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Energy Conversion

Generating Electricity from Water through Carbon Nanomaterials

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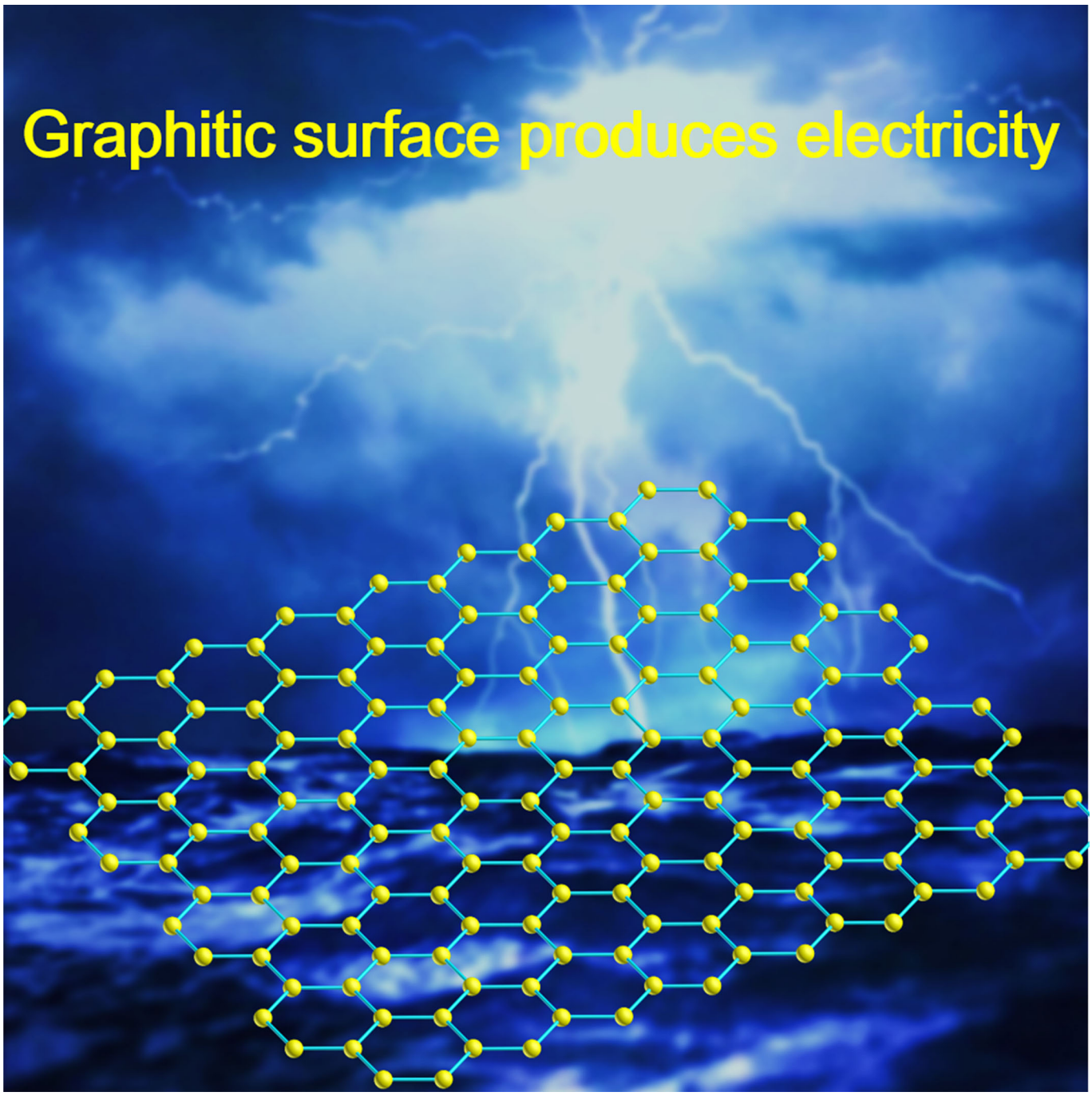

Abstract: Over the past ten years, electricity generation from water in carbon-based materials has aroused increasing interest. Water-induced mechanical-to-electrical conversion has been discovered in carbon nanomaterials, including carbon nanotubes and graphene, through the interaction with flowing water as well as moisture. In this Concept article, we focus on the basic principles of electric energy harvesting from flowing water through carbon nanomaterials, and summarize the material modification and structural design of these nanogenerators. The current challenges and potential applications of power conversion with carbon nanomaterials are finally highlighted.

