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Harvesting Water Drop Energy by a Sequential Contact-Electrification and Electrostatic-Induction Process

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In the past decades, global warming and climate change have become the most important environmental issues because of the rampant consumption of fossil fuels. As fossil fuels are a finite resource, the price will continue to increase as reserves dwindle. Therefore, increased efforts have been devoted towards renewable and low-carbon emission energy sources. The nanogenerator, which is a new invention that can harvest energy from different sources in the environment including mechanical vibration,[1,2] heat,[3,4] and wind,[5,6] is capable of fulfilling the above mentioned features and has captured the worldwide attention.

Contact electrification, also called triboelectrification, is a well-known phenomenon that occurs when two materials are brought into contact, and has been demonstrated in applications such as metal ions reduction,[7–9] electrostatic charge patterning,[10,11] chemical sensors,[12,13] and laser printing.[14] Recently, this phenomenon has been used to collect energy from environmental sources, in the form of the triboelectric nanogenerator (TENG).[15–18] The working mechanism of the TENG is based on a combination of contact electrification and electrostatic induction. Contact between two materials with different triboelectric polarity will cause surface electron/ion/material transfer and create an electric potential difference after separation. By cyclically contacting and separating materials with oppositely charged surfaces, electrons can be driven to flow through the external load and generate a continuous output. Normally, the TENG needs a relatively dry environment to provide a stable output,[19] as surface triboelectrification would be reduced owing to the presence of water. However, the water-related energy sources including ocean waves, waterfalls, and rainwater in the environment have abundant amounts of energy, which are inexhaustible and can be good alternatives to solar energy.[20–22] Therefor, it is our intention to explore a new prototype TENG that can convert the water-related energy into electricity. In our last study, we demonstrated that water itself could be one of the materials to generate triboelectricity. The contact electrification between water and insulating polymer films has been used to harvest wave energy and detect the temperature and alcohol concentration in water.[23]

A water drop carries two types of energy. The first is mechanical impact energy when it falls on a substrate. The second is electrostatic energy generated during the contact electrification process with air/pipes. Our current research is studying how to collect both of them, especially the latter. In 1867, the Kelvin water dropper was invented to collect electrostatic energy from water and illustrate the natural generation of electricity in atmospheric phenomena such as thunderstorms.[24] The Kelvin water dropper uses the initial instantaneous non-equilibrium charge distribution of falling water to generate an electric potential difference through electrostatic induction between two interconnected, oppositely charged systems. In this paper, we develop the water-TENG with a superhydrophobic micro-/nanostructured polytetrafluoroethylene (PTFE) surface to harvest the water-related energy from flowing water and water drop, which is substantially different from the working mechanism of the Kelvin water dropper. By studying the relationship between the motion of single water drop onto the water-TENG and the measured output, a sequential contact-electrification and electrostatic-induction process is proposed to describe the working mechanism of the water-TENG. The output of the water-TENG generated from a 30-μL water drop can achieve a peak voltage of 9.3 V and a peak current of 17 μA. A maximum output power of 145 μW is observed when the water-TENG is connecting to a load resistor of 5 MΩ. The water-TENG is also utilized to collect energy from flowing tap water from a kitchen faucet, and the output current and instantaneous power densities reach 1.5 μA cm^{-2} and 20 mW cm^{-2}, respectively. The output can be used to directly drive 20 light-emitting diodes (LEDs) instantaneotusly. Moreover, we design a system containing the water-TENG and a packaged contact-TENG to effectively collect electrostatic and mechanical energy from spray water drop. The rectified outputs have been demonstrated to charge commercial capacitors. Our study shows the great potential of using the water-TENG to harvest the energy from flowing water and raindrop.

The fabrication process of the superhydrophobic hierarchical structures is displayed in Figure S1.[25,26] First, microstructures were fabricated by blasting an Al foil with sand particles using compressed air. The sand-blasted Al foil was further anodizing in a 0.3 M oxalic acid solution to obtain an anodic Al oxide (AAO) template with nanometer-sized holes (**Figure 1**a). Then a PTFE solution was poured into the AAO template and any remaining air was removed from the

