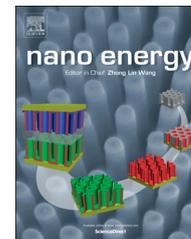


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RAPID COMMUNICATION

Self-powered cleaning of air pollution by wind driven triboelectric nanogenerator

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Abstract

Air pollution is one of the major challenges faced by the human kind, but cleaning of air is a horrendous task and hugely expensive, because of its large scope and the cost of energy. Up to now, all of the air cleaning systems are generally driven by external power, making it rather expensive and infeasible. Here, we introduce the first self-powered air cleaning system focusing on sulfur dioxide (SO₂) and dust removal as driven by the electricity generated by natural wind, with the use of rotating triboelectric nanogenerator (R-TENG). Distinguished from traditional approach of electrostatic precipitation by applying a voltage of thousand volt, our technology takes the advantages of high output voltage of R-TENG, typically in the order of a few hundreds volt. This self-powered air cleaning system not only adsorbs dust particles in air, but also oxidizes SO₂ without producing byproducts. Therefore, it could be potential for easing the haze-fog situation, which is one of the most important directions in self-powered electro-chemistry.

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Introduction

With the growing threat of air pollution and the crisis of energy, the search for cost-effective, renewable and green methods to clean air is one of the most urgent challenges. Especially in China that is experiencing a fast development, people have long been tormented by polluted air, which is

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mainly due to particulate matter (PM), sulfur dioxide and nitrogen oxide (NO_x). In Beijing, during 2000-2014, the annual average concentration of PM₁₀ was never lower than 100 µg/cm³. Various health problems are believed to be associated with long-term exposure to polluted air [1], such as respiratory diseases, rise in numbers of deaths from cardiovascular and respiratory disease among older people [2], and increase of lung cancer and cardiopulmonary mortality [3]. Therefore, it is essential to find a cost-effective way to resolve this issue in order to improve the quality of people's life.

Multiple approaches have been extensively demonstrated to remove dust and SO₂. For dust removal, methods such as simple filtration, adsorption with solid sorbents [4,5] pulsed electron beam, reaction with negative ion [6] and electrostatic precipitation [7] have been reported. Among these de-dusting approaches, electrostatic precipitation has been widely used in industries and indoors [8]. But a major drawback of this approach is that it requires an external power even at a high voltage, making it expensive and not adequate for large-scale outdoor usage. Moreover, some of the methods rely on corona effect for electrically charging particulates, which could generate ozone and NO₂ as byproduct [9]. The filtration is very simple and effective, but it could be easily clogged by submicron solid aerosol particles [10]. As for SO₂ removal, there are also many methods such as electro-catalytic and metal catalytic oxidation [11]. However, electro-catalytic oxidation needs external power, and the metal catalytic oxidation relies on novel metals which are very expensive and are rather limited.

Currently, many self-powered electronics or electrochemical applications have been achieved through triboelectric nanogenerators or piezoelectric nanogenerators such as splitting water [12] or degrading methyl orange [13] or sensing [14]. However, air cleaning as a big issue has not been realized using this self-powered method.

Herein, we innovatively fabricated a very simple and practical system for self-powered clearing of toxic materials in the air by electrostatic precipitating of flying dust particles and oxidizing SO₂. Instead of utilizing an external electricity power, we utilize the electricity generated from natural wind with the use of R-TENG. The output voltage of the R-TENG is about 300 V, which is not too high to generate byproducts such as ozone and NO_x [15], as produced in traditional electrostatic precipitation by using high voltage discharge. In addition, no noble metals are needed to electrochemically oxidize SO₂, since the generated current is high enough to electro-catalyze the oxidation of SO₂. Last but not the least, the self-powered SO₂ and flying dust removal system is cost-effective, versatile in size and easy to be applied, making it feasible for environmental cleaning and indoor air purification. It is also worth mentioning that the method of oxidizing SO₂ may have a great influence on sulfuric acid production, and this principle could be used in many other electrochemical reduction or oxidation applications.