Introduction

With the approaching exhaustion of traditional fossil energy, harvesting energy from the environment, like wind, solar, geothermal and water energy, has been considered as an effective solution for the increasing energy crisis.[1–3] Among these renewable energy sources, converting the potential mechanical and electrochemical energy stored in water into electricity is both attractive and promising, as water is abundant and widely available in rivers, lakes and oceans as well as in our body.[4,5] Mankind has utilized hydropower for more than one hundred years. Large dams and water wheels have been built to harvest the enormous energy from flowing water for power supply and as a basis for social advancement and life. However, these facilities are usually large and complex, a novel design of miniaturized and highly efficient nanogenerators is desired to satisfy the working needs for small size or even implantable feasibility in personal and biomedical devices. Carbon nanomaterials have been considered as promising candidates since power generation of carbon nanotubes (CNTs) from flowing water was discovered in 2003.[6] Nanoscale graphitic surfaces enable fast transport of water inside and unique nanofluidic phenomena.[7] According to electrokinetic effects, the flow through the nanochannels can induce electrical power generation by means of streaming currents.[8,9] In this Concept article, we will briefly discuss electricity generation from water using carbon nanomaterials, including CNTs, graphene and their derivatives with emphasis on working mechanisms.

Nanogenerators Based on Carbon Nanotubes

Flowing electricity generation by individual metallic CNTs at nanoscale was predicted in 2001 through theoretical calculations.[10] Two mechanisms, momentum transfer and fluctuating Coulombic fields, were proposed for the generation of flowing potential. However, energy harvesting from water flow by using fluidic nanogenerators based on bundles of CNTs was not experimentally realized until 2003.[6] An output voltage of 2.67 mV was produced in pure water at a flowing velocity of 1.8 mm s^{-1} (Figure 1 a). The potential difference (1 mV) at the interface of the metal electrodes with the CNT bundles was taken into consideration, and was subtracted from the output voltage measured in flowing water. The experimental results showed that voltage generation had rather weak dependence on liquid viscosity in the calculation process of momentum transfer. In fact, logarithmic dependence of the voltage output on the flowing velocity and the polar nature as well as the ion concentration of the liquid were observed. The authors proposed a mechanism based on one-dimensional periodic ratchet potential, where the potential resulted from the fluctuating imbalance in local charge neutrality produced by the velocity gradient near the liquid–solid interface.

However, the fluctuating imbalance should be perpendicular to the flow direction, and did not agree well with the output voltage measured along the flowing direction. The distribution of free charges was also uniform in the flowing direction, and no voltage would be generated in this way. It was worth noting that the CNT purification treatment of long exposure to HCl in this work might leave the CNT surface charged. The electricity generation was more likely to be attributed to an ionic mechanism. A more prosaic working mechanism of streaming potential in porous materials was suggested for the flowing voltage generation, that is, the flowing liquid carried counter-ions accumulated in a Debye layer near the liquid–solid interface.[11] The acid-purification treatment resulted in excess positive charge at the Debye layer of the CNT surface, which was consistent with the experimental voltage signs.[6]

Besides the ions, the interaction between water molecules and CNTs has also been considered to play an important role in producing flowing electricity. Weak coupling between CNTs and water dipoles was proposed as another possible mechanism for the flowing voltage generation in water-filled CNTs.[12] Through the charge redistribution resulting from the interaction between the charge carriers and the water dipole chains in the CNT, an output voltage of 17.2 mV was predicted. The hypothesis was in agreement with the previous experimental results, as a voltage generation of $\approx$8 mV was observed in water-filled single-walled CNTs.[13] Water molecule chain formed in the one-dimensional confinement of single-walled CNTs through hydrogen bonds, and the electrostatic field of water molecules polarized the CNTs and caused a charge distribution.

The discovery of flowing electricity in CNTs provided a novel prospect to harvest green energy. However, the output voltage generation of CNTs in the early studies was generally limited to a few millivolts due to the small sizes of the devices. The macroscopic assembly of CNTs in an aligned structure can well maintain the remarkable properties of individual CNTs, which provides an efficient method to greatly enhance the output electricity. Therefore, aligned CNT materials such as sheets and fibers represent promising candidates.

