Improving energy conversion efficiency for triboelectric nanogenerator with

capacitior structure by maximizing surface charge density

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Fig. S1. (a) FE-SEM photograph of the PDMS maxed with GPs. (b) FE-SEM photograph of the GPs. (c) XRD patterns of PDMS film with different GP contents.



Fig. S2. (a) The maximum output current of the CFNG, (b) a partially enlarged view of (a) and lighted LEDs (c).



Fig. S3. The dependence of the output voltage, current and instantaneous power on the resistance of the external load with the CFNG devices made of different films, (a) pure PDMS, (b) PDMS@0.01 g GPs, (c) PDMS@0.05 g GPs, (d) PDMS@0.08 g GPs, (e) PDMS@0.1 g GPs, (f) PDMS@0.2 g GPs.



Fig. S4. The open-circuit voltage of CFNGs using the composite films with different GP contents.



Fig. S5. (a-b) The output current of CFNGs (size: $2 \text{ cm} \times 2 \text{ cm}$, thickness: 0.45 mm) at different frequencies of oscillation. The maximum output current is at 2.0 Hz.



Fig. S6. The schematic diagram of CFNG's cross section (a), the charge distribution and working steps (b) in one operating cycle (II-III-IV-V-II). Current-time pulse curve of one operating cycle of the device (c).

The working mechanism of the CFNG is as follows. In its initial state, there is no charge on any part of the CFNG, as is shown in Fig. S6a (I). When the upper electrode is pushed to contact the PDMS@GPs film, it causes the friction between the composite film and electrode. Because the tendency of aluminum to lose electrons is larger than that of PDMS, the PDMS@GPs film is negatively charged by rubbing against the electrode, resulting in a net negative charge density ($-\sigma_0$) on its surface and

a net positive charge density $(+\sigma_0)$ on the electrode (II). When the upper electrode separates from the composite film, the charge on the upper electrode transfers to the bottom electrode through the external circuit which generates a current (III). The current disappears when the electrode reaches the top position (IV). When the upper electrode gets close to the composite film, the charge on the bottom electrode transfers to the upper electrode through the external circuit which generates a current (V). The current disappears when the electrode reaches the bottom position (II). The whole cycle is II-III-IV-V-II. The output current corresponding to the pushing and releasing processes is shown in Fig. S6c.



Fig. S7. The way to form the gap between the top electrode and the PDMS film.

We insert the PET film with the thickness of 0.1 mm between the top electrode and the PDMS@GPs film to form the gaps. And the way of controlling the size of the gap is as follows: $n_1 = 0.2$ mm, $n_2 = 0.5$ mm, $n_3 = 0.8$ mm respectively corresponds to 2 layers of PET film, 5 layers of PET film, 8 layers of PET film. We firstly fixed the CFNG according to the following Fig.S7 when testing the device. Then, the PET films were pulled out of the CFNG, so the gap was formed. In this measurement, one of the Al foil electrodes is fixed on the hitting part of electric oscillator.



Fig. S8. Schematic diagram of the setup and measurement system for testing the performance of CFNG.

Table S1. The relative dielectric constant (ε_r) of the PDMS composite films mixed with different kinds of conductive carbon, graphite particles (GPs), graphenes (G) and carbon fibers (CFs).

| film | None | GPs | Gs | CFs |
|--------------|-------|-------|-------|-------|
| ${\cal E}_r$ | 3.000 | 2.973 | 2.732 | 2.686 |