Nano Energy (2014) 10, 37–43

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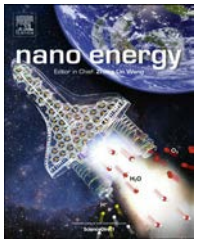

RAPID COMMUNICATION

Enhancing the performance of triboelectric nanogenerator through prior-charge injection and its application on self-powered anticorrosion

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Received 11 July 2014; received in revised form 21 August 2014; accepted 28 August 2014
Available online 6 September 2014

KEYWORDS
Energy harvesting;
Nanogenerator;
Charge injection;
Self-powered system;
Anticorrosion

Abstract
Corrosion is a large damage for industry, infrastructure and transport equipments such as vehicle and freighter. If the energies in environment can be harvested into electricity for the prevention of corrosion, it will be very valuable. Here, we enhanced the property of triboelectric nanogenerator and designed a self-powered anticorrosion system to protect iron from corrosion. Nanostructures fabricated through a simple method and prior-charge injection into the space between the surface of the friction layers and the electrodes were used to greatly enhance the property of triboelectric nanogenerator. The charge density was increased by 48% owing to the nanostructure and further increased by 53% owing to the prior-charge injection process. The output open-circuit voltage reached 1008 V, the short-circuit current density reached 32.1 mA/m^2 and the charge density reached 121 μC/m^2. On the basis of this high performance TENG, we developed a self-powered anticorrosion system to successfully protect a piece of iron sheet soaked in saline water from corrosion.

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http://dx.doi.org/10.1016/j.nanoen.2014.08.017
2211-2855/

Introduction

Nanogenerator (NG) is a kind of energy converter that converts mechanical energy in the environment into electric energy to power some micro/nano devices [1–5]. Recently, triboelectric nanogenerators (TENGs) have attracted much attention for high performance, durable, little maintenance and low cost [6–8]. A general TENG is made of two pieces of flexible substrates covered by materials with different triboelectric characteristics. Triboelectric charges with opposite signs are generated on the surface of the friction layers of the device by triboelectrification, and mechanical movement causes the polarization of the tribocharges, resulting in the output of electric power. So, charge density is one of the most conclusive parameters that determine the performance of a TENG. In previous works, two methods have been used to improve the charge density, using materials easier to gain/loss charges [4] and fabricating micro/nanostructures on the friction layers [5,9]. All their works increased the charge density on the surface of the friction layers and improved the performance of the TENG greatly, exhibiting wide application prospect. However, the long-term development of TENG will be limited by studying on increasing the charge density only on the surface of the friction layers. It is beneficial for enhancing TENG's performance to make full use of all the space, not only the surface but also the space between the surface and the electrodes in TENG. Therefore, we expect that, if charges can be injected into TENG directly, the performance of TENG will be improved significantly.

With the development of TENG, it has been applied in many fields such as sensors [10,11], electronics [12,13] and electrochemistry [14]. When the performance of TENG is enhanced to higher level, it is possible to be further applied in more fields even in engineering field. Materials corrosion is a large problem exists almost in all fields of national economy [15] such as energy, transportation, machinery, chemical industry. Many methods, such as physical anticorrosion [16,17], chemical anticorrosion [18,19] and electrochemical anticorrosion [20,21], have been developed to prevent corrosion. Comparing with other methods, the impressed current cathodic protection [20] is the most durable one because it does not cost other materials, but the constantly electric energy consuming leads to the problem of high energy consumption. Developing a self-powered anticorrosion technology will turn over a new leaf for the field of anticorrosion.

In this paper, we explored a very simple and cheap method to fabricate nanostructures on friction layers, and further introduce a new method which injects charges directly into the space between the surface of the friction layers and the electrodes. The charge density was increased by 48% owing to the nanostructure and further with additional 53% increase owing to the charge injection process. A new TENG with high performance was developed. The open-circuit voltage reached 1008 V and the short-circuit current reached 51.4 μA, corresponding to current density of 32.1 mA/m^2. Furthermore, we demonstrated a self-powered anticorrosion system (SPAS) on the basis of the impressed current cathodic protection using the high performance TENG as the power source. A piece of iron sheet from rusting in simulated seawater was protected successfully. The SPAS exhibits a good prospect to protect materials from rusting with low energy cost.

Material and methods

Preparation of polyamide (Nylon) and Poly (vinylidene fluoride) (PVDF) solutions

2 g Nylon was added into 18 g formic acid in a 50 mL triangular flask. The solution was stirred for 5 min to dissolve Nylon completely. 3.75 g PVDF was added into 8.5 g N.N-dimethylacetamide (DMAC) and 12.75 g acetone in a 50 mL triangular flask. The solution was stirred at 60 °C for 30 min and cooled to room temperature to dissolve PVDF completely. All the reagents above were analytically pure and used without any further purification.

Fabrication of TENG with nanostructures

Nylon and PVDF solutions were spin-coated on two pieces of Polyethylene terephthalate (PET) film, respectively, and Ag electrodes with the size of 4 cm × 4 cm were deposited on the other side of the PET films by magnetron sputtering. The PET films were fixed together at two edges with electrodes outward. The device was heated to 80 °C under bending state and then cooled down to make the device arch-shaped. The thickness of the TENG is 400 μm.

Method of prior-charge injection

The TENG was connected to a high voltage DC power, with the positive end connected with the electrode on the PET sheet with PVDF film, and negative end connected with the other electrode. The device was pressed to make the two films contact with each other and 10 kV (25 V/μm) voltage was applied on the device. After one hour, the device was released and the voltage was removed.

Results and discussion

The performance of TENG before charge injection

Figure 1a sketches the structure of the TENG and Figure 1b shows the TENG's photograph. As explained in previous studies [14], the TENG works on the coupling of contact electrification and electrostatic induction. For our TENG, Nylon and PVDF were selected as the friction because they are low cost, durable and easy to fabricate, meanwhile they have large difference in the ability of trapping electrons. When the TENG was pressed, Nylon film and PVDF film contacted and rubbed against with each other. Electrons transferred from Nylon to PVDF subsequently, which generated positive charge on the surface of Nylon film and negative charge on the surface of PVDF film according to the triboelectric series. As shown in Figure S1, when the device was pressed, the PVDF film with negative charge contacted with Nylon film with positive charge, leading to the potential of the electrode on the substrate with PVDF rise while the potential of other electrode drop. The potential difference between the two electrodes drove the charge flow forward. When the pressure released, the potential of the electrode on the substrate with PVDF dropped while the potential of other electrode raised, leading to the charge flow back.