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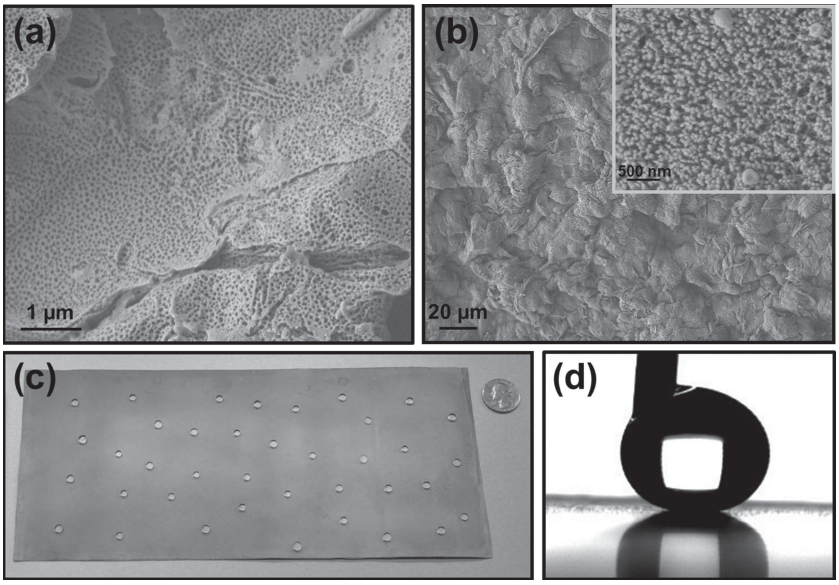

Figure 1. (a) SEM image of the AAO template with irregular microstructures and nanometer-sized holes. (b) SEM image of the prepared PTFE thin film with hierarchical micro-/nanostructures. Inset is an SEM image with higher magnification. (c) Photograph of a large-sized PTFE thin film (13 cm × 33 cm) with uniform superhydrophobicity. (d) Advancing contact angle of the PTFE thin film.

nanoholes through a vacuum process. After the curing at ambient temperature for one day, the solvent evaporated and a PTFE thin film with hierarchical micro-/nanostructures formed. Finally, the PTFE thin film was peeled off from the AAO template using a double-sided tape and attached to a poly(methyl methacrylate) (PMMA) substrate coated with a Cu electrode on the top to complete the water-TENG.

Figure 1b shows a scanning electron microscopy (SEM) image of the PTFE thin film. The surface of the PTFE thin film is composed of irregular microstructures, which is further covered by high-density nanorods. The mean length and diameter of the nanorods determined from SEM images are around 360 nm and 45 nm, respectively. We also demonstrated that the approach can be utilized to prepare large-sized PTFE thin films with a uniform superhydrophobic property (Figure 1c). This is very important for the application of this concept to harvest the water-related energy in the environment. The hydrophobicity of the PTFE thin film was investigated by measuring the contact angle of water drop on it (Figure 1d). Because the hierarchical micro-/nanostructures contain trapped air, it will reduce the actual contact area between the surface and water drop, making the surface superhydrophobic. The average measured contact angle of the PTFE thin film is 169° and can be determined as a superhydrophobic surface (>150°).[27]

The working mechanism of the water-TENG is based on the triboelectricity generated from the contact electrification process with air/pipes (**Figure 2**a) and/or the PTFE thin film (Figure 2b). The operation of the water-TENG will be explained as single-electrode mode. Figure 2a shows before the contact with the water-TENG, the water drop already contains triboelectric charges on its surface because of the friction with air/pipes. Previous studies have shown that when a water drop is falling from the sky or flowing through an insulating tube, triboelectricity will be