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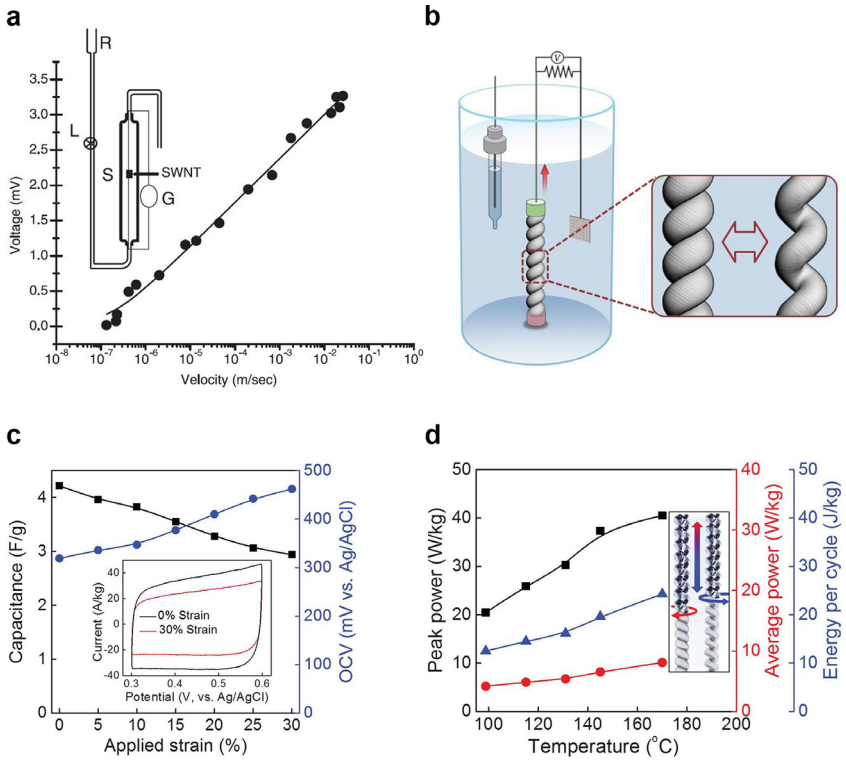

Figure 1. a) Dependence of the output voltage on the velocity of water. Reproduced with permission.[6] Copyright 2003, American Association for the Advancement of Science. b) Schematic illustration to a coiled CNT yarn electrode, reference and counter electrodes in an electrochemical bath. The enlarged section shows the coiled CNT before and after stretching. c) Dependence of the capacitance and open-circuit voltage (OCV) on the applied strain. Inset: cyclic voltammetry (CV) curves under 0% and 30% strain. d) Dependence of peak power, average power and energy per cycle on the temperature of a coiled nylon artificial muscle connected to the CNT yarn which stretched and twisted the yarn nanogenerator during heating. Inset: the yarn nanogenerator stretches and up-twists during heating and the process is reversed during cooling. Reproduced with permission.[14] Copyright 2017, American Association for the Advancement of Science.

Nanogenerators Based on Aligned Carbon Nanotubes

Similar electricity production was achieved by nanogenerators based on macroscopic aligned CNT yarns and sheets with much improved harvesting performance. An aligned CNT yarn harvester that converts tensile or rotational energy of the yarn into electricity in static solution was previously proposed.[14] An elastic coiled structure was obtained by inserting extreme twist into a CNT yarn. Output voltage was generated by the capacitance decrease of the coiled-CNT-yarn under applied strain in aqueous electrolyte (Figure 1 b, c). The reversible variation of the capacitance was caused by the change of yarn density under untwisting and twisting in the stretching and releasing process. If the homochiral coiled yarn was stretched, the yarn coiling would be partially converted to yarn twist that increased the yarn density, decreased the capacitance and increased the open-circuit voltage. The voltage decreased when a heterochiral yarn was stretched. The mechanical-to-electrical energy conversion efficiency was 1.05% during cycling of a coiled CNT yarn to 20% strain at 1 Hz in 0.1 M HCl. Through integration with a thermally annealed coiled nylon fiber artificial muscle,[15] the CNT yarn could convert thermal energy into electrical energy when being up-twisted and stretched by heating of the nylon muscle (Figure 1 d).

A novel fiber-shaped fluidic nanogenerator (FFNG) based on aligned multiwalled CNT sheets was also discovered with high flexibility and stretchability.[16] Flowing electric output was derived from the relative movement between the FFNG and saline water. A model based on an electric double layer (EDL), also known as the Debye layer, was summarized for the working mechanism of electric output generation (Figure 2). The EDL is formed at the interface between the CNT and the fluid as the fluid contacts the FFNG. The net charge of the stern

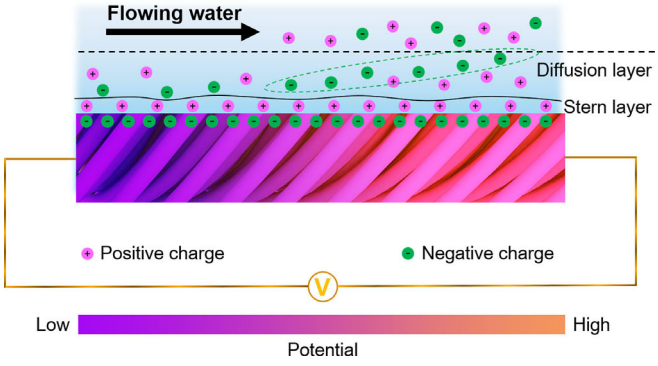

Figure 2. Schematic illustration to the working mechanism of the generated flowing potential in the FFNG. Reproduced with permission.[16] Copyright 2017, Wiley-VCH.