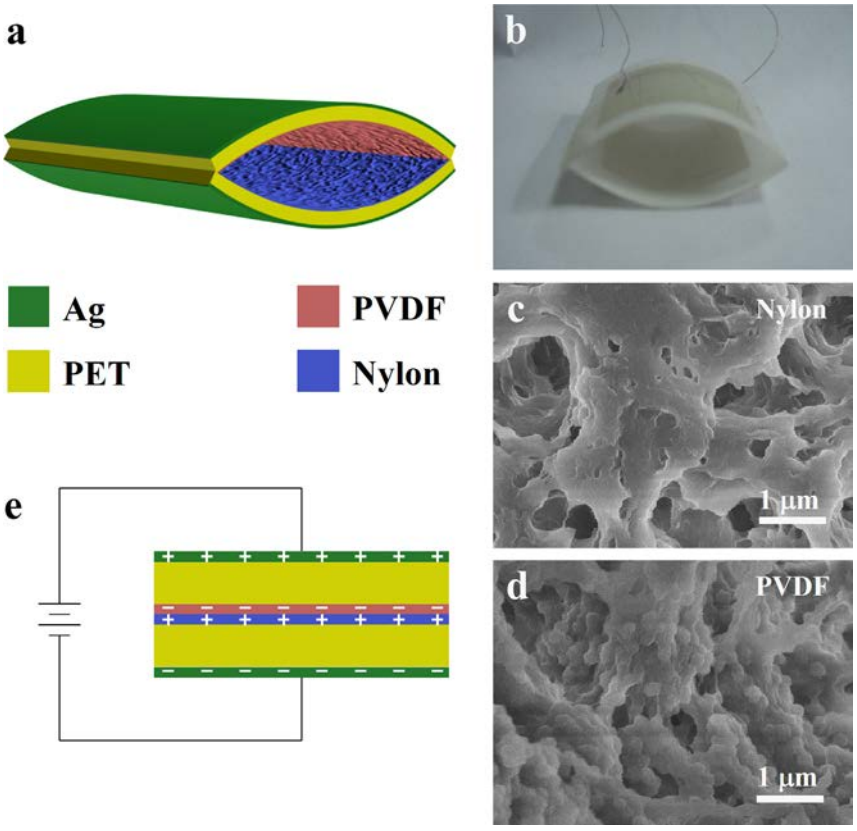

Figure 1 Fabrication of the high performance TENG. (a) Structure of the high performance TENG. (b) Photograph of the high performance TENG, the side length of the electrodes in the image is 4 cm. (c, d) Top view SEM image of the Nylon and PVDF film. (e) Schematic image of the equipment used for charge injection.

Increasing the charge density of TENG is the most important way to improve the performance of a TENG. In this work, we tried to increase the charge density by two ways. Previous studies have demonstrated that micro/nanostructures such as nanowires, nanorods, nanoparticles existed on the surface of a TENG contributed greatly to higher performance of a TENG [5-9,14]. In our work, nanostructures were made on the surface of Nylon and PVDF film spontaneously when they were spin-coated on the PET film as the solvents evaporated rapidly. Scanning electron microscopy (SEM) images of the nanostructures were shown in Figure 1c, d. We make a TENG with the same structure except for smooth Nylon and PVDF surface (Figure S2a, b) in order to check the effect of the nanostructure. The TENG with/without nanostructures was driven by a linear motor with the same amplitude and frequency. As shown in Figure S2c, d, the short-circuit current of the TENG with smooth surface was 11.1 μA, corresponding to current density of 6.9 mA/m^2, and the charge flowing through the circuit per peak was 77 nC, corresponding to charge density of 48 μC/m^2. Here, the charge density was calculated by dividing the charge flowing through the circuit per peak by the area of the electrode. For the TENG with nanostructures, the corresponding values were 15.4 μA, 9.6 mA/m^2, 114 nC and 71 μC/m^2, indicating that the charge density was increased by 48% owing to the existence of nanostructures on the surface of the friction layers. Comparing with other fabrication processes of micro/nanostructures shown in previous works [5,7,12], this process is convenient and low cost.

Nanostructures on the surface enlarge the surface area of the film and enhance the rubbing between the friction layers. Thus, the charge density on the surface of the friction layers is increased and the performance of the TENG is improved effectively. However, the space between the surface of the friction layers and the electrodes has not been utilized in previous works. If we can take advantage of this space, the output power of the TENG will be further enlarged to a new level.

Enhancing the TENG's performance through prior-charge injection

Our second method was making a design to utilize this ignored space by injecting charge into the films. Figure 1e shows the TENG's prior-charge injection. The process of charge injection can be explained by dielectric breakdown [22,23]. The dielectric between electrodes tends to breakdown when the TENG is under pressed and a sufficient high direct electric field is applied. The air gap between Nylon and PVDF will preferential breakdown because the dielectric constant of the air gap is the smallest. Subsequently, the ionized positive charge and negative charge will be injected into the friction layers or even into the PET films separately due to the direction of the external electric field. Thus, the space in TENG is fully used. The injected charge will work along with the triboelectric charge to improve performance of the TENG. This method could be

applied to the TENGs introduced in most previous works to increase their charge density [4-9,12-14].

Figure 2 shows the effect of the injected charge on the performance of nanogenerator. We measured the open-circuit voltage and short-circuit current of a TENG without injected charge (State 1), with positive charge injected into the film with PVDF (State 2) and with negative charge injected into the film with PVDF (State 3) driven by a linear motor under the same frequency and amplitude. The charge distribution of these states is shown in Figure 2a. The output open-circuit voltage was 584 V for State 1 (Figure 2b I), −214 V for State 2 (Figure 2b II) and 1008 V for State 3 (Figure 2b III). And the output short-circuit current was 24.5 μA for State 1 (Figure 2c I), −7.9 μA for State 2 (Figure 2c II) and 51.4 μA for State 3 (Figure 2c III), corresponding to current densities of 15.3 mA/m^2, −4.9 mA/m^2, 32.1 mA/m^2. Here, the negative value indicates the reversed voltage or current direction at the same work stage. The charge flowing through the circuit per peak under the three states were 126 nC, −51 nC and 194 nC, corresponding to charge densities of 79 μC/m^2, −32 μC/m^2 and 121 μC/m^2. The voltage, current and charge density were increased for 73%, 110% and 53%, respectively, indicating that the charge was injected into the films successfully and the injected charge contributed to improving the performance of the TENG. For our TENG, the short-circuit current of State 2 reduced with the working time, which further confirms the existence of the injected charge as shown in Figure S3.

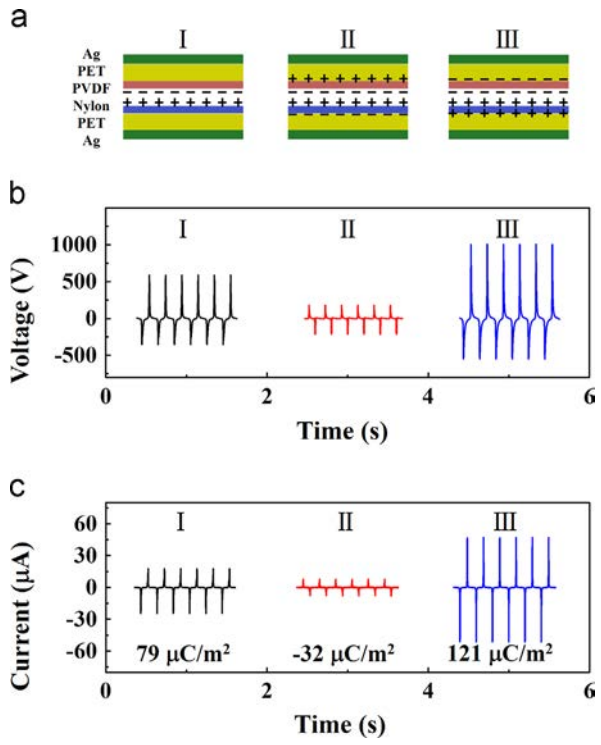

Figure 2 The TENG's performance at different charge injection states. In this figure, (I), (II) and (III) indicate the state without charge injection, the state with positive charge injected into the film with PVDF and the state with negative charge injected into the film with PVDF film, respectively. (a) Charge distribution of the three states after the device working for some time. (b) Open-circuit voltage of the three states. (c) Short-circuit current of the three states.

Further study on prior-charge injection parameters

Figure 3a shows the performance of the TENG with different charge injection time by figured out the charge density under these states. The charge density increased rapidly from 91 μC/m^2 to 144 μC/m^2 under the voltage of 10 kV (25 V/μm) in the first 2 min and reached a stable value of 148 μC/m^2 after 4 min. The charge density changing with the injected voltage was shown in Figure 3b. Here, each voltage was added for one hour to reach the saturation charge density. We can clearly see that the charge voltage began to increase at 2 kV (5 V/μm) and reached a stable value at 7 kV (17.5 V/μm). Therefore, we can conclude that the highest performance of TENG could be obtained by adding an electric field higher than 17.5 V/μm for more than 4 min.

Application on self-powered anticorrosion system

In consideration of the superior performance of the TENG after charge injection, it is expected that our TENG can be further applied in more aspects even in engineering field such as anticorrosion. In this work, we tried to solve some engineering problem by this TENG. Corrosion exists everywhere and always causes serious problems. Corrosion of materials will lead to the huge waste of natural resources and of course, the huge economic losses. What's more, great security threats will be brought by corrosion, so anticorrosion is an urgent problem to solve. The impressed current cathodic protection is one of the methods which are widely used in the anticorrosion of steel. In seawater, electrochemical corrosion is a main process in marine corrosion. Fe will be oxidized to Fe (II) with the standard electrode potential of −0.44 V. By adding a negative voltage on Fe, this electrode process will be prevented. According to the international standard ISO 15589-2:2012(E), the potential difference between saturated calomel electrode and Fe must be higher than 0.88 V to prevent Fe from rusting. As there is a constant current flowing through the circuit, this method makes considerable energy expenditure every year. In order to solve this problem, we try to make an SPAS on the basis of the impressed current cathodic protection with a TENG as the power source. In this case, the TENG must be durable enough to work continuously in a long term, and can provide a constant current and voltage higher than the critical value at a wide working frequency. Figure 4a showed the TENG's short-circuit current under 1-10 Hz working frequency while Figure 4b showed the peak value of the short-circuit current and the corresponding charge density changes with the working frequency. The short-circuit current of the TENG was basically proportional to the working frequency for the TENG providing equal charge per peak despite the working frequency. In the SPAS, the TENG should be connected to a rectifier bridge and a capacitance to supply a constant current and voltage, as the device provide equal charge per cycle at different working frequency, the current was proportional to working frequency, so the device could provide enough energy at working frequency higher than a critical value. Moreover, the long team measurement showed the short-circuit current of the

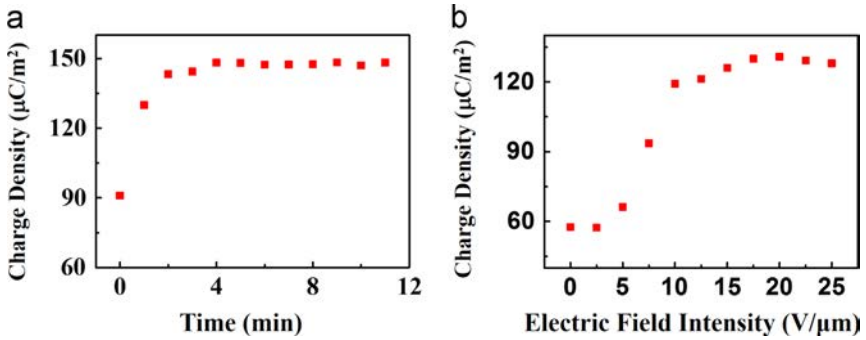

Figure 3 The TENG's performance changes with charge injection parameter. (a) Charge density changes with charge injection time under 10 kV injection voltage (25 V/μm). (b) Charge density changes with charge injection electric field intensity with 1 h injection time. The charge density is calculated by the short-circuit currents.

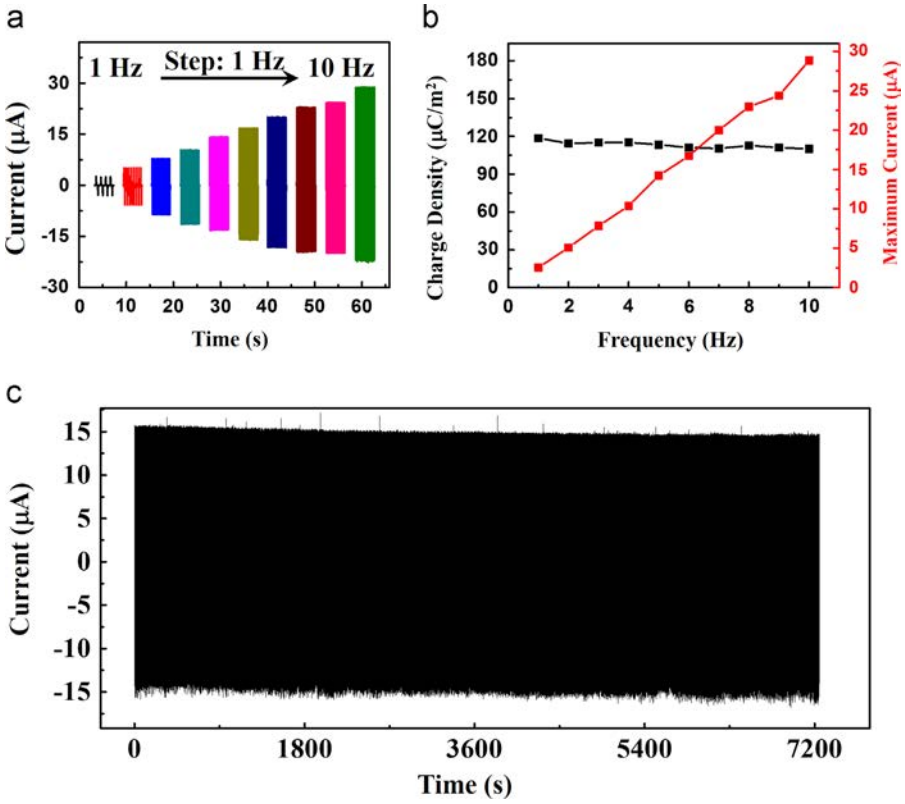

Figure 4 Stability of the high performance TENG. (a) Short-circuit current of the TENG working at frequency ranges from 1 Hz to 10 Hz, and the frequency difference between two neighboring curves is 1 Hz. (b) Peak short-circuit current and output charge per pulse of the TENG at different working frequency. (c) Short-circuit current of the TENG working continuously for 2 h.

TENG did not change in two hours at 5 Hz working frequency (shown in Figure 4c). These results indicated that the TENG can meet all the requirements discussed previously well.

Figure 5a sketches the design for SPAS in our work. The TENG was firstly connected to a rectifier bridge and then connected in parallel with a 100 μF capacitor as the power source aiming to provide constant voltage and current after the SPAS works stably. In order to keep the electrode potential stable, the power source was connected with a three-electrode system soaked in simulated seawater, 3.5% sodium chloride solution. The system contained an saturated calomel electrode, a carbon electrode and an iron sheet with a diameter of 3 mm working as the reference electrode (the iron sheet was polished by sand paper and one side of the iron sheet was exposed to solution), the counter electrode and the working electrode, respectively. As shown in the schematic image, the reference electrode was connected to the positive pole of the power source together with the counter electrode providing a stable electric potential relative to the solution (the salt bridge was used to limit the current flowing through the reference electrode), and the iron sheet was connected to the negative pole of the power source. As discussed previously, the voltage between the reference electrode and the iron sheet should be higher than 0.88 V in order to protect the iron sheet from corrosion, which was real-time measured in SPAS.

In the experiment, the TENG was driven by a linear motor with 7.5 Hz working frequency, which was a little larger than the critical value. Before the iron sheet was soaked into the

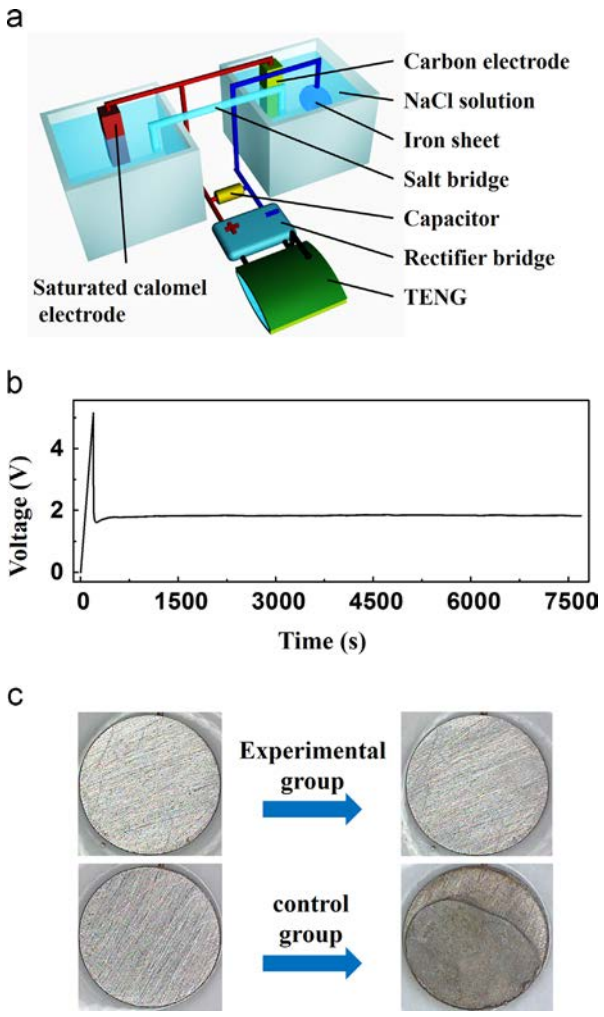

Figure 5 The SPAS based on the high performance TENG. (a) Schematic image of the SPAS. (b) The voltage between the iron sheet and the carbon electrode. (c) Iron sheets before and after soaked in the solution for 2 h. The experimental group indicates the iron sheet connected in the system while the control group indicates the iron sheet does not connected in the system.

solution, the TENG was working for 3 min to storage some charge in the capacitor, in order to keep the voltage between the reference electrode and the iron sheet should be higher than 0.88 V throughout the experiment. The iron sheet was soaked into the solution as the experimental group after 3 min while another iron sheet without connecting into the circuit soaked into the solution at the same time as the control group. The voltage decreased from 5.1 V to 1.6 V rapidly after the iron sheet soaked into the solution because the electrical double layers formed near the three electrodes in the solution, which consumed some charge in the capacitor. Then it stabilized to 1.8 V in 5 min (shown in Figure 5b). In the control group, a small rust spot can be observed on the iron sheet after 15 min, the area of the rust spot on the iron sheet in the control group enlarged gradually, and the iron sheet rusted seriously on most area of the iron sheet after 2 h. While in the experimental group, the iron sheet did not change in the whole process. The optical images of the two iron sheets before and after soaked in the solution for 2 h were shown in Figure 5c. These results indicated that the SPAS works effectively and can be further used on steamers, offshore pipelines and so on, exhibiting a good application prospect in engineering field.

Conclusions

In summary, we fabricated a new kind of TENG with high performance through two steps. Nanostructures fabricated simply by spin-coating method improved the charge density of the TENG by 48%, and charge injected by high voltage polarization further increased the charge density by another 53%. The output open-circuit voltage reached 1008 V and the short-circuit current reached 51.4 μA, corresponding to current density of 32.1 mA/m^2. The corresponding charge density reached 121 μC/m^2. This TENG is easy to fabricate, low cost, durable and can be commercialized. Prior-charge injection is a universal method to make full use of the space in the device and this method could be used on TENGs introduced in most previous works. On the basis of this high performance TENG, we designed an SPAS and protected a piece of iron sheet soaked in simulated seawater from corrosion successfully. The SPAS exhibits a good prospect to protect materials from rusting with low energy cost and it is the first approach to use the TENG in engineering field.

Acknowledgments

We sincerely thank the financial support from National Natural Science Foundation of China (NSFC) (No. 51322203, 51472111), Fok Ying Tung Education Foundation (No. 131044), PCSIRT (No. IRT1251), and the “thousands talents” program for pioneer researcher and his innovation team, China. The Fundamental Research Funds for the Central Universities (No. lzujbky-2014-m02, lzujbky-2013-k04, lzujbky-2013-34, lzujbky-2013-230), National Natural Science Foundation of China (NSFC) (No. 51302120), Scholarship Award for Excellent Doctoral Student granted by Ministry of Education, Scholarship Award for Excellent Doctoral Student granted by Lanzhou University.

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at http://dx.doi.org/10.1016/j.nanoen.2014.08.017.

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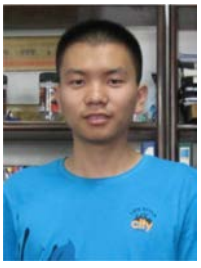

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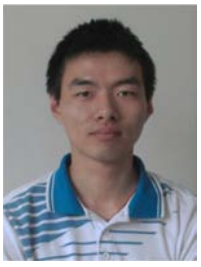

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