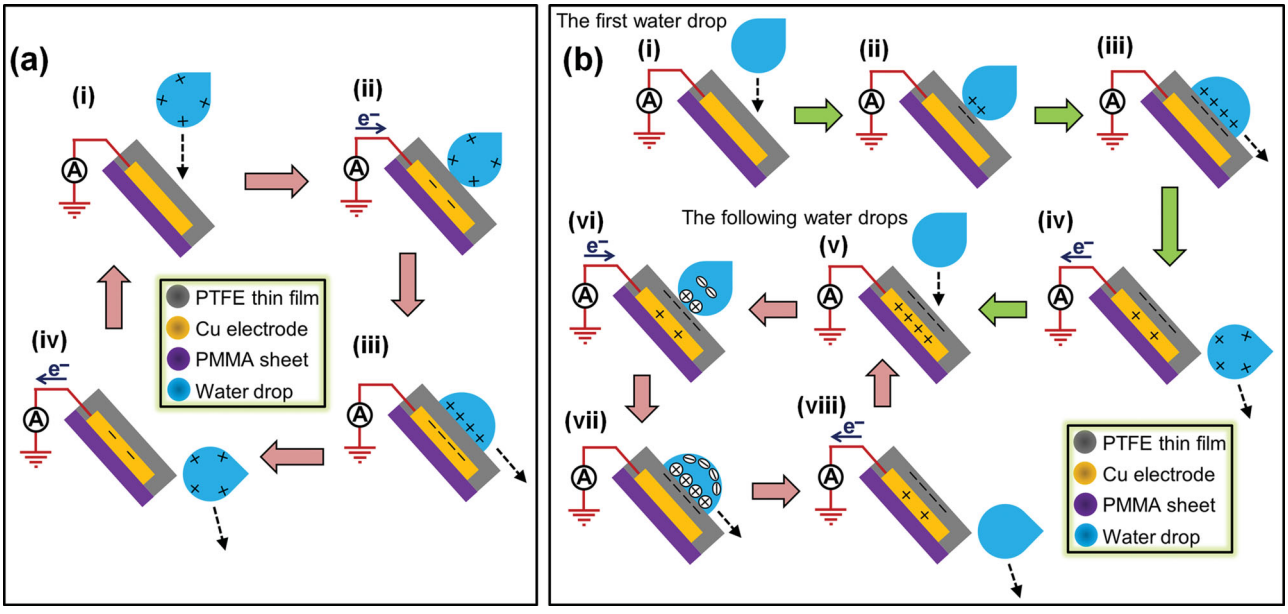

Figure 2. (a) Working mechanism of the water-TENG when the generated triboelectricity is dominated by the contact electrification process with the air/pipes. (b) Working mechanism of the water-TENG when the generated triboelectricity is dominated by the contact electrification process with the PTFE thin film.

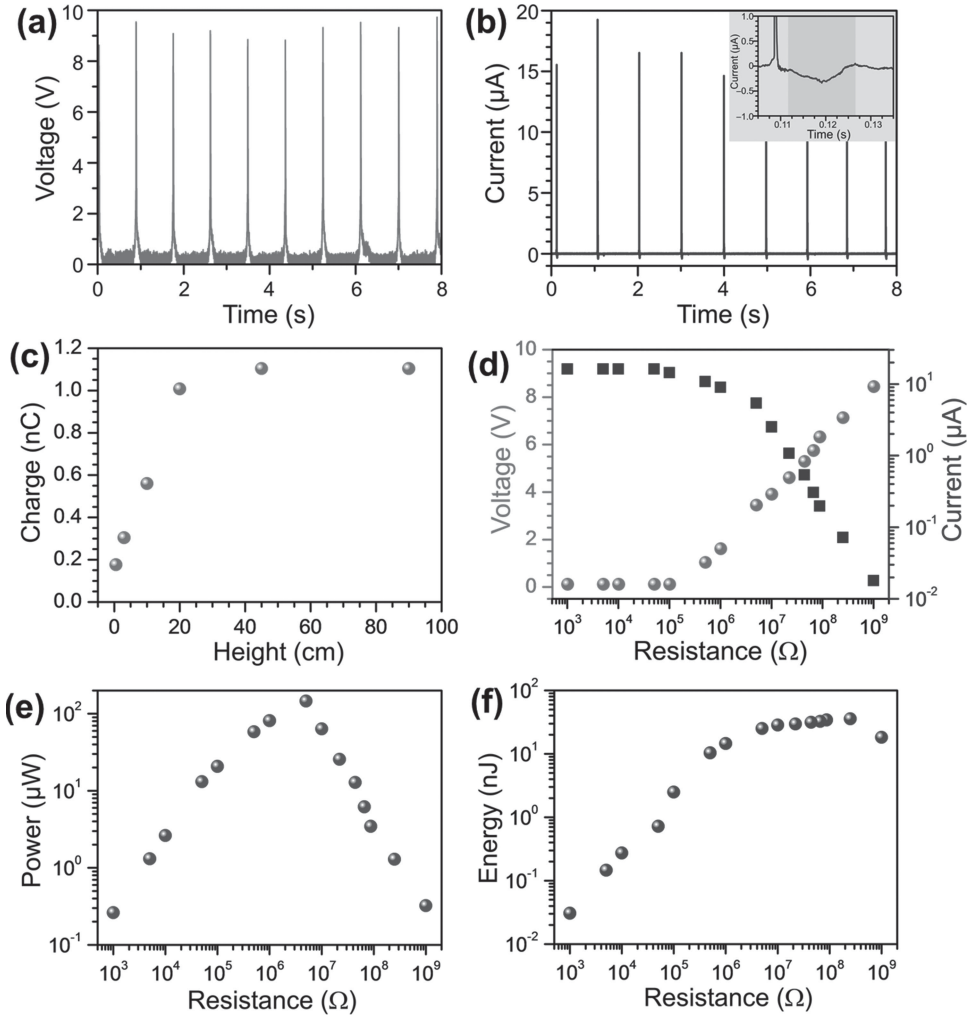

Figure 3. (a) Output voltage and (b) output current of the water-TENG generated from water drop. Inset of (b) is the magnified output curve in one cycle. The volume of each water drop is fixed at 30 μL and fell from a height of 90 cm and the water-TENG did not connect to external resistance when measuring the electrical output in (a) and (b). (c) Impact of water drop falling height on the inductively transferred charges of the water-TENG, which is proportional to the triboelectricity generated during the contact electrification processes with air and PTFE thin film. (d–f) Dependence of output voltage (d), output current (d), output power (e), and output energy (f) of the water-TENG on the resistance of external load.

generated and contributes to the charged surface of water drop.[28–32] For example, a surface charge density of 4.5 μC m^{-2} is measured from each water drop pipetted from a PTFE coating tip.[28] It is worth noting that the charge on the water drop could be positive or negative depending the counterpart the water drop interacts with. According to the data we measured in **Figure 3**, we use the example of positive charge on water drop to illustrate the process of converting electrostatic energy to electricity. As the positively charged water drop approaches the PTFE thin film (Figure 2a, ii), a positive electric potential difference is created between the Cu electrode and ground. In the short-circuit case, electrons are transferred from ground to the Cu electrode to balance the potential difference and finally reach equilibrium (Figure 2a, iii). This process produces an instantaneous positive current. The charge density on the drop surface affects the inductively transferred charges on the Cu electrode, which will determine the output of the water-TENG. Once the water drop moves off the surface of PTFE thin film, it creates a negative electric potential difference and therefore, electrons flow from the Cu electrode to ground (Figure 2a, iv), until a new equilibrium (Figure 2a, i) is reached. This process produces an instantaneous negative current. If water is periodically dropped and contacted with the PTFE thin film, an AC output can be continuously generated. To obtain the maximum electrical output, the water drop should be completely removed from the surface of PTFE thin film before the next one drops, necessitating superhydrophobicity.

Figure 2b represents the case that the triboelectric charges generated when the water drop falls on the PTFE thin film. It is known that when a water drop is dripping and sliding on a polymer surface, triboelectricity will be generated and contributes to the charged water drop and polymer surface.[33,34] When a water drop falls down and contacts with the PTFE thin film, the ionization of surface groups on PTFE will cause PTFE to be negatively charged[12,28] and cause a positively charged electrical double layer (EDL) on the contact surface of the water drop

to maintain electrical neutrality (Figure 2b, iii). As the water drop is leaving the PTFE thin film, a negative electric potential difference will be established between the Cu electrode and ground. In the short-circuit case, electrons are transferred from the Cu electrode to ground (Figure 2b, iv) and reach equilibrium (Figure 2b, v). This process produces an instantaneous negative current. Because the triboelectric charges on the PTFE thin film can be retained for a long time, once another water drop is falling to contact with the negative charged PTFE thin film, the negative charges on the PTFE will attract counter ions from the water drop to form another positively charged EDL, and establishes a positive electric potential difference. Therefore, electrons will flow from ground to the Cu electrode (Figure 2b, vi) until reaching a new equilibrium (Figure 2b, vii). This process produces an instantaneous positive current. When the water drop is leaving the PTFE thin film, a negative electric potential difference will be established between the Cu electrode and ground. Electrons are transferred from the Cu electrode to ground (Figure 2b, iv) and another new equilibrium is achieved (Figure 2b, v). Once the following water drops are contacting with the PTFE thin film periodically (Figure 2b, v-viii), a continuous ouput will be obtained.

To measure the electrical output of the water-TENG, we used a programmable syringe pump to control the volume of each water drop. The volume of each drop was fixed at 30 μL and fell from a height of 90 cm. To demonstrate future application, untreated tap water was chosen as the water source. Figure 3a shows a peak output of 9.3 V when contact is made. The output current reaches a value of 17 μA and exhibits AC behavior (Figure 3b), with an equal number of electrons flowing in the opposite direction in each cycle. The experimental data validates the working mechanism proposed in Figure 2. It is observed that the current peak corresponding to separation has a smaller magnitude but lasts longer than that for contact (inset in Figure 3b). This can be explained by the fact that separation is a slower process and thus contributes to a smaller but wider current signal.

Because the triboelectricity generated during the contact electrification process with air and the PTFE thin film will impact the output of the water-TENG, it is required to discuss the relationship between the triboelectricity and falling distance. By integrating the output current peak of the water-TENG obtained for different heights from the contact surface, we can deduce the inductively transferred charges on the Cu electrode, which is proportional to the triboelectricity generated during the energy harvesting process. It is clearly shown that the inductively transferred charges are increased when enlarging the falling distance, and finally saturate at 20 cm (Figure 3c), corresponding to the maximum output of the water-TENG that a water drop can provide. This verifies that triboelectricity does exist when the water drop travel through the air and contact with the polymer film, and also indicates that the concept can be applied to harvest the raindrop energy in the environment.

We then measured the electrical output of the water-TENG after connecting loads of different resistances while keeping the falling height constant at 90 cm. As shown in Figure 3d, when the resistance is below 0.1 MΩ, the voltage remains close to 0 and the current does not change significantly. When the resistance is raised from 0.1 MΩ to 1 GΩ, the voltage through the load increases, but the current across the load follows an opposite trend. Therefore, the instantaneous power on the load (Figure 3e) remains small when the resistance is below 0.1 MΩ and reaches a maximum of 145 μW at 5 MΩ for a single water drop. The output energy is also calculated by the time integral of I^2R (Figure 3f). The curve shows that the output energy increases with the resistance, and gradually saturates at a resistance of 22 MΩ (30 nJ), which is 3 orders of magnitude larger than that of 1 KΩ (0.03 nJ). After the calculation, we find the energy conversion efficiency from the water-related energy to electrical energy for single water drop is around 0.01% at a resistance of 22 MΩ. Although right now the energy conversion efficiency is small, we believe it will be improved by collecting the secondary energy of the water drop in the near future. The concept we have also demonstrated in Figure 5.

To demonstrate that this design can be used to harvest the water-related energy in the environment, flowing tap water from a kitchen faucet is first evaluated. The flow rate was set at around 30 mL/s and the distance between the water-TENG and faucet was 20 cm. This demonstration could be important because it represents not only in the future we can collect the wastewater energy from the environment in daily life but also we are moving forward about discovering and facilitating new renewable energy source. To highlight the potential application, the generated output now will be divided by the water-TENG dimensions (4 cm × 4 cm). **Figure 4**a shows the output current density of the water-TENG. It is noticed that the maximum output current density can reach 1.5 μA cm^{-2} and the signal varies due to the unstable flow of tap water from the faucet. The output signal drops to zero after we turn off the faucet, revealing the output is generated from the faucet flowing water. The AC output could be transformed to unidirectional pulse output by attaching a full-wave rectifying bridge (Figure 4b). The rectified output can be utilized to drive 20 commercial LEDs instantaneously (Figure 4c and Video S1) and charge a 33-μF capacitor (Figure 4d). The LEDs are lighted up as soon as the flowing water contacts with the water-TENG and totally go out when we turn off the the faucet. The instantaneous peak to peak power density is calculated as about 20 mW cm^{-2} (Figure S2) when the water-TENG was connected to a 5 MΩ load resistor. To effectively harvest the energy from flowing water, we further designed the water-TENG as a multilayer device (Figure S3a). Figure S3b shows that the double-layered water-TENG can provide twice the output compared to that of single-layered water-TENG (Figure 4a). When the superhydrophobic PTFE film was replaced with either a hydrophilic Nylon film or a smooth, untreated hydrophobic PTFE film, there was a noticeable decrease in output (Figure S4), showing the importance of superhydrophobic surface. In the cases of flowing water or spray water drop like raindrop, it is necessary to consider there could be more water remaining on the surface of the water-TENG during the energy harvesting process. Here we need to explain that under those conditions, the water-TENG can still generate output is because of the amount of water on the water-TENG surface is not constant. Like the water flowing from a kitchen faucet is not fully continuous. The water flow has some fluctuation and causes the amount of water on the water-TENG surface to be constantly changing. Besides, the superhydrophobic property of the