layer could not be immediately counteracted by the anions at the front end of the fluid, inducing charge imbalance along the FFNG which drew electrons from the CNT to balance the excess charges. Unbalanced charges were further accumulated at the front half of the FFNG during the flowing process, inducing a potential difference. As the contact length of the solution with the FFNG no longer increased, the excess charge would be gradually balanced by the ions and the potential difference decreased. During the flowing process, only one electrode of the FFNG contacted the solution, and no obvious output voltage could be detected when both of the two electrodes were immersed, indicating a different working mechanism to previous reports. It should be noted that the electric output did not return to zero after the solution ceased flowing, and the output voltage could be attributed to the galvanic nature of carbon–metal nanomaterial junction, which will be further discussed below. Therefore, both the flowing electric energy based on the EDL working mechanism and the chemical energy of carbon–metal nanomaterial junction should contribute to the total electric output of the FFNG.

The energy-harvesting process of the FFNG was highly efficient, and it showed a high power conversion efficiency (PCE) of 23.3%, which was much higher than the other fiber-shaped generators, including solar cells, electrostatic and triboelectric nanogenerators. The PCE was calculated as the ratio of the total electric work output of the FFNG to the flowing mechanical input of the fluid. The energy harvesting performance demonstrated both high stability and durability, as evidenced by well-maintained output voltage after 1 000 000 deformation cycles. Both high peak output voltage of 2.3 V and peak output current of 0.34 mA were produced by connecting twelve FFNGs in series and in parallel, respectively. Moreover, based on their unique one-dimensional shape, FFNGs might desirably be implemented in the human body to harvest flowing energy from body fluids and blood.

Nanogenerators Based on Graphene

Compared to the one-dimensional structure, two-dimensional carbon nanomaterials such as graphene have aroused growing interest recently owing to less lossy interfaces and less complicated ion transport.[17] Graphene shares a similar component and structure with CNTs, and therefore it is expected that energy harvesting performance can also be achieved by the water–graphene system based on similar working mechanisms. A flowing aqueous solution of 0.6 M HCl over few-layered graphene was first reported to generate a peak output voltage of 30 mV.[17] Since a nonlinear response of the induced voltage to the flowing velocity was observed, momentum transfer and streaming potential models could not be used to interpret the experimental results. On the basis of molecular dynamics simulations, a working mechanism based on surface ion hopping was proposed for the flowing power generation. But the flowing voltage generation of the graphene was questioned in the next year.[18] It was stated that the induced voltage should be attributed to the interaction of the ions with the metallic electrode connected to the graphene, and the immersed graphene itself could not produce the flowing voltage in the ionic water. In contrast, the graphene was more likely to behave as a load connected to the two electrodes which transported the charge carriers, rather than a fluidic nanogenerator, since no measurable voltages could be detected when the electrodes were sealed and prevented from touching the fluid. The conclusion was further confirmed in a probe of graphene transistor for streaming potential, as no flow-induced currents were detected in graphene.[19]

However, open-circuit voltage of 0.1 V and short-circuit current of 11 μA were reported in the graphene-sheet-based fluidic nanogenerator, and in this work, the electrode connected to the graphene sheet in the fluid was carefully protected by silicone to avoid exposing to the fluid.[20] Unlike the previous work where the graphene was completely immersed in the water, here the fluidic nanogenerator was only partly immersed, indicating a different working mechanism. A working mechanism based on moving boundaries of EDL was proposed for the power generation of graphene sheet. The output potential arose from charge transfer in the graphene sheet driven by boundary movement of an EDL between saline water and graphene sheet. During the immersion process of the graphene sheet in saline water, an EDL consisting of adsorbed Na^+ layer and the attracted Cl^- layer would form near the liquid–gas boundary on the graphene. By moving the boundary of the EDL, the local hole concentration was raised, inducing a higher local potential and drove a current from the liquid–gas boundary to the immersed graphene deeper in the fluid. Once the graphene sheet was completely immersed, the voltage would drop to a resting value since no liquid–gas boundary existed, indicating that the hole redistribution was caused by moving the EDL near the boundary.

