T,i}(t) = -\frac{Q_i(t)}{C_{T,i}[x_i(t)]} + V_{\text{OC},i}[x_i(t)] \quad (1)$$

After periodic averaging, the rectified TENG can be approximately expressed by Thevenin's Theorem, effective voltage V_{eq} and output resistance R_{eq} . Ignoring the leakage of the storage capacitor, its charging behavior is given by the following formula

$$C_L \frac{dV_C}{dt} = \frac{V_{eq} - 2V_D - V_C}{R_{eq}} \quad (2)$$

to

$$V_C(t) = (V_{eq} - 2V_D) \left[1 - \exp\left(-\frac{t}{R_{eq} C_L}\right) \right] \quad (3)$$

Therefore, the time constant $\tau = R_{eq} C_L$ obtained from the experimental charging curve provides a quantitative measurement of the effective output impedance. The energy stored in the capacitor is calculated as $E_C = \frac{1}{2} C_L V_C^2$

Importantly, segmentation itself does not generate additional energy. Under the ideal uniform synchronous contact condition, S-TENG and M-TENG with the same total contact area should generate roughly the same total charge. Therefore, the M-TENG enhancement observed in the experiment comes from the reduction of non-ideal scaling loss. In large area S-TENG, substrate deformation, spatial uneven pressure, residual air gap and asynchronous local contact will reduce the effective contact coefficient and promote charge dissipation. By dividing the same total area into mechanical compliant units, M-TENG improves local contact consistency and increases the proportion of frictional charges transferred to external circuits.

This effect can be described using an effective charge collection coefficient. For the

S-TENG, $Q_S = \eta_S \sigma A_S$, whereas for the M-TENG, $Q_M = \sum_{i=1}^N \gamma_i \eta_i \sigma A_i$.

Here, η_i represents local contact and charge retention efficiency, and γ_i represents synchronization and rectification loss. The average value of $\gamma_i \eta_i$ may exceed the effective coefficient η_S of a large-scale single-cell device because smaller cells conform to the applied pressure more effectively. Therefore, in the same charging time, M-TENG shows higher rectified charging current, smaller effective charging time constant and more energy stored in the capacitor. This analysis determines the reduction of spatial electromechanical inhomogeneity, rather than a separate geometric division, as the physical origin of the observed multi-cell enhancement.