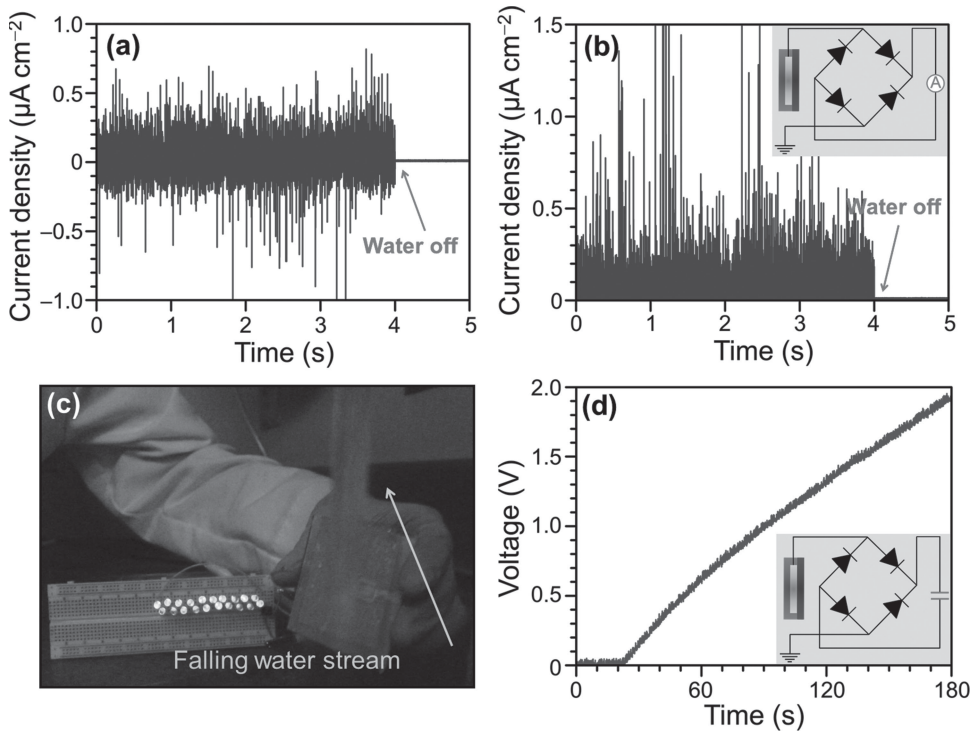

Figure 4. (a) Output current density of the water-TENG generated from flowing tap water. (b) The AC output transformed to unidirectional pulse output by a full-wave rectifying bridge. (c) Photograph of the water-TENG used as a direct power source to light up 20 LEDs. (d) The rectified output used to charge a commercial capacitor of 33 μF. Insets of (b) and (d) are the sketches of the corresponding circuit connection polarities.

water-TENG also enables the change of water amount on the surface.

Water drop energy such as raindrop energy is another interesting energy source for low power applications such as sensors and portable electronics.[35] Recently, piezoelectric materials-based harvester composed of polyvinylidene difluoride (PVDF) and lead zirconate titanate (PZT) have shown the potential to convert mechanical impact energy of water drop into electricity.[36,37] However, there is still no TENG designed for collecting of raindrop energy. Herein, we not only show that the water-TENG can be applied to harvest water drop electrostatic energy but also develop an integrated system including another packaged contact-TENG to collect the water drop energy more effectively (**Figure 5**a). The packaged contact-TENG was purposely used to harvest the water drop mechanical impact energy. Besides that, we also increased the water-TENG dimensions to 15 cm × 15 cm in order to reveal our approach with the great potential to harvest raindrop energy. A household shower jet system was utilized to spray water drops. Figure 5b displays the maximum output current density the water-TENG can achieve is 0.15 μA cm^{-2}. The rectified output also successfully powered 20 commercial LEDs instantaneously (Figure 5c) and charged a 33-μF capacitor (Figure 5e). Also from Figures 5d and 5e, we can observe that the packaged contact-TENG also successfully collects mechanical impact energy of spray water drops. We also determined that the generated output of the water-TENG will not be impacted by a higher water temperature of 65 °C (Figure 5f).

In summary, a prototype water-TENG with superhydrophobic and self-cleaning features is invented and demonstrated with the potential to harvest the water-related energy in the environment. By observing the relationship between the motion of single water drop on the TENG and the generated output, we also successfully explain the working mechanism of the water-TENG. The generated output from a 30-μL water drop can reach a peak voltage of 9.3 V and a peak current of 17 μA, demonstrating its applicability to real life. The water-TENG was utilized to collect the energy of flowing tap water from a kitchen faucet, and the output current and instantaneous power densities of 1.5 μA cm^{-2} and 20 mW cm^{-2}, respectively, have been achieved, which can directly drive 20 LEDs. Moreover, we have developed an integrated energy collection system consisting of the water-TENG and a packaged contact-TENG to effectively collect the water drop energy. The rectified outputs have also been demonstrated to charge commercial capacitors. All these results clearly show that the novel concept and design of the water-TENG will serve as the stepping stone for future related TENG studies and inspire the development of TENG toward discovering and facilitating new renewable energy sources from the environment in daily life. Furthermore, the water-TENG can serve as a sensor to detect water/liquid leakage from a container/pipe.

Experimental Section

Water-TENG Fabrication: The fabrication process of the water-TENG is starting from the preparation of PTFE thin film with superhydrophobic hierarchical structures, which is illustrated schematically in Figure S1. First, an AAO template with hierarchical micro-/nanostructures is needed. The microstructures were formed by blasting an Al foil with sand

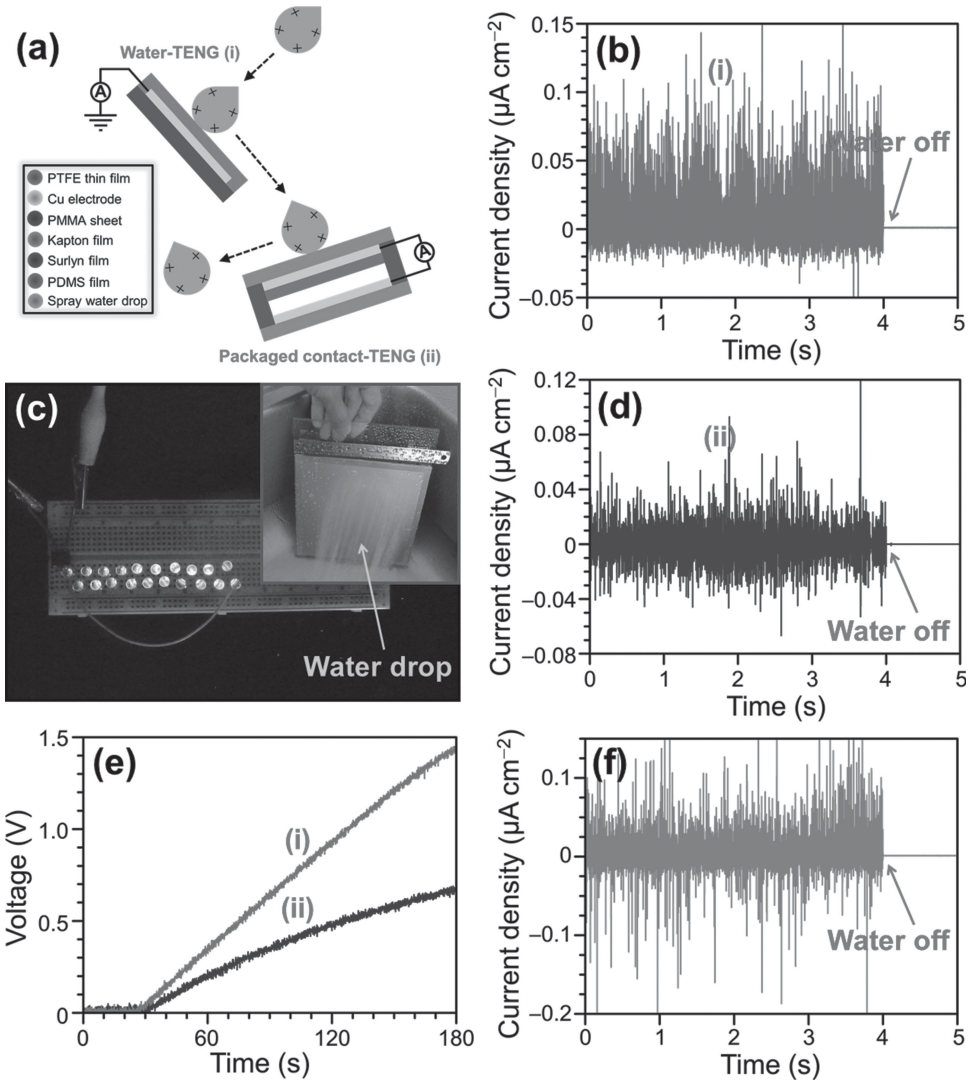

Figure 5. (a) Schematic diagram of the integrated TENG system for the spray water drop energy harvesting. The integrated TENG system is composed of the water-TENG (i) and a packaged contact-TENG (ii). (b) Output current density of the water-TENG for primary energy harvesting of spray water drop. A household shower jet system was utilized to imitate the spray water drop. (c) Photograph of the water-TENG used as a direct power source to light up 20 LEDs. (d) Output current density of the packaged contact-TENG for the secondary energy harvesting of spray water drop. (e) Both the rectified outputs used to charge commercial capacitors of 33 μF. (f) Output current density of the water-TENG generated from the spray water drop at a temperature of 65 °C.

particles (diameter around 30 μm) using compressed air (4 kgf cm^{-2}). Then the sand-blasted Al foil was put in an oxalic acid solution (0.3 M) as the anode. And another flat Al sheet was used as the cathode. The distance between those two electrodes is 5 cm. The operating voltage was set to 40 V by using a computer-interfaced power supply. During the anodization process, the solution was maintained at a temperature of 15 °C by a circulator and an AAO template with nanometer-sized holes was obtained (Figure 1a). After cleaning the AAO template with water, a commercial PTFE precursor (AF 601S2, 6 wt%, DuPont) was poured into the AAO template and a vacuum process was applied to remove the air remaining in the nanoholes. After curing at ambient temperature for one day, the solvent was evaporated, leaving a PTFE thin film with hierarchical micro-/nanostructures. Cu thin films (thickness around 100 nm) were deposited on PMMA substrates by a RF magnetron sputtering deposition system. The effective dimensions of the water-TENG discussed in this paper were 4 cm × 4 cm and 15 cm × 15 cm. Conducting wires were connected to the Cu thin films as leads for subsequently electrical measurements. Finally, the prepared PTFE thin film was peeled off from the AAO template by using a double-sided tape and bonded to the top of Cu thin film as a complete the water-TENG. The hydrophilic Nylon film and hydrophobic PTFE film were purchased from American Durafilm.

Packaged Contact-TENG Fabrication: The packaged contact-TENG fabrication starts with a PDMS film with a patterned pyramid array (Figure S5). Firstly, a Si wafer was molded using photolithography. Then the wafer was etched through a dry etching process, resulting in the Si wafer mold with notched pyramid features. After cleaning with acetone and ethanol, the prepared Si wafer mold was treated with chlorotrimethylsilane to avoid the adhesion between PDMS and Si in the next step. PDMS mixture that consists of PDMS elastomer and cross-linker in a 5:1 ratio was spin-coated (1000 rpm, 1 min) on the Si wafer mold and then incubated at 60 °C. After 24 h, a uniform PDMS film with patterned pyramid array was formed. Next, 100 nm of Cu thin films were deposited on two Kapton substrates (127 μm, DuPont) by a RF magnetron sputtering deposition system. Conducting wires were connected to the Cu thin films as leads for subsequent electrical

measurements. The formed PDMS film with patterned pyramid array was peeled off from the Si wafer mold and then placed onto a Cu thin film-coated Kapton substrate with uncured PDMS mixture on top, then incubating at 60 °C for another 24 h. Finally, those two Kapton substrates were assembled by using a 60-μm-thick hot-melt ionomer film (Surlyn 1702, DuPont) under heating at 150 °C for 10 min and a fully packaged contact-TENG was obtained.

Characterization: A Hitachi SU8010 field emission scanning electron microscope (SEM) was used to measure the size and shape of PTFE hierarchical micro-/nanostructures and the pore diameter of AAO template. For the measurement of typical electrical output of the water-TENG, a programmable syringe pump (PHD 2000, Harvard Apparatus) was applied to control the volume and dropping speed of each water drop. The volume of each water drop is fixed at 30 μL and the dropping speed is around 1 m/s when the water drop is falling from a height of 90 cm. The electrical outputs of the water-TENG were measured using a programmable electrometer (Keithley Model 6514) and a low-noise current preamplifier (Stanford Research System Model SR570).

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements

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