Fabrication and characterization of R-TENG

The multi-layered R-TENG is mainly composed of a rotator and a stator, as shown in sketch diagram Figure 1a and optical image Supplementary Figure S1. The key components of the

stator and rotator are top grating and bottom grating electrodes which are fabricated by the print circuit board (PCB) technology [16]. Both of the electrodes are clusters of radially arrayed segments, made of copper, with radius of 70 mm and a central angle of 1°. The difference is that the top electrode has 180 sector units separated by an interval of 1°; while the bottom electrode is composed of two complementary patterned electrode networks which are separated by fine trenches in between. Each group of patterned electrode networks is respectively connected at one end. The rotator rotates under the driving force from wind blowing; while the stator is fixed on a bracket. For the fabrication of rotator, as depicted in Figure 1b, the top grating was just needed to be immobilized onto the top substrate (made of PMMA). As to the stator, the bottom grating was firstly immobilized onto the bottom substrate, and then adhered with a Kapton film (thickness of 30 µm) (Figure 1c).

The working principle of this kind of R-TENG has been reported previously [17,18]. Because of the PCB technology, the tribo-electrification units are miniaturized, and the output current is increased remarkably, yet the volume has not been enlarged. Moreover, the rotating design is very convenient for the exploitation of wind energy. Because of the light weight of the materials for making the rotator, the R-TENG could be easily driven by wind.

To characterize the optimized performance, the fabricated R-TENG was mechanically driven by a rotary motor with a rotating speed of 600 rpm. The open-circuit voltage (V_{oc}) was measured by a low-noise voltage preamplifier (Keithley 6514 System Electrometer) and the short-circuit current (I_{sc}) of the R-TENG was measured by a low-noise current preamplifier (Stanford Research SR570). As illustrated in Figure 1d and e, the measured I_{sc} and V_{oc} are 3.4 mA and 320 V respectively. For supplying the air cleaning system, the alternating current (AC) output of the R-TENG should be rectified into direct current (DC) output (supplementary Figure S2a-S2b). To indicate the capability of the R-TENG as a power source, it was connected to conventional light bulbs after rectification. With the wind gusting up to 8.5 m/s (5 BF in Beaufort wind force scale), the spot lights on the panel were all lightened up, as illustrated in Supplementary Movie S1.

Supplementary material related to this article can be found online at <http://dx.doi.org/10.1016/j.nanoen.2014.12.013>.

Experiments of self-powered SO₂ removing and de-dusting

To drive the two systems using electricity harvested from wind energy, the R-TENG was connected to a miniaturized wind cup structure that was driven by air blower at a wind speed of 15.1 m/s (7 BF). The oxidation of SO₂ and adsorption of flying dust were all performed in a closed transparent cubic chamber of 125 L with an air inlet, an air outlet and two needle-like copper wires across the wall at ambient temperature. Through wires, the R-TENG outside the chamber was connected to two paralleled copper meshes inside. For the experiment of SO₂ oxidation, the chamber was filled with SO₂ beforehand. Then the copper meshes were connected through a sink of saturated NaHSO₃ solution (Figure 3a). At given time intervals, the SO₂

