Capturing energy from the environment provides the hope for clean energy and enables the formation of self-powered systems. Nanostructured functional materials can interact with water to generate electrical energy, greatly expanding the technical capabilities of water energy harvesting, while those derived from sustainable biomass for this purpose are still in the infancy. Herein, a series of thin self-standing amylopectin-derived membranes of several micrometers can output hydrovoltaic electric energy in the ambient environment. One single-unit flat device (around 0.78 cm²) can generate an instant voltage of up to 0.95 V from high ambient humidity. The underlying mechanism for generating electricity from amylopectin-derived membranes is attributed to the fast adsorption and desorption of water molecules on the membrane surface based on the results of dynamic vapor sorption. Novel moisture-induced miniwindmills as electric generators are fabricated, thanks to these outstanding features such as being self-standing, flexible, lightweight, and having ease of scale production. Such miniwindmill devices with a membrane layer thickness of ≈10 μm can be used to harvest energy with a sustained voltage of around 0.45 V from ambient environment. These results pave the way for developing energy-harvesting powerful miniwindmill devices that exploit water gradients prevalent in nature with biomass materials.

1. Introduction

With the constant threat of the energy dilemma and global warming caused by the overuse of fossil fuels, seeking for green and sustainable energy is one of the most pressing challenges facing human civilization. To address this problem, capturing energy directly from the environment has been deemed as a promising approach as existing abundant low-grade energy can be harnessed directly to generate electricity.[1] As a recyclable resource, water on earth exists as liquid and vapor form in ocean, rivers, lakes, clouds, rain, and as moisture in air. Abundant energy imbued flowing water can be transferred from gravitational or kinetic energy into electricity.[2] Even the energy of dispersing water molecules as moisture in air can be transformed and harvested as electric energy, thanks to the development of nanotechnology.[3] Recent studies have demonstrated that nanomaterials on exposure to atmosphere moisture can generate electricity, and numerous efforts have been made to design moisture-induced devices using various nanomaterials, including carbon materials,[4] metal-derived materials,[5] biofibers,[6] polymers,[7] and composites.[8] Abundant surface functional groups, including out-of-plane large π-bonds, hydroxyl or carboxyl groups, directly determine the polarity and the potential of the ions in the electrical double layer (EDL) and the Debye length.[9] Reversible adsorption and desorption of water moisture in these functional materials brought an anisotropic effect and gave an impressive electric response.[10] When water molecules pass through the narrow capillary channels
formed by the charged active materials, they can induce the oppositely charged counter ions in the EDL within the capillary channels to move at the same time, thereby generating an electric potential. Therefore, the surface properties of such solid materials, which can be modulated by introducing various functional groups, are the main factors affecting their interaction with moisture and resulting output power.

Natural polysaccharides with readily modifiable hydroxyl groups and polymer features, as exemplified by starch, can be excellent candidates as sustainable materials for preparing defined nanostructures. At the same time, they are associated with many advantages, such as abundant source, low cost, complete biodegradability, as well as renewability. The starch macromolecules consist of repetitive anhydroglucose units connected by $\alpha$-1, 4-linkages in amylose chains and additional $\alpha$-1,6-linkages in branched amylopectin. In particular, amylopectin as the major component in starch is a high-density multiple-branched polysaccharide with a cluster of pending chains, which provide the high flexibility and mobility for chemical modification and structure formation. Nevertheless, the use of unmodified amylopectin for harvesting energy from moisture can be severely restricted due to the undesirable textural changes during the syneresis between the glucan chains and water phases. The introduction of functional groups into amylopectin macromolecules could reinforce the ions migration at the water–solid interface on account of the release of protons derived from the reaction of water molecules with surface functional groups. However, the use of such biomass-derived compounds for hydrovoltaic energy generation is still in the infancy. On the other side, the combination of sustainable biomass and such economically promising energy generation is of great interest for our current societal development.

Herein, we demonstrate a series of membranes with chemically modified amylopectin, which undergo an invertible electric response upon moisture and generate electricity. To realize energy harvesting triggered by moisture, a stationary electric generator was fabricated that showed successive electricity-generating behavior upon periodic moisture trigger. The underlying mechanism of amylopectin-derived membranes for generating electricity was analyzed with the help of DVS. Based on the flexibility, robust, and readily shape-programming characters of these amylopectin-derived membranes, a type of self-standing miniwindmills was fabricated to collect electricity utilizing artificial sea wind with high relative humidity. The results demonstrated for the first time that flexible amylopectin-derived membranes could be applied for constant moisture-induced electric generation. We believe that such flexible amylopectin-derived membranes would open new doors to a wide variety of applications in portable smart devices using sustainable biobased compounds.

2. Results and Discussion

2.1. Fabrication of Amylopectin-Based Membranes

Amylopectin as a polysaccharide is a typical bio-based macromolecule with branched chains composed of anhydroglucose units (Figure 1a). They contain abundant secondary hydroxyl groups which can be easily oxidized by periodate into aldehyde groups for further functionalization. The obtained 2,3-dialdehyde amylopectin (DAP) can easily form Schiff base bonds with aromatic amines under basic conditions. Melamine with three primary amines was selected to react with aldehyde groups in DAP. The membranes were obtained by solvent casting the mixture of DAP and different amounts of melamine. For 100 mg DAP, the amount of melamine of up to 10 mg led to transparent and continuous membranes, while too much melamine of 15 mg was not conducive for membrane formation (Figure S2, Supporting Information). The thickness of obtained DAP-MA membranes was controlled to be around 10 $\mu$m (Figure 1a).

![Figure 1. Schematic illustration for moisture-induced electric generation using DAP-MA membranes. a) Photo and SEM image of a DAP-MA membrane with the chemical structure of Schiff base between melamine and amylopectin. Diagrams showing b) the flexible free-standing membranes and c) fixed membranes as devices for moisture-induced electric generation and experimental setup for measurement.](image-url)
and an internal dense structure in the cross section. The DAP-MA membranes contain C=N groups according to Fourier-transform infrared spectroscopy (FTIR) measurement (Figure S1, Supporting Information). The peaks at \( \approx 1601 \text{ cm}^{-1} \) in the FTIR spectrum of DAP-MA membranes belong to C=N stretching vibrations, and the intensity of the peak at \( 1731 \text{ cm}^{-1} \) decreases due to the consumption of aldehyde groups.

These amylopectin-derived DAP-MA membranes were further used for moisture-induced electric generation. They are generally flexible with moisture-responsive behaviors (Figure 1b). In order to avoid the influence of the movement of the membranes on the electricity generation,[17] the DAP-MA membranes were sandwiched between two current collectors and fixed on the glass to further investigate their response to moisture and thus the generation of electricity (Figure 1c). A representative optical image is shown in Figure S3, Supporting Information.

### 2.2. Moisture-Induced Electricity Generation

A sequence of moisture-induced constant open-circuit voltages of \( \approx 0.41, 0.62, \) and \( 0.95 \text{ V} \) was delivered by membranes made of DAP, DAP-MA5, and DAP-MA10, respectively. The moisture-induced voltage rose obviously with higher melamine amounts, when the variation of relative humidity (\( \Delta \text{RH} \)), that is, by spraying moisture toward membranes and letting them evaporate to environmental RH, was adjusted to \( \approx 69\% \) with an interval of 30 s (Figure 2a). Because the membranes were fixed during the electric generation, the moisture-induced electric generation process should be directly related to the interaction between the amylopectin-derived membranes and water molecules. The difference in the degree of hydration of the two sides of the DAP-MA membranes leads to the gradient distribution of hydrons, which will move directionally with the deeper penetration of water. In comparison, the charged groups attached to the amylopectin are immobile. Hence, substantial confinement of charged groups and hydron transport achieve positive/negative charge separation to induce an electric field. The generated electricity is transmitted to the external load through the copper and aluminum electrodes, which act as current collectors and are satisfied for the test. This is analogous to the electric generation of carbon-based materials subjected to moisture diffusion or water evaporation.[18] The amylopectin-derived membranes could spontaneously dissociate and release moveable hydrogen ions with each moisture stimulus on the exposed side, thus generating an immediate electric signal. After removing the moisture stimulus, the moisture gradient across the membranes tends to be balanced out, resulting in the disappearance of the signal.

![Figure 2](https://www.advancedsciencenews.com)

Figure 2. Performance of various DAP-MA membranes for moisture-induced electric generation. a) Open-circuit voltages of DAP-MA membranes with distinct amounts of melamine. Open-circuit voltages of DAP-MA10 membranes b) with different spraying time intervals with the \( \Delta \text{RH} \) of 64.3\% and c) in response to different \( \Delta \text{RH} \) from 64.3\% to 32.4\% with the same spraying time interval of 10 s. d) The performance of the DAP-MA10 membranes compared with other representative materials, for example, GO, TiO2, PSSA, GQDs, and 3D PPY, for moisture-induced electric generation.
The reversible electricity generation of DAP-MA membrane can thus be achieved by water dehydration and subsequent water adsorption repeatedly. In particular, the sharp peaks for the electrical response signals induced by repeated moisture delivery demonstrate that water molecules should be bound to amylopectin-derived membranes, passed through, and/or desorbed quickly within a very short period of time.

In order to investigate the adsorption–desorption process of water molecules and their effects on the electric generation, different frequencies for the delivery of water moisture toward the DAP-MA10 membranes were applied with different time intervals. It is worth to emphasize that the time intervals of 10, 20, or 30 s for spraying moisture onto the membrane did not strongly affect the voltage output (Figure 2b). Therefore, the water moisture can quickly adsorb and leave the surface, leading to the voltage signal, as long as the simulated humidity difference appears. The signal, and thus the membrane surface potential, can be restored to the original state within 10 s due to the desorption of the water molecules on surface.

In addition to the time interval of spraying moisture as incoming stimuli representing various desorption speeds of water molecules from the membrane surface, the extent of ΔRH on the membrane surface, that is, the amount of evaporated moisture from ≈100% RH after delivering moisture and subsequent evaporation to environmental RH, was also essential for the generation of voltage output. These different ΔRH were realized by controlling the surrounding RH with saturated salt aqueous solutions (LiBr of 6.3%, LiCl of 11.1%, CH₃COOK of 22.5%, K₂CO₃ of 43.0%, or NaCl of 75.4%), and the accuracy was ±2.0% under ambient temperature. Before each experiment, the DAP-MA10 membranes as devices were treated at 80 °C for 30 min to remove the inside moisture and equilibrated in surroundings with corresponding RH. Once these DAP-MA10 membranes were subjected to ΔRH by taking the sprayed moisture and following desorption of water molecules, the open-circuit voltage output was observed. Subsequently, when the sprayed moisture disappeared, the moisture gradient on the surface went back to the initial state and the voltage signal disappeared simultaneously. Even when the membranes were exposed to less ΔRH of 52.9%, 36.7%, or 32.4%, DAP-MA10 membranes still could capture moisture and deliver the output voltages of correspondingly 0.65, 0.33, and 0.21 V (Figure 2c). Notably, as-prepared DAP-MA10 membranes showed comparable or superior voltage output, compared with currently widely used materials for moisture-induced electric generation (Figure 2d).

### 2.3. Dynamic Moisture-Induced Process

DVS analysis was carried out to elucidate the adsorption–desorption capacity of DAP-MA10 membranes. The water moisture sorption increased rapidly at relatively low RH ranges originating from the occupation of the primary sorption sites (Figure 3a). The curve of sorption process was fit by the Guggenheim–Anderson–de Boer (GAB) model,[20] which is usually expressed as

\[
M = \frac{CKM_0a_w}{(1-Ka_w)(1-CKa_w)}
\]

where \(M\) is the equilibrated water content (g/100 g solid), \(a_w\) is water activity (RH), \(M_0\) is the water content in monolayer (g/100 g solid), \(C\) is the Guggenheim constant that is related to the different enthalpy of monolayer and multilayer sorption, and \(K\) is a constant related to the different sorption heats of multilayer water and bulk liquid water. The sorption curve of DAP-MA10 membranes was fit by the GAB model with \(R^2\) of 0.998 and \(C, K, M_0\) of 17.38, 0.65, and 10.21, respectively. The \(M_0\) value obtained by GAB fitting is larger, indicating that there are a large number of primary adsorption sites for monolayer adsorption of water molecules on DAP-MA10 membrane surface and polymer chain surface. This could be explained by the presence of sufficient polar groups, such as remaining hydroxyl and primary amine groups. Thus, the membranes were able to capture water easily from moist air. On the other side, the DAP-MA10 membranes exhibited very weak hydrophilicity with a static water contact angle of 81.7° ± 2.4°, which indicates effective water desorption capacity. Moreover, the moisture sorption of DAP-MA10 membranes rose gradually without a distinctive “knee” point, which implied the continuous deposition of water molecules onto the immobilized water layers. The rapid increase of sorption capacity in a high-RH range was mainly attributed to the existence of bilayer hydrated shell, and the equilibrated water...
content of adsorption reached 24.8 wt% at 92% RH. During the following desorption process, water molecules were slowly released at first at high-RH regions and more rapidly at low-RH regions. Finally, the desorption curve ended up in the negative range at 0% RH, which should be due to water sorption within the DAP-MA10 membrane during storage in the ambient environment.

This absorption of water molecules into and their release from DAP-MA10 membranes resulted in electric generation. According to the definition of ion gradient diffusion, an ionic current is produced when these charged ions move in response to a gradient in the concentration of surface functional groups or as a result of the asymmetric introduction of liquid into the system.\[^{10c,21}\] Within the DAP-MA10 membranes, the water molecules absorbed onto the polymer chain surface should have interacted with surface functional groups, such as –OH and –NH\(_2\) groups, and liberated polarized ions instantly, which should go back to initial states, accompanying the release of water molecules. During this short-term dissociation and reassociation of polarized ions, the electrical output is generated and enhanced by introduced asymmetric moisture concentration across the membrane (Figure 3b). Since the electrical output of moisture-induced electric generation should be based on ion diffusion, the strategies by enhancing the production of polarized ions via the reaction with surface functional groups and adjusting the concentration gradient of the polarized ions can be used to maximize the diffusion current.\[^{19a}\]

### 2.4. Moisture-Responsive Mechanical Properties of DAP-MA Membranes

The unique compact structure and perfect compatibility endowed the self-standing DAP-derived membranes with superior mechanical properties (Figure 4a). It is obvious that the DAP-MA membranes showed improved tensile strengths and Young's modulus. With higher amount of melamine, DAP-MA10 membranes showed higher tensile strengths of around 75 MPa and Young’s modulus of around 2.5 GPa. The DAP-MA membranes were generally mechanically robust, which was ascribed to the crosslinked networks of DAP by melamine molecules. Dynamic mechanical thermal analysis (DMTA) was further adopted to study the effect of moisture on the dynamic mechanical properties of these self-standing membranes (Figure 4b). DAP-MA membranes showed generally higher storage modulus than DAP membranes over the whole range of RH between 20% and 90%. At the same time, the storage moduli (\(E'\)) and loss moduli (\(E''\)) maintained invariability with the increase of RH from 20% to 90%, at 25 °C and a frequency of 10 Hz (Figure 4b). These amyllopectin-derived membranes exhibited excellent stability even after heating in water at 80 °C for 10 h (Figure S4, Supporting Information). Therefore, enhanced mechanical properties of DAP-MA membranes due to the crosslinking with the melamine remained stable even in atmospheres with high RH, demonstrating their high stability and thus the application potential in surroundings with high moisture contents.

It’s worth noting that the shape of the membrane can be programmed as required. The moisture responsiveness of the membranes can be initiated in an environment with RH gradient and by designing self-standing functional devices for special movements, such as lifter and windmill. For example, a lifter was fabricated by origami folding using the DAP-MA10 membrane strips as “arm” (Figure 4c and Supporting Movie S1, Supporting Information). It could lift and put down objects using moisture as trigger. A sponge with a weight of 32.3 mg, which is ≈13 times of the weight of DAP-MA10 strips (≈2.5 mg), was

![Figure 4](https://example.com/figure4.png)

**Figure 4.** Moisture-responsive mechanical properties. a) Tensile strength and b) humidity sweeps at 25 °C with a frequency of 10 Hz of DAP, DAP-MA5, and DAP-MA10 membranes. c) Photographs showing moisture-driven lifting up of an object by DAP-MA10 membrane under ambient conditions. Scale bars: 1 cm.
lifted up using moisture as stimuli and returned to its initial status once the moisture was removed. This readily programmed shapes of DAP-MA10 membranes with their maintained moisture-responsive mechanical properties as well as their stability over a wide range of RH allow them to be further used in suitable shapes for moisture-induced energy harvesting.

2.5. Flexible Energy-Generating Miniwindmills

Miniwindmills were then made of DAP-MA10 membranes and applied for flexible moisture-induced electric generation (Figure 5). Such miniwindmills of DAP-MA10 membranes were assembled via origami folding, thanks to the robustness of DAP-MA10 membranes (Figure 5a,b). The miniwindmills did not obviously output energy when they were kept in ambient surroundings without RH change. Once exposed to moisture using artificial sea wind, the miniwindmill rotated immediately and delivered output signal that finally reached a voltage of 0.4 V (Supporting Movie S2, Supporting Information). By alternatively delivering artificial sea (‘on’) wind and stopping (‘off’) the delivery, the miniwindmills can correspondingly start and stop the energy generation for many cycles (Figure 5c). In addition, after the “on–off” cycles, the energy-generating miniwindmills can still maintain stable output performance for a long time, as long as the artificial sea wind was continuously breezed to the miniwindmills (Supporting Movie S2, Supporting Information). These results demonstrate that the miniwindmills of DAP-MA10 membranes could be used for stable moisture-induced electricity collection from sea wind with high moisture contents.

3. Conclusion

In summary, we fabricated flexible and robust amyllopectin-derived membranes for moisture-induced energy harvesting directly from the ambient environment. At ΔRH of 69%, one single-unit of DAP-MA10 membrane with an area of 0.78 cm² delivered an instant constant voltage of up to 0.95 V. The underlying mechanism of electric generation from amyllopectin-derived membranes triggered by moisture should be due to the fast absorption and desorption of water molecules according to DVS measurement. Significantly, these amyllopectin-derived membranes are mechanically flexible, which makes them unique among moisture-induced electric generators and therefore allows to program their shapes easily for different application scenarios. Accordingly, novel miniwindmills of DAP-MA10 membranes, as demonstrated for practical applications in wind power devices, delivered a sustained voltage of around 0.45 V for a long time. Combining these advantageous properties, amyllopectin-derived membranes are promising sustainable biomass-based substrates for collecting moisture-induced electric energy from ambient environment.

4. Experimental Section

Materials: Amylopectin from maize, sodium periodate (NaIO₄), melamine, sodium chloride, ethylene glycol, and hydroxylamine hydrochloride were received from Sigma-Aldrich (Steinheim, Germany). All chemicals were of analytical grade or higher and used without any purification. Preparation of 2,3-Dialdehyde Amylopectin (DAP): DAP was prepared by sodium periodate oxidation of amyllopectin according to our previous
work. In brief, 4 g of amylopectin, 8.5 g of NaIO₄, and 10.1 g of NaCl were dispersed in 200 mL of deionized water. The mixture was stirred at 75 °C for 3 h in dark, followed by adding ethylene glycol (20 mL) to quench oxidation. Obtained suspensions were dialyzed in a dialysis membrane with a molecular weight cutoff of 3500 Da. Obtained white suspensions were heated at 90 °C for further 3 h until an almost transparent solution emerged. The small number of remaining solids was removed by centrifugation at 14 000 rpm (25 °C) for 30 min. Resulting transparent DAP solutions were then concentrated to 20 mg mL⁻¹ under reduced pressure and stored at 4 °C for further use. The degree of oxidation of the obtained DAP was measured to be 1.81 ± 0.04 by titration with hydroxylamine hydrochloride.

Melamine-Modified DAP Membranes: The DAP-derived membranes were fabricated by solvent casting of a mixed solution of DAP with melamine. In brief, a certain amount of melamine powder (0, 5, 10, 15 mg) was added into 5 mL of DAP solutions. The mixtures were vortexed for 30 min and ultrasonically treated for another 5 min. Then, the solutions were pipetted into a Petri dish for solvent casting at ambient temperature. To obtain the membranes DAP-MAx (x = 0, 5, 10, 15) with uniform thickness, the Petri dishes were adjusted to a nontilting state using a bubble level. The thickness of the obtained membranes was >10 μm.

FTIR Measurement: FTIR was implemented on a FTIR spectrometer (Bruker, Germany) ranging from 4000 to 500 cm⁻¹ and accumulated 24 scans.

Scanning Electron Microscope: The images of the samples were captured by using SEM Leo SUPRA 35 (Cal Zeiss SMT GmbH, Oberkochen, Germany). A layer of gold/palladium was sputtered on the surface of the samples before observation.

Dynamic Mechanical Thermal Analysis (DMTA): DMTA was carried out on a DMTA system (GABO EPLEXOR, NETZSCH) equipped with a 150 N force sensor. The DAP-derived membranes with the size of 20 × 5 mm were placed in the machine with a clamp distance of 8 mm for measurement. The uniaxial stretching was performed at a constant rate of 2 mm min⁻¹ until rupture. The mechanical performances were recorded as the average values from the stress–strain curves of at least three parallel trials that were fractured in the middle of the specimens.

Dynamic Vapor Sorption (DVS): The dynamic water vapor sorption behaviors of DAP-derived membranes were evaluated on a DVS apparatus (DVS Advantage, Surface Measