Electronic Supplementary Information for

Surface Microstructural Engineering of Silicone Elastomers for High Performance Adhesive Surface-Enabled Mechanical Energy Harvesters

Haiyan Fu, Jianliang Gong,* Hao Zhong, Bao Yang, Zuchang Long, Jiqing Zeng, Zhiyu Cheng, Jialin He, Bingang Xu, Yiwang Chen*

1. The $V-Q-x$ relationship for AS-TENGs

Since the areal size (*S*) of used CNFs is much larger than their separation distance (the sum of SE layer thickness (d_{SE}), air gap (x_{air}) and VPA layer thickness (d_{SE})), it is reasonable to assume that the two electrodes are infinitely large and their charges induced on the metal electrodes are uniformly distributed. Therefore, the electrostatic fields at different regions between the two electrodes can be simply estimated based on Gauss's Law. Inside the dielectric layer of SE

$$E_{SE} = \frac{-\sigma_{EI}}{\varepsilon_0 \varepsilon_{SE}} \tag{1}$$

Inside the air gap

$$E_{air} = \frac{\sigma_{CE} - \sigma_{EI}}{\varepsilon_0} \tag{2}$$

Inside the dielectric layer of VPA

$$E_{VPA} = \frac{-\sigma_{EI}}{\varepsilon_0 \varepsilon_{VPA}} \tag{3}$$

where ε_0, ε_{SE}, and ε_{VPA} are the vacuum permittivity and the relative permittivity of SE and VPA materials, respectively.

The instantaneous electric potential difference (i.e. voltage) between the two electrodes can be given by the following equation

$$V(t) = E_{SE}d_{SE} + E_{air}x_{air}(t) + E_{VPA}d_{VPA} \tag{4}$$

Substituting Equations (1)–(3) into Equation (4)

$$V(t) =- \frac{\sigma_{EI}(t)}{\varepsilon_0}\left(\frac{d_{SE}}{\varepsilon_{SE}} + \frac{d_{VPA}}{\varepsilon_{VPA}} + x_{air}(t)\right) + \frac{\sigma_{CE}}{\varepsilon_0}x_{air}(t) \tag{5}$$

where $V(t)$ and $\sigma_{EI}(t)$ are the instantaneous voltage and induced charge density during the contacting or separating process of two surface-electrified materials at a separation distance of $x_{air}(t)$, respectively

Combining ${d_{SE}}/{\varepsilon_{SE}}+{d_{SE}}/{\varepsilon_{SE}}$ as the effective thickness constant d_0, the Equation (5) for calculating instantaneous voltage can be further simplified as follow

$$V(t) =- \frac{\sigma_{EI}(t)}{\varepsilon_0}\left(d_0 + x_{air}(t)\right) + \frac{\sigma_{CE}}{\varepsilon_0}x_{air}(t) \tag{6}$$

When shorting or connecting an external load (R) between their electrodes, the electrostatic potential difference created during the contacting and separating process can drive free electrons flow across the external load forth and back. Defining the amount of the transferred electrons between the two electrodes as Q, it is equal to the instantaneous amount of charges induced on the electrode:

$$Q(t) = \sigma_{EI}(t)S \tag{7}$$

where S is the areal size of electrode.

Substituting Equation (7) into Equation (6), a $V-Q-x$ relationship for AS-TENG based on vertical contact-separation mode during the separating process can be given by

$$V(t) =- \frac{Q(t)}{S\varepsilon_0}\left(d_0 + x_{air}(t)\right) + \frac{\sigma_{CE}}{\varepsilon_0}x_{air}(t) \tag{8}$$

The electricity generation performance of AS-TENGs can be evaluated by measuring the output voltage across the external load (R). Based on Ohm's law, its corresponding instantaneous output current ($I(t)$) can be expressed as

$$I(t) =- \frac{Q(t)}{RS\varepsilon_0}\left(d_0 + x_{air}(t)\right) + \frac{\sigma_{CE}}{R\varepsilon_0}x_{air}(t) \tag{9}$$

2. The relationship of separation distance between two CE surfaces with triggering frequency and time

To enable CE surfaces repetitively contact and separate for continuous electricity generation, an external force was applied on the top electrode for driving it to undergo a sinusoidal motion. The separation distance between two CE surfaces ($x_{air}(t)$) can be given by the following equation

$$x_{air}(t) = A_0 \sin(\omega t + \theta) + A_0 \quad (10)$$

where A_0 , ω and θ are the motion amplitude, angular velocity and initial phase angle, respectively.