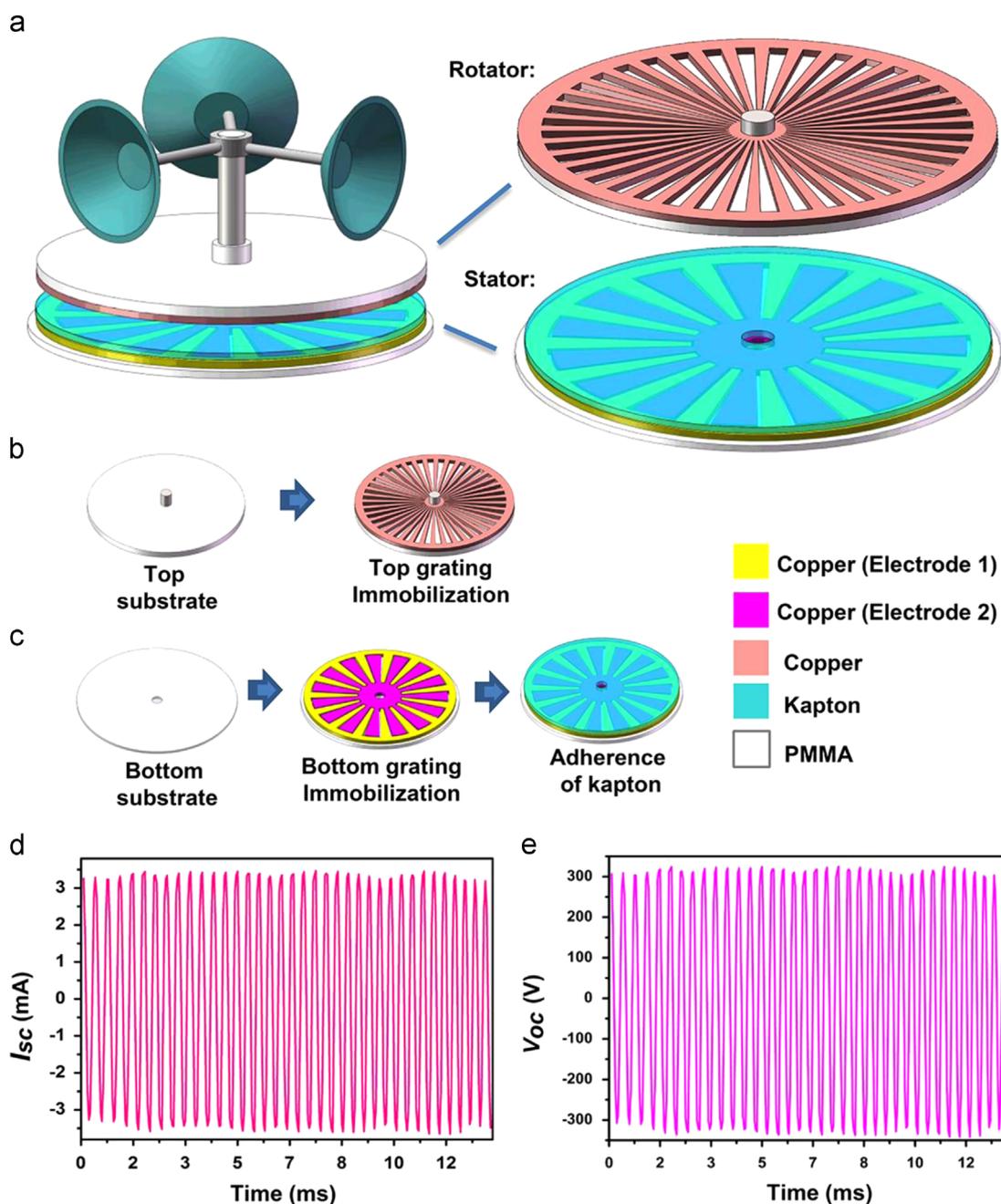


Figure 1 The schematic diagram and fabrication process of the fabricated R-TENG driven by wind energy. (a) Schematic illustrations of R-TENG with wind cup structure and enlarged view of Rotator and Stator. (b) Rotator and (c) Stator fabricating process flow. (d) The short-circuit current (I_{sc}) and (e) open-circuit voltage (V_{oc}) of R-TENG at 600 rpm.

inside the chamber was extracted by vacuum sample bags, and assayed by gas chromatography (GC) to determine the concentration change, which was then compared (for both setups with and without air cleaning system) to evaluate the air cleaning system's effectiveness. To verify that SO_2 is oxidized into sulfuric acid in the chamber with R-TENG, the anodic copper mesh is characterized by scanning electron microscope (SEM), stereomicroscope and energy dispersive spectrometer (EDS). For the experiment of dust removal, the chamber was replenished with dust aerosol through the inlet by solid aerosol generator series SAG 410. The weight of the dust adsorbed to

the meshes was measured with and without R-TENG in real time to determine adsorption efficiency.

Principle for self-powered removal of SO_2 and dust

According to the principle about electrochemical reaction, the mechanism of SO_2 oxidation can be proposed as follows:



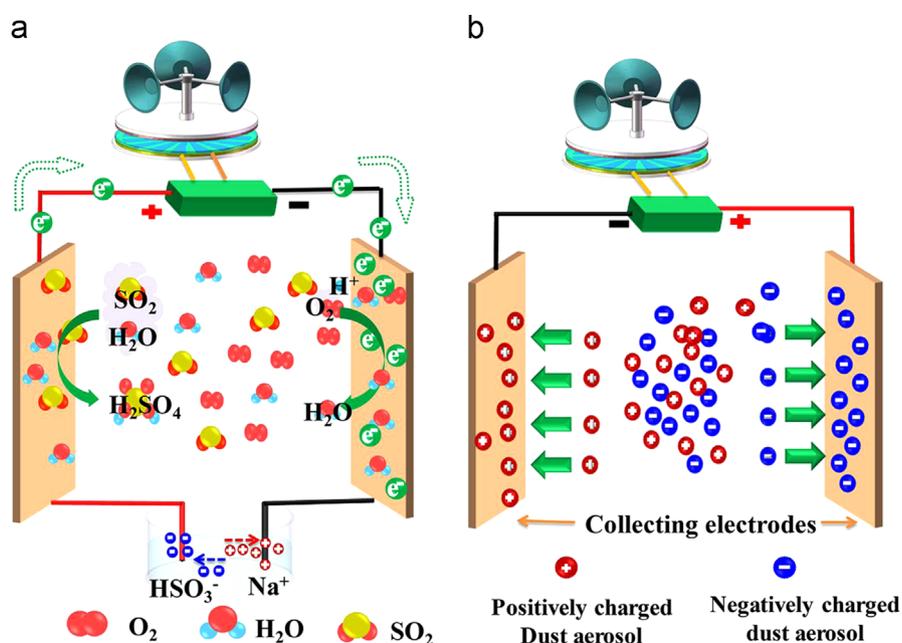
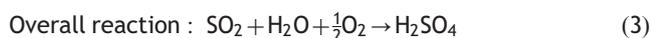
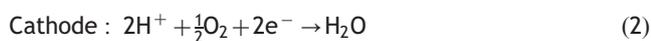


Figure 2 Schematic illustrations of the principles for SO₂ (a) and dust (b) removal based on electrostatics.



During the process of electrochemical reaction, electrons move from anode to cathode. At the anode, SO₂ loses two electrons and turns into sulfuric acid in water film that is coated on the surface of copper mesh. Therefore, the concentration of SO₂ in the chamber is decreased and sulfuric acid on the anode is detected. The schematic illustrations of the principle for SO₂ removal are described in Figure 2a. At the cathode, oxygen gains two electrons and turns into water.

As for dust removal, the principle is based on electrostatic precipitation. The mechanism can be explained by the following equations [19]:

$$E = \Delta\Phi/d \quad (4)$$

$$F = Eq \quad (5)$$

where E is the electric field between two plates; $\Delta\Phi$ is the potential difference between two plates, which approximately equals to the open circuit V_{oc} generated by R-TEG; d is the horizontal distance separating the plates; q is the charges on the dust particles. When generated from solid aerosol generator, dust aerosol is either positively or negatively charged due to triboelectric effect between the particles and air [20]. Along with compressed air, the charged aerosol is pumped into a chamber through an inlet by air pump. Under the Coulombic force of attraction and repulsion, charged dust particles are drawn onto two electrodes. As demonstrated in Figure 2b, the negatively charged dust particles are pulled onto the positive electrode; while the positively charged dust particles are drawn onto the negative electrode. Thus, the weight of copper mesh is increased.

The most prominent distinction between de-SO₂ and de-dust principle is that de-dusting depends on electric field density, while SO₂ removal needs a closed circuit. Although

the SO₂ and dust removing mechanisms are well established, the most innovative thing here is that the energy required for carrying out the processes is using energy harvested from wind. Owing to the renewability of wind energy, there is no need to worry about energy consuming problem during air cleaning. As long as wind exists, air cleaning system could execute its function, resulting in the decrease of cost.

Self-powered oxidation of SO₂

A schematic diagram of the system for self-powered oxidation of SO₂ is sketched in Figure 3a. The R-TEG captured wind energy with the assistance of wind cups, generating AC electricity. In order to oxidize SO₂ continuously, the generated AC electricity is converted into DC electricity through a rectifying bridge. The saturated NaHSO₃ solution is connected with two copper meshes in between to circulate the current and eliminate the interference of SO₂ dissolving for the reason that saturated NaHSO₃ solution has certain conductivity and that SO₂ is almost insoluble in saturated NaHSO₃ solution. NaCl was added into the saturated NaHSO₃ solution till no dissolving to improve the conductivity. Through the evaporation of the solution, water film is coated on the copper meshes to participate in the degradation of SO₂, as shown in the mechanism of SO₂ degradation. The optical image of the system is shown in Supplementary Figure S3. When the system works, SO₂ can be oxidized into sulfuric acid on anode, as verified by Figure 3b where a cloudy white precipitate appeared in the BaCl₂-HCl solution after addition of rinse solution of copper mesh with R-TEG, while no changes occurred in the same situation without R-TEG. Because of the generation of sulfuric acid, the pH of anodic copper mesh's rinse solution decreased (Figure 3c) and the corrosion of copper mesh in SO₂ atmosphere accelerated (Figure 3d by SEM and Supplementary Figure S4 by stereomicroscope) compared to the system without R-TEG, thus proving our assumption. The increase of sulfur

