Electronic Supplementary Information

Functional sponge-based triboelectric nanogenerators with energy harvesting, oil-water separating and multi-mode sensing performance

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Experimental section

Materials

Carbon iron (CI) particles (CN-type) with the diameter of about 7 µm were bought from BASF Aktiengesellschaft, Germany. Multi-walled carbon nanotubes (MWCNTs, 8-13 nm in diameter, 3-12 µm in length) were purchased from conductive materials of Heluelida Power Sources Co., Ltd, Xinxiang City, Henan Province, China. Silicone rubber Ecoflex 00-20 was provided by Smooth-On, PA, USA. Polydimethylsiloxane (PDMS) prepolymer and its crosslinking agent (Sylgard 184) were supplied by Dow Corning GmbH, USA. Sugar granules were commercial products.

Preparation of the conductive MWCNT/CI-PDMS composite

First, MWCNTs with a mass fraction of 2% were introduced into PDMS prepolymer, followed by stirring for 2.5 h. Then, CI particles of 50% and the crosslinking agent with ratio to the prepolymer of 1:30 were added, and the total mixture was homogeneously stirred for 0.5 h. Afterwards, it was degassed in a vacuum chamber for 10 min and vulcanized at 90 °C. Thus, the conductive MWCNT/CI-PDMS composite was yielded.

Fabrication of the flexible CI-Ecoflex sponge-based TENG

First, sugar granules were placed in a prepared mold using a facile wetting-evaporating process: spreading a layer of sugar granules with a thickness of 0.25 mm, and then spraying about 1 mL deionized (DI) water, followed by drying at 50 °C for 20 s. After repeating the process, the sugar-filled skeleton template was acquired. Next, Ecoflex and CI particles with a mass ratio of 1: 1 were poured to infiltrate the template, followed by degassing in a vacuum chamber for 10 min. Then, the mixture was cured for 4 h. Afterwards, the sugar template was dissolved in an ultrasonic cleaner with DI water, followed by drying process. Hence, the flexible CI-Ecoflex sponge with three-dimensional interconnected porous structure could be prepared. Finally, the sponge was assembled with a copper wire and un-vulcanized MWCNT/CI-PDMS composite. After being vulcanized at 90 °C, this novel triboelectric nanogenerator (TENG) was obtained.

Magnetorheological effect and oil absorbency of the CI-Ecoflex sponge

To investigate the magnetorheological property, the sponge was molded into a 1 mm (diameter) \times 20 mm (height) cylinder, and the testing gap was kept at 1 mm. The oscillation strain and frequency was set to 0.1% and 1 Hz, with a constant applied temperature of 25 °C. The magnetic field ranging within 0-1000 mT was provided by an external coil, and thus the magnetorheological effect was defined as

$$MR = \frac{G_s - G_0}{G_0} \times 100\%$$

in which G_0 and G_s were the initial and maximum storage modulus during the measurement.

To evaluate the oil adsorption capacity, the dried sponge was immersed in various oils/organic solvents for 30 min. After taking out, the oil was wiped from the surface by filter paper. Ultimately, the oil absorbency (M) was determined as follows:

$$M = \frac{m - m_0}{m_0} \times 100\%$$

where m_0 and m were the mass of the sponge before and after oil absorption, respectively. For accuracy, the average value was obtained after three individual determinations.

Characterization

The micro-morphologies were observed by a scanning electron microscope (SEM, Gemini 500, Carl Zeiss Jena, Germany). The magnetorheological performance was explored by a rheometer (Physica MCR 301, Anton Paar Co., Austria) with an external magnetic field system. The mechanical properties were studied using an electroforce dynamic system (TA ElectroForce 3220, TA Instruments) and a micro/nano-mechanical testing system (FT-MTA02, Femto Tools, Switzerland). The triboelectric performance was conducted using an oscillator (JZK-10, Sinocera. Piezotronics. INC, China), and the output electric signals were measured via digital multi-meters (DMM6001). To eliminate environmental and equipment noise, all voltage signals were processed by a band stop filter approach. A magnetic property measuring system

vibrating sample magnetometer (SQUID, Quantum Design Co., America) was utilized to measure the magnetic hysteresis loops. The magnetic flux density ranging from 0 to 540 mT was applied through a commercial electromagnet, of which the coil excitation current was regulated by a DC power supply (ITECH IT6724). The calculation model of the porous structure was established by DIGIMAT software, and the finite element simulation was calculated by COMSOL software.



Fig. S1 (a-d) Preparation process of the CI-Ecoflex-70 sponge, in which the high viscosity impeded its flowability.



Fig. S2 (a) The current to voltage (I-V) curves and (b) steady shear viscosities of a series of MWCNT/CI-PDMS composites with different MWCNT and CI contents. (c) The triboelectric output performance at 40.0 N of different MWCNT/CI-PDMS based TENGs.



Fig. S3 (a-d) Simulated electric potential distributions when an object was approaching the TENG.



Fig. S4 Real-time voltages of TENG subjected to varying frequency of 10-15 Hz.



Fig. S5 Various external vertical forces applied to the TENG for triboelectric performance measurement.



Fig. S6 Schematic diagram for monitoring human gaits.



Fig. S7 The calculated magnetic field monitoring sensitivity, which was defined as the increment of voltage caused by the change of unit magnetic flux density.

Movie S1 Lighting a bulb by the TENG.

The TENG device was used to charge the capacitor and then light the bulb via a full bridge rectifier circuit.

Movie S2 Powering a stopwatch by the TENG.

A commercial stopwatch was powered after the capacitor was fully charged by converted direct current (DC) output of the TENG.

Movie S3 Wireless passive sensing system.

The output signal of the passive sensation test was successfully received and transmitted to the mobile phone, with the aid of a self-made integrated wireless sensing system.