Interestingly, a voltage of a few millivolts was generated by moving a droplet of ionic liquid (such as NaCl) on graphene.[21] According to density functional theory calculations, a hydrated cation of Na^+ would be adsorbed on the graphene surface that contacted the droplet because of its positive adsorption energy. With the increasing adsorbed Na^+, a thin layer of accumulated electrons extended along the contacted surface of the graphene, and a pseudocapacitor was formed at the graphene–liquid interface (Figure 3a, b). It was driven forward by moving the liquid droplet, which drew the electrons in the graphene. Compared with the static state, the ions were adsorbed at the front end and desorbed at the rear, decreasing or increasing the electron density behind or ahead of the droplet, respectively, and thus producing a higher potential at the front (Figure 3c). The output voltage was found to be dependent on the species and concentration of the ions in the droplet, and showed a linear relationship with the flowing velocity. No detectable voltage output could be generated for deionized water. Notably, moving two droplets in opposite directions did not produce flowing voltage since individual electricity generation was counteracted.

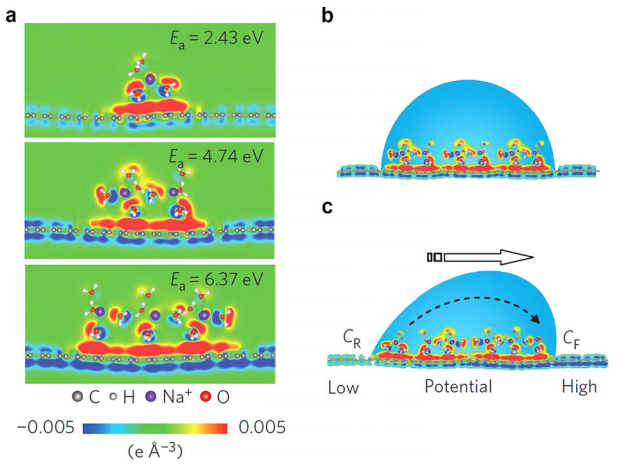

Figure 3. a) Density functional theory calculation results for the charge distribution near the graphene surface caused by adsorbing one, two and three hydrated Na^+ and the adsorbing energy. b) Schematic illustration of a pseudocapacitor formed by a droplet on graphene. c) Schematic illustration of the potential difference generated by moving a droplet. Reproduced with permission.[21] Copyright 2015, Nature Publishing Group.

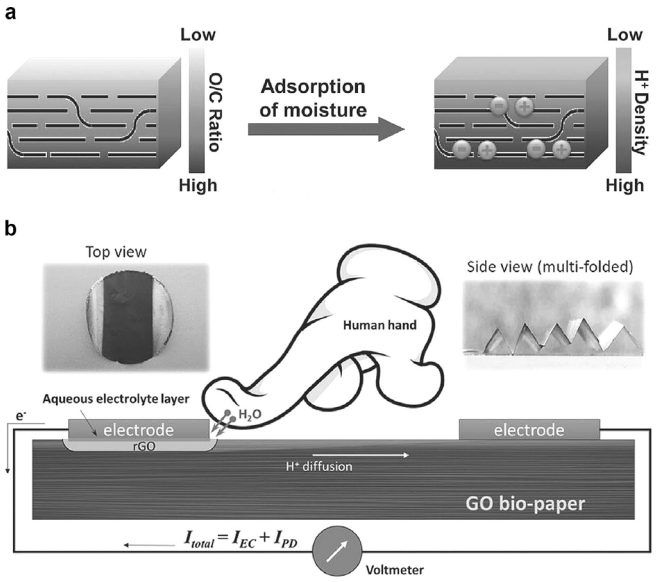

Figure 4. a) Schematic illustration of the gradient distribution of free H^+ produced by the oxygen-containing group after adsorbing moisture. Reproduced with permission.[25] Copyright 2015, Wiley-VCH. b) Schematic illustration of the GO–metal junction and the working mechanism. The left-hand side shows the top view, and the right-hand side shows the side view. Reproduced with permission.[27] Copyright 2016, Wiley-VCH.

Nanogenerators Based on Graphene Oxide

Salts dissolved in water are required for the power generation of graphene in the above-mentioned work. However, in nature, pure water exists across a wider geographic area compared to saline water, and nanogenerators which can harvest energy directly from pure water are favorably desired. Graphene oxide (GO) with oxygen-containing groups, such as carboxyl and hydroxyl groups, has shown unique responsive actuations owing to the sensitivity to pure water.[22,23] Furthermore, by varying the relative humidity, the electrical and ionic conductivities of the GO were controllable.[24] Therefore, a more universal nanogenerator harvesting energy from pure water can be prepared through chemical modification of graphene to produce electricity from water vapor. The power-generating phenomenon of GO film with an oxygen-containing group gradient under moisture has indeed been observed.[25] The exposed GO film could adsorb water molecules from the moisture or vapor due to the strong hydrophilicity of oxygen-containing groups. As the water molecules accumulated in the O-rich part of the GO film, free H^+ would be released from the oxygen-containing groups owing to the hydrogen bond weakened by the regional solvation effect. The gradient distribution of oxygen-containing groups resulted in the concentration gradient of free H^+ (Figure 4a). Therefore, potential difference and free electron movement in the external circuit were produced by the diffusion of free H^+, and the formed electric field drove the H^+ back to the high concentration end. Eventually, it reached a dynamic balance with the diffusion process, and the output voltage was stabilized. It was not surprising that the GO-based nanogenerator could harvest energy from our breath and an output voltage of 18 mV was produced, as the breath of humans and animals is one constant and steady source of moisture tide. Beyond energy harvesting, the GO-based nanogenerator could also function as a self-powering device to track real-time body conditions through our respiratory moisture, converting it into electric signals.