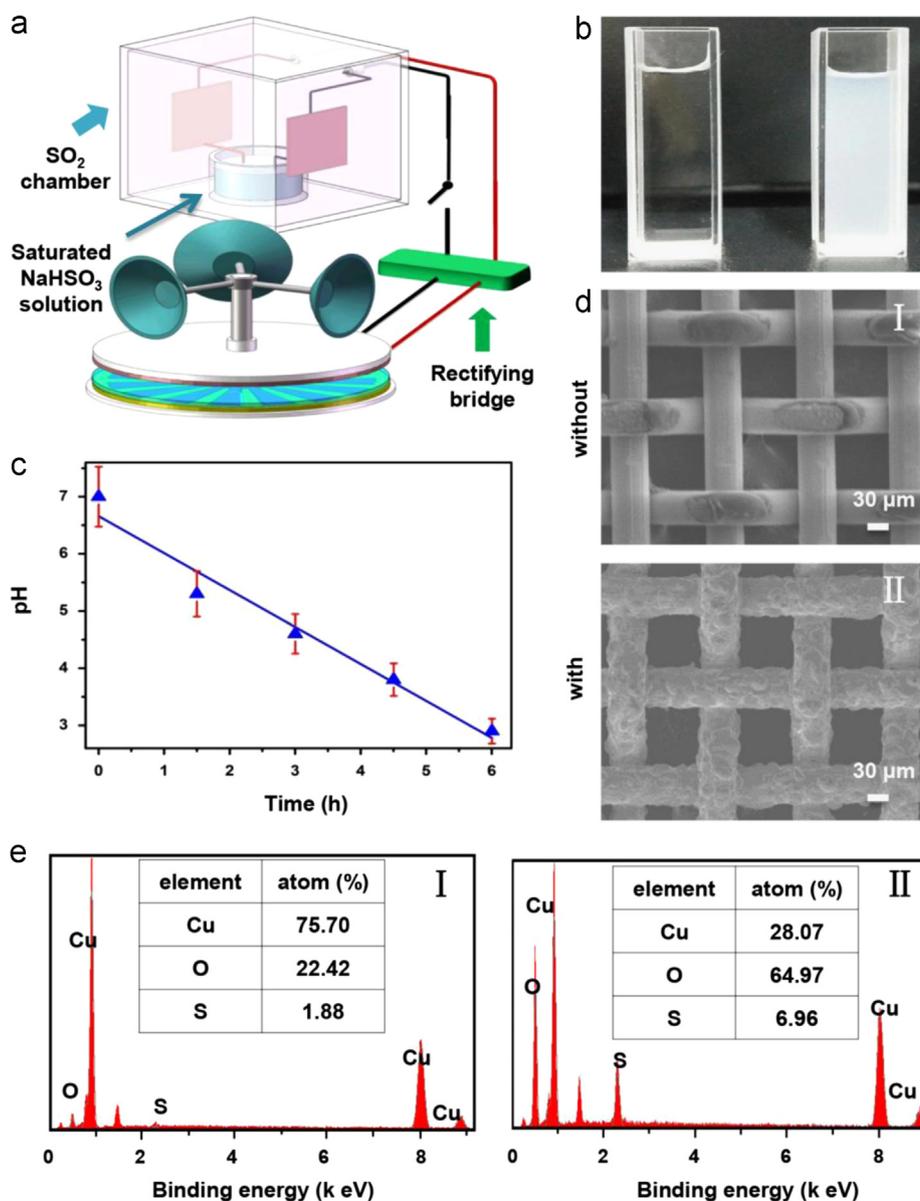


Figure 3 Self-powered oxidation of SO_2 . (a) Schematic diagram of the system. (b) The optical images of the $\text{BaCl}_2\text{-HCl}$ solution after addition of rinse solution of copper mesh without and with R-TENG. (c) pH of rinse solution of anodic copper mesh under a wind speed of 15.1 m/s (7 BF). SEM image (d) and EDS spectra (e) of copper mesh without (I) and with (II) R-TENG. Wind speed, 15.1 m/s (7 BF); working time, 6 h.