The angular velocity of the motion is dependent on impact frequency (f) of one triboelectric material to contact with another triboelectric material, which can be given by

$$\omega = 2\pi f \quad (11)$$

Substituting Equation (11) into Equation (10)

$$x_{air}(t) = A_0 \sin(2\pi f t + \theta) + A_0 \quad (12)$$

3. Calculation of triboelectric charges and electric energy based on output voltage performance of TENGs

The amount of free electrons moving from one electrode to another electrode was equal to the amount of charges induced on the electrode, while the latter was determined by triboelectric charges generated *via* CE. Therefore, the CE performance of different triboelectric material pairs can be evaluated and compared based on calculating the amount of charges (electrons) transferred across the resistance load between two electrons(Q). It can be calculated by time integral of output current using the equation below

$$Q = \int_{t_1}^{t_2} I dt = \int_{t_1}^{t_2} \frac{U}{R} dt \tag{13}$$

where Q is the amount of charges generated by the TENG, I is the instantaneous output current, U is the output voltage, R is the load resistance, and t_1 and t_2 represent the start time and end time of a single contact, respectively.

Assuming that the external load was a pure resistance, the electric energy (E) equal to the Joule heating energy also can be calculated by the following equation

$$E = \int_{t_1}^{t_2} \frac{U^2}{R} dt \tag{14}$$

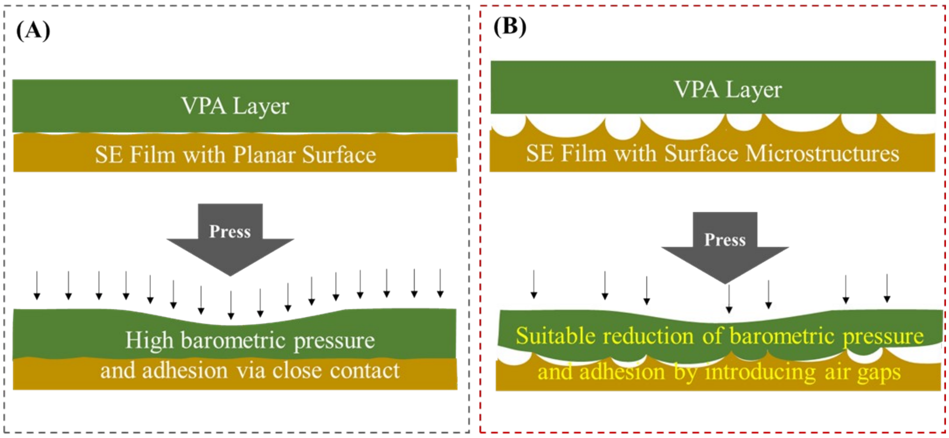

Figure S1 Schematic comparison of interface adhesive strength between VPA layer and SE film with (A) planar surface and (B) surface microstructures

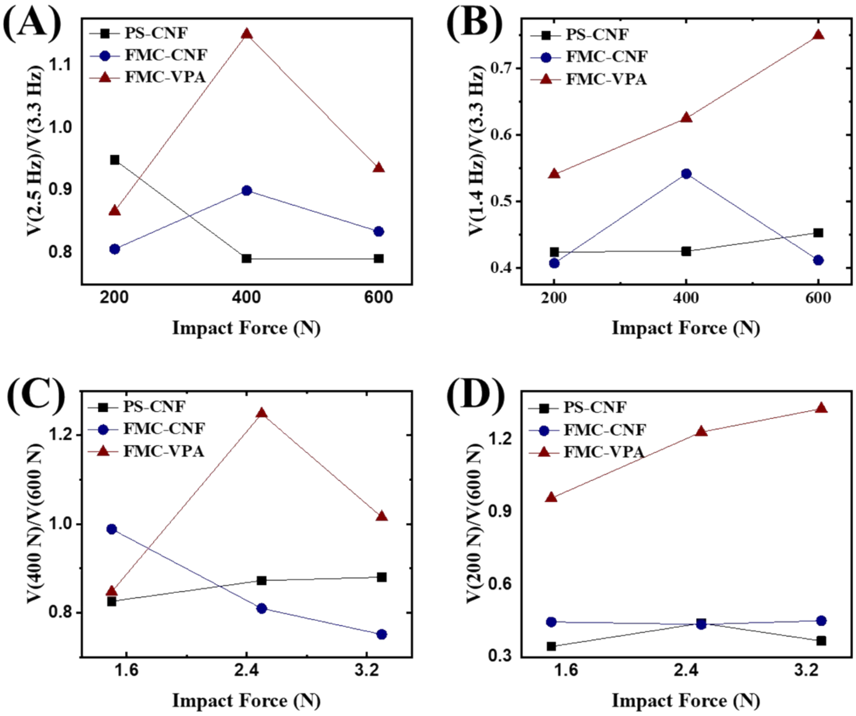

Figure S2 Performance degradation comparison of AS-TENG based on FMC-VPA and NAS-TENGs based on PS-CNF and FMC-CNF at different impact forces and triggering conditions

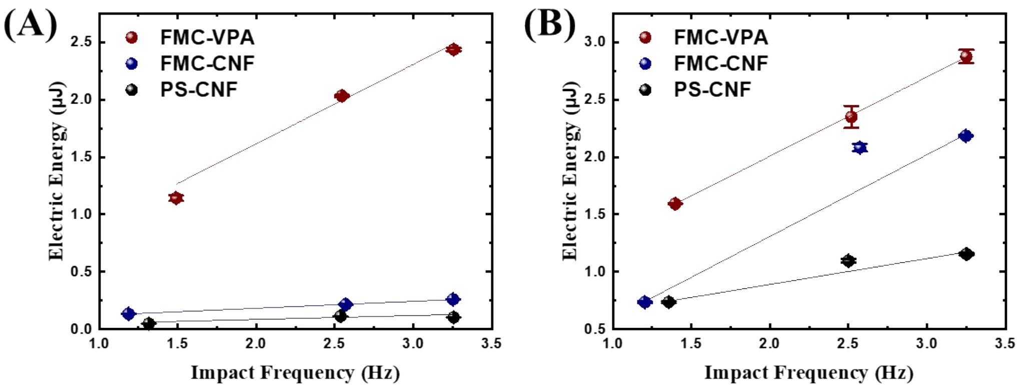

Figure S3 The dependence of triggering (impact) frequency on electric energy of AS-TENG (FMC-VPA) and NAS-TENGs (FMC-CNF and PS-CNF) at (A) 200 N and (B) 600 N, respectively

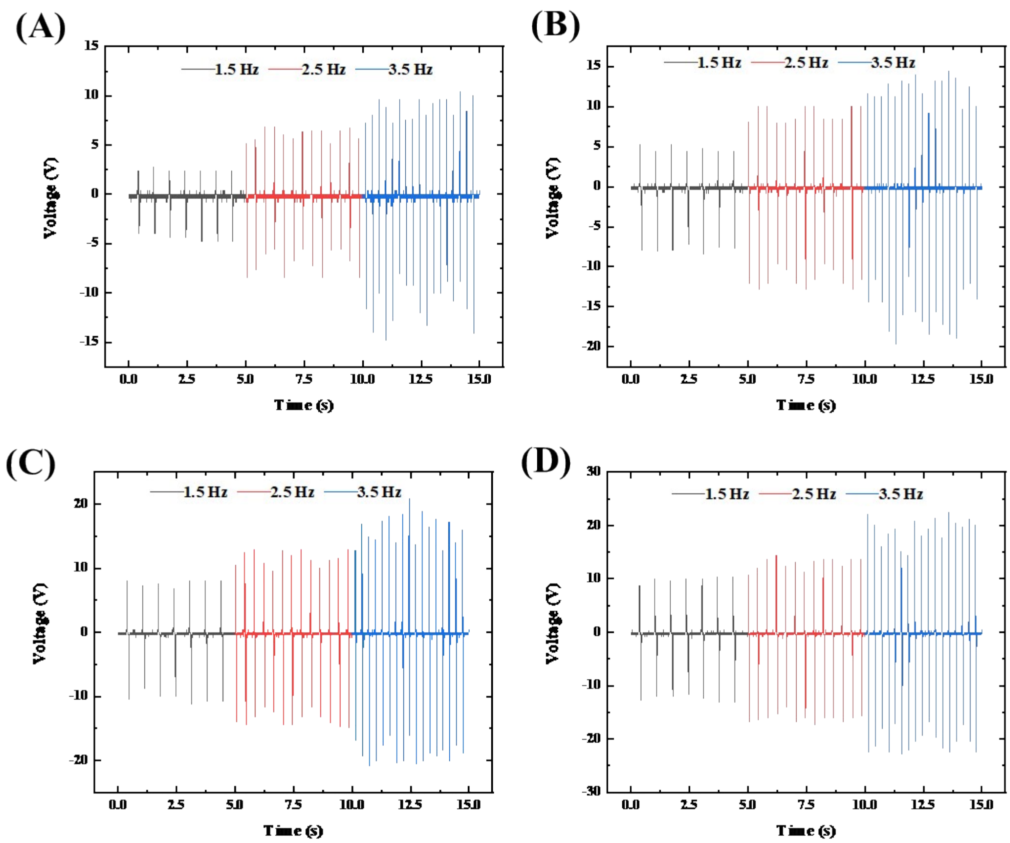

Figure S4 The output performance of AS-TENGs with an effective CE area of 3 cm ×3 cm triggered by (A) 10 N, (B) 20 N, (C) 30 N, and (D) 40N at 1.5 Hz, 2.5 Hz, and 3.5 Hz, respectively

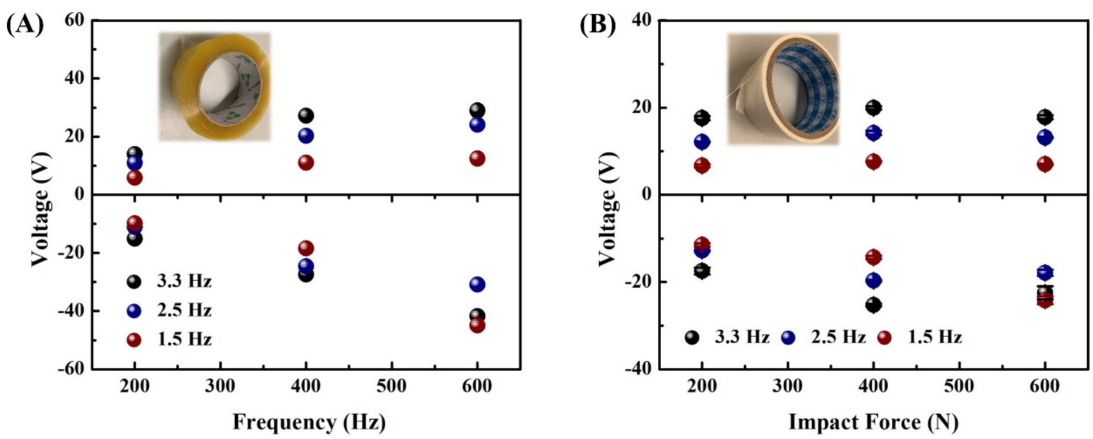

Figure S5 Output performance of AS-TENGs fabricated based on (A) transparent adhesive tape and (B) double-sided adhesive tape commercially available under different triggering forces and frequencies

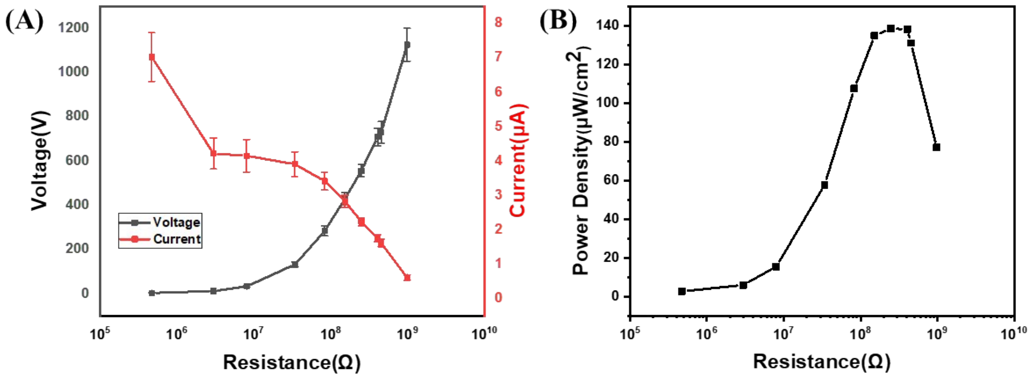

Figure S6 The influence of load resistance on the (A) output voltage, current and (B) power density of AS-TENGs