Based on a similar working mechanism, a more efficient moisture-induced electricity generation was also proposed from a superhydrophilic three-dimensional (3D) framework of the GO.[26] An improved output voltage of 0.26 V was produced with a relative humidity variation of 75%. As discussed above, the water adsorption was a critical factor for the power generation. One single layer of GO could adsorb a water weight content of 12%, capturing water molecules by the oxygen-containing groups. In contrast, the 3D framework of the GO provided a highly porous structure and fast permeation with nanoscale GO sheets for the adsorption of water molecules, showing a higher water adsorptance of 33.1% within 1 s under relative humidity of 80%. The 3D skeleton structure also enabled a more efficient ion transport through the layered structures. These all benefited the energy-harvesting performance of the nanogenerator based on 3D GO. By connecting five units of the nanogenerators in series and parallel, an output voltage of 1.32 V and current of 10.5 mA were produced.

However, in the above-two studies, the effect of the electrode connected to the GO was neglected in the energy harvesting from moisture. A flexible nanogenerator based on GO–metal hybrid materials which harvest energy from the touch of human fingers was further investigated.[27] When the GO–metal junction was made wet by pressing with a bare finger, an output voltage of hundreds of millivolts was produced with polarity depending on the touching location (Figure 4b). The authors suggested a working mechanism of the electrochemical reaction between the active metals (like copper and aluminum) and the GO after the adsorption of water from the moisture on the GO–metal junction, generating electrochemical po-

tential. For the materials inert to GO, only the diffusion of H^+ contributed to the voltage generation. It was confirmed by the fact that the GO–Al junction produced a high output voltage of 800 mV, and the output voltage for the GO–Au junction was 250 mV. Owing to the touching voltage generation of the GO–metal nanogenerators, a self-powering electronic skin for tactile sensing was fabricated with individual nanogenerators positioned in an array, displaying a promising future for interfacial sensors in soft robots.

Nanogenerators Based on Other Carbon Materials

The oxygen-containing group plays a critical role in moisture-enabled electricity generation, and it is expected that other carbon materials with functional groups can also generate electricity from moisture. In fact, besides GO, this energy harvesting performance has been observed in other carbon materials with oxidative modification. For instance, it has been detected with moisture in porous carbon films with two sides of different contents of oxygen-containing groups (Figure 5a).[28] To avoid contacting water vapor, the copper electrodes connected to the carbon films were covered with epoxy resin (Figure 5b). A constant output voltage of 68 mV was achieved after its exposure to the environment with high relative humidity of $>95\%$ for 6 h, and the voltage decreased to zero when the relative humidity decreased to below 83%. The reproducibility of this process indicated a strong dependence of the output voltage on the relative humidity. Notably, carbon films with two sides of the same content of oxygen-containing groups could not induce significant electric potentials. Combined with ab initio molecular dynamics and grand canonical Monte Carlo simulation, a working mechanism similar to the moisture-induced voltage generation in graphene was proposed. The imbalance of oxygen-containing groups and water adsorption caused the H^+ concentration gradient, and the output voltage was attributed to free H^+ transfer driven by the concentration difference (Figure 5c).

Unlike high humidity conditions, natural evaporation of water is a more common water power source, and carbon black sheet was reported as a highly efficient water-evaporation-induced nanogenerator (Figure 6a).[29] The carbon black sheet with a thickness of $\approx$70 μm consisted of carbon nanoparticles (average size of 20 nm) which were treated by annealing and plasma to be hydrophilic. The carbon black sheet output an increasing open-circuit voltage of up to 1 V when inserted into a beaker with the bottom end covered by deionized water, and the output voltage could be well maintained for 8 days (Figure 6b). It was noticed that the output voltage achieved its maximal value when the capillary water reached the maximal height along the carbon black sheet; the output voltage decreased when the beaker was sealed and the water evaporation was saturated. Therefore, there existed a strong relation between voltage generation and water evaporation. The electricity generation was derived from the evaporated water flowing inside the porous carbon black film with hydrophilic functional groups. According to density functional theory simulations, a specific EDL formed at the interface between the carbon nanoparticles and adsorbed water, and the electron redistribution resulted in possible electrokinetic effects. However, the working mechanism for electricity generation was still not clear enough, and other models such as momentum transfer, fluctuating Coulombic fields, coupling of water molecules and movement of free H^+ might also contribute to the electricity generation. More efforts are required to reveal the exact principle behind this phenomenon. Through parallel and series connections of four nanogenerators, the induced output voltage and current could be scaled up to 4.8 V and 380 nA, respectively; they were high enough to light a liquid crystal display (Figure 6c, d). The porous carbon film could also be prepared by using a printing method for low-cost industrial scale-up production, and a controlled deposition

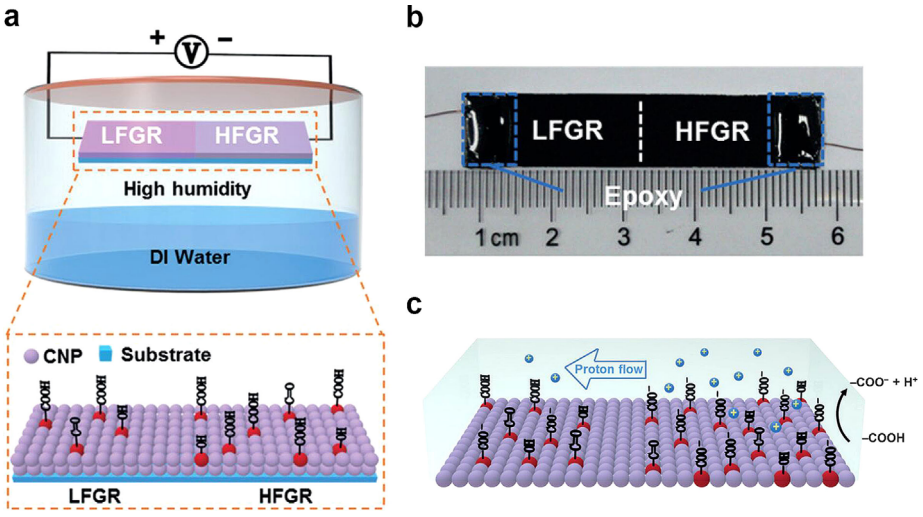

Figure 5. a) Schematic illustration of the experimental setup. b) Photograph showing the carbon film nanogenerator. The two electrodes were covered with epoxy. c) Schematic illustration of the proposed mechanism for voltage generation. The proton flow was derived from the imbalance of released H^+. Reproduced with permission.[28] Copyright 2016, Wiley-VCH.

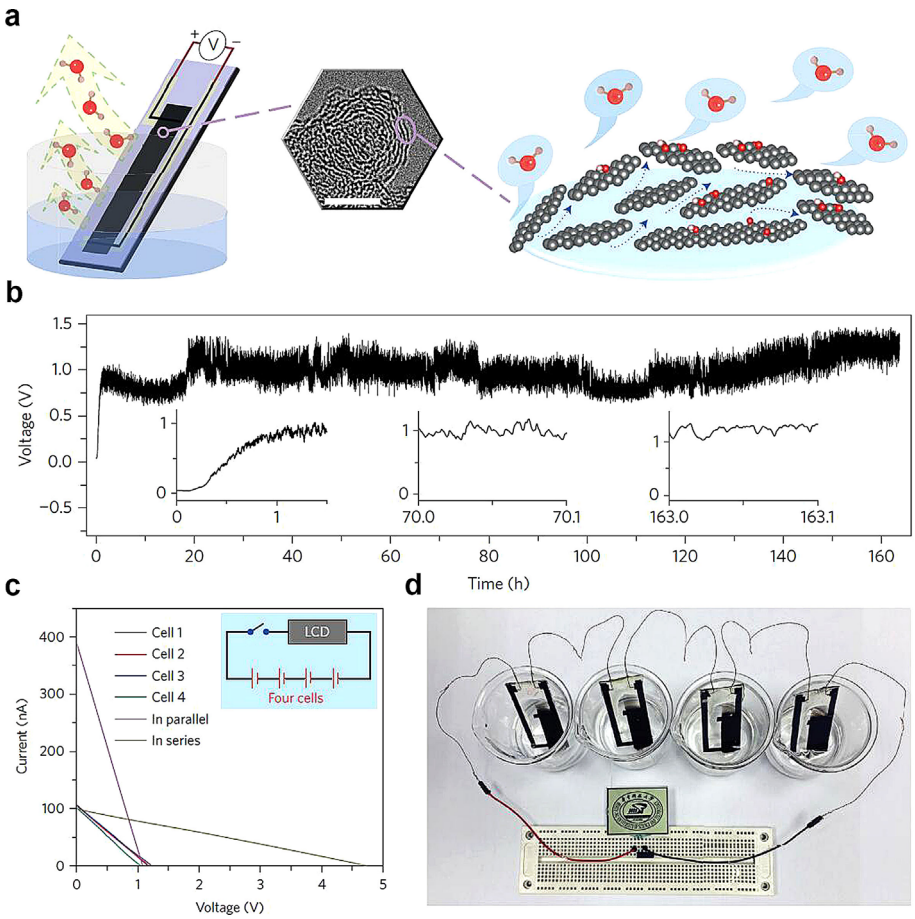

Figure 6. a) Schematic illustration of the experimental setup. The nanogenerator consisted of a carbon black sheet with two CNT electrodes, and it was inserted into deionized water for voltage generation. b) Output open-circuit voltage of the carbon black nanogenerators under fluctuating relative humidity between 53.5% and 66% was well maintained for 163 h. The insets show the measured curves of 0 to 1.5 h, 70.0 to 70.1 h and 163.0 to 163.1 h. c) Current-voltage curves of four nanogenerators connected in series and in parallel. Inset: circuit diagram. d) Photograph showing an LCD powered by four nanogenerators connected in series. Reproduced with permission.[29] Copyright 2017, Nature Publishing Group.

of silver by using the nanogenerator as a power source was realized.[30]

Conclusions and Perspectives

In recent years, water energy harvesting of carbon nanomaterials has made great progress both in fundamental research and technology. Materials with larger surface areas to store more charge can increase output voltage and current in the devices, and the output power has been thus greatly improved by scaling-up the carbon nanomaterials such as from micrometer to centimeter. However, at the current stage, the use of nanogenerators is still far away from practical applications. Some issues remain to be solved. One key issue is that unclear working mechanisms must be clarified to further improve the energy harvesting performance. Well-designed experimental setups and proper molecular simulation studies are both required to solve the contradictions, and the role of the metal electrode and the carbon nanomaterials should be addressed.

Another critical issue is that the output voltage and current by the carbon nanomaterials are still not sufficient for large-scale applications. It can be further enhanced by more efficient chemical modification and physical incorporation of the carbon nanomaterials as well as scaling-up assembly. By means of in-series and in-parallel connections, the electric output can be improved, and an efficient structural design of connection and integration are demanded. By integrating the energy harvesting nanogenerators with energy storage devices such as lithium-ion batteries and supercapacitors, the output electricity of the nanogenerators is effectively stored and can be used at any place and time.

Nevertheless, the majority of the results so far demonstrate a promising future of harvesting energy from water in miniaturized and integratable devices. The mechanical and electrochemical energy of water both in a flowing liquid and a floating gaseous state can be converted into electrical power. The concept of nanogenerators based on carbon nanomaterials could possibly lead to the development of energy-harvesting devices with high performance, flow sensors with high sensitivity, miniaturized energy supplies for implantable biomedical devices, and may open up a new direction in microelectronics.

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Conflict of interest

The authors declare no conflict of interest.

Keywords: carbon · carbon nanotubes · energy harvesting · graphene · nanogenerator

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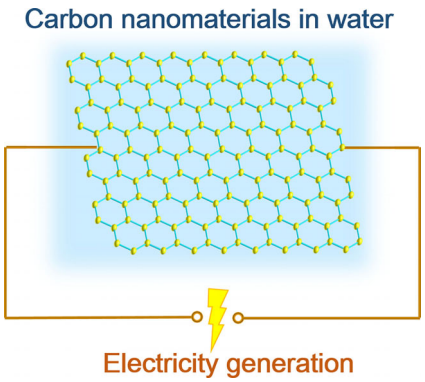
CONCEPT

Electric avenues: Water-induced voltage generation in carbon nanomaterials through the interaction with flowing water as well as moisture has aroused increasing research interest. In this Concept article, the basic principles of the unique power generation and harvesting properties of these nanogenerators are highlighted.



Energy Conversion

Y. Xu, P. Chen, H. Peng**

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Generating Electricity from Water through Carbon Nanomaterials

Electricity generation from water was discovered in carbon-based materials more than ten years ago. Carbon nanomaterials such as carbon nanotubes and graphene can output considerable voltage and current through flowing water and moisture. The working mechanism can be attributed to the interaction among water molecules, ions and the graphitic surface of the carbon nanomaterials. In their Concept article on page ■■ ff. H. Peng, P. Chen and Y. Xu introduce the basic principles of this unique energy-harvesting technique as well as material modification and structural design of these carbon nanomaterial nanogenerators.