and oxygen elements in the EDS result (Figure 3e) of the copper mesh with R-TENG compared to the system without R-TENG further demonstrates the assumption from another point of view.

To figure out the defacto current for oxidizing SO_2 , the current output of R-TENG with NaHSO_3 solution and copper meshes is also measured. As shown in Figures S5 and 4a, there is a correlation between the rectified current and wind speed in conditions where R-TENG is under load or not. When the wind speed rises, the current increases. This result can be explained by the change of charge transfer rate in the triboelectrification surfaces. Due to $I = \Delta Q / \Delta t$, the higher the wind speed is, the shorter the charge transfer duration, which means a larger current. Through the comparison of Figures S5 and 4a, it also can be seen that the defacto current magnitude in Figure 4a decreased to μA , that is because saturated

NaHSO_3 and NaCl solution have larger resistance. Though the defacto current is very small, the oxidation of SO_2 could proceed continuously, since the electrochemical reaction relies on electrode potential.

To demonstrate the effectiveness of the system and the influence of wind speed on SO_2 removal, a systematic experiment without and with R-TENG under different wind speeds was conducted. Through GC peak intensity in Figure 4b-f, we could see that after the same time intervals the SO_2 concentration in the chamber without R-TENG almost remains same with time going by; in contrast, the SO_2 concentrations in the chamber with R-TENG decreases under wind speed of 10.2 m/s (5 BF) and 15.1 m/s (7 BF), indicating that the self-powered SO_2 removing system is effective. Figure 4g depicts the corresponding SO_2 concentration change in Figure 4b-f under different working time and wind speed, showing that the SO_2

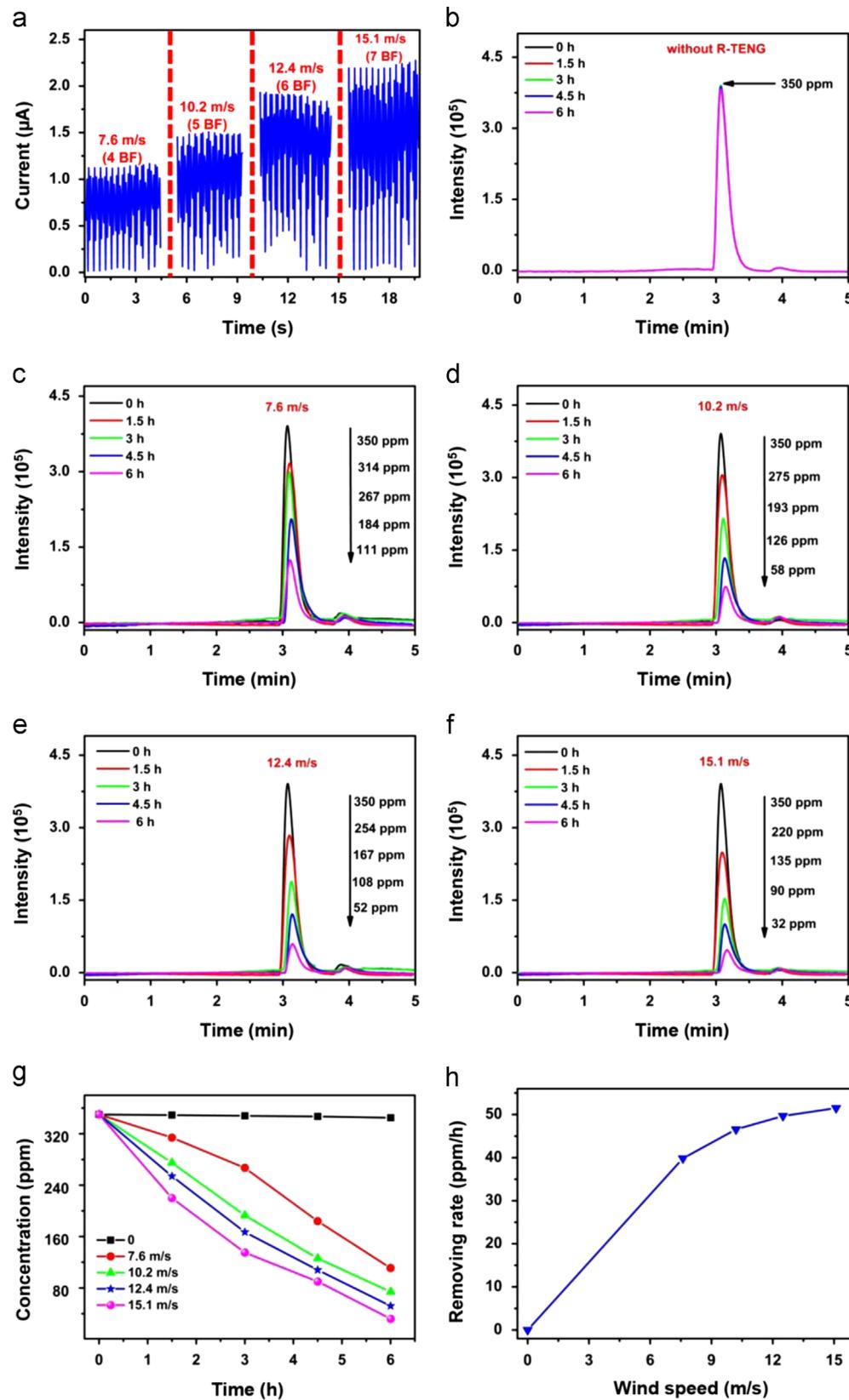


Figure 4 (a) The output current of R-TENG under load under different wind speeds from 7.6 m/s (4 BF) to 15.1 m/s (7 BF). (b)-(f) The GC peak intensity of SO₂ in the chamber under different time intervals without and with R-TENG under wind speed of 7.6 m/s (4 BF), 10.2 m/s (5 BF), 12.4 m/s (6 BF), 15.1 m/s (7 BF). (g) The SO₂ concentration as a function of wind speed. (h) SO₂ removing rate versus wind speed.

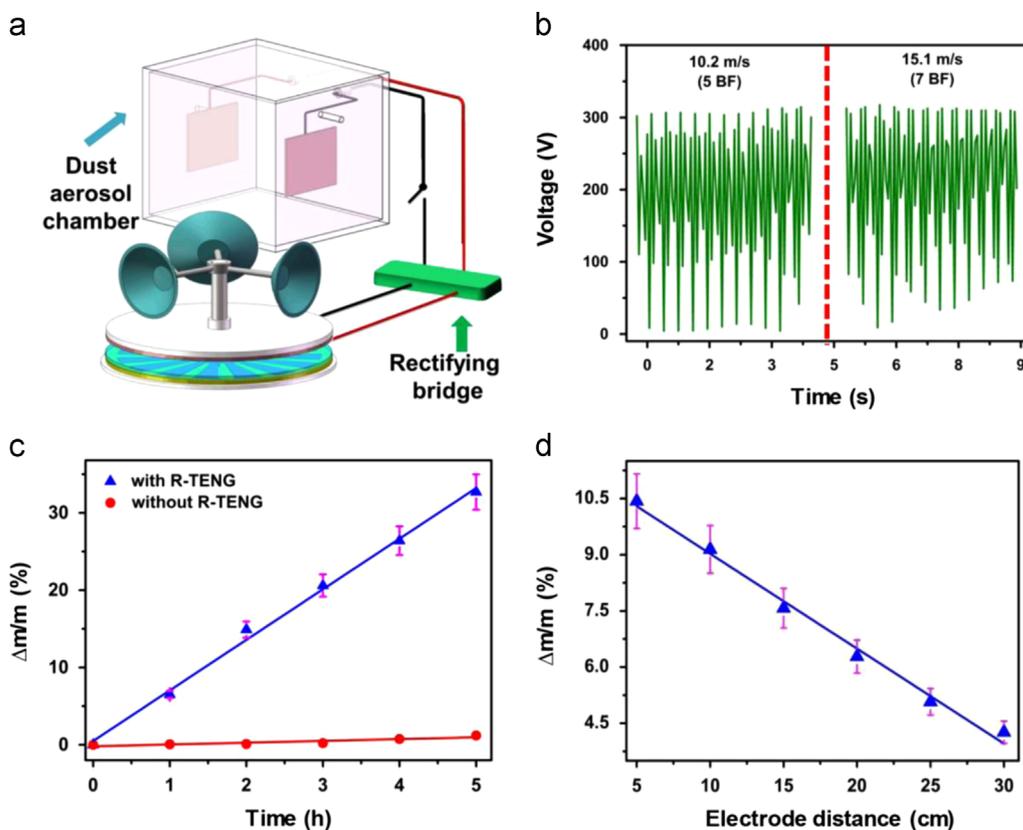


Figure 5 Self-powered precipitation of dust particles. (a) Schematic diagram of the system. (b) Rectified voltage of R-TENG connected with copper meshes under different wind speeds from 10.2 m/s (5 BF) to 15.1 m/s (7 BF). (c) Weight increase of copper mesh with and without R-TENG. Distance of collecting plates, 15 cm. (d) Dust adsorption rate as a function of collecting plates' distances of 5 cm, 10 cm, 15 cm, 20 cm, 25 cm, and 30 cm. Wind speed, 15.1 m/s (7 BF).

concentration decreases more quickly under higher wind speed, which also can be seen more clearly through SO_2 removing rate in Figure 4h. The reason for this phenomenon is that the SO_2 oxidation rate depends on anodic current density while anodic current density depends on rotating speed. Hence, high wind speed is beneficial for SO_2 removal.

Self-powered adsorption of dust

Figure 5a illustrates the schematic diagram of the system for self-powered adsorption of flying dust, among which electricity supplying and electrode components are the same as in Figure 3a for SO_2 removal. The optical image of the system is shown in Supplementary Figure S6. Figures 5b and S7 implies that the voltage of R-TENG connected with or without copper meshes remains a saturated value of 300 V when wind speed rises. This result can be explained by the following two facts: firstly, though the wind speed rises, the contact area of the tribo-electrification surfaces has not been changed, so the voltage output remains 300 V [17]. Secondly, for dust adsorption in the electrostatic field, the circuit is open, the voltage between two copper meshes is equal to the output voltage of R-TENG, thus making the measured voltages in Figures 5b and S7 almost the same.

In order to confirm that the increase of dust absorption is due to the electrostatic precipitation, a comparative

experiment was performed. As we can see from Figure 5c, the weight increase of the dust in the condition without R-TENG at the same time intervals is much lower than that with R-TENG where the weight increase of dust goes obviously with working time. After 5 h, when the wind speed is 15.1 m/s (7 BF), 1 g copper mesh could adsorb 0.327 g dust, proving the feasibility of this de-dusting system.

Besides time, distance between two copper meshes is another influence factor for dust removal. As implied in Figure 5d, the weight increase goes up with the reduction of the distance. It is because electric field is inversely proportional to distance. With the decrease of distance between two collecting plates, electric field between two copper meshes rises correspondingly, and the Coulombic force exerted on flying dust rises consequently, resulting in the enhancement of weight increase on copper mesh, which has been discussed above.

Conclusion

In summary, we have introduced a novel self-powered air cleaning system that is capable of effectively removing SO_2 and flying dust particles in air. By using grating electrodes as electrical performance enhancer, the as-developed R-TENG could achieve a high voltage of 320 V and a high power density

of 3.4 mA. Through the fabricated R-TENG, wind energy was harvested, and then immediately used to oxidize the SO₂ and adsorb the flying dust in the air. The working principle of the air cleaning system has been elucidated through conjunction of schematic illustrations and experiments. Not only the air cleaning system makes full use of the wind energy, but also shows advantage in simplicity, low cost and effectiveness. Thus, this self-powered dust and SO₂ removing system holds great potential for cleaning and prevention of air pollutants. As most polluting molecules can be oxidized or reduced through electrochemical method, and most polluting ions could be absorbed through electrostatic interaction, we could expect that this work may inspire the development of R-TENG toward other pollutants control, such as sewage treatment and formaldehyde removal, in the near future. This is a new direction in self-powered electrochemistry.

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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.2014.12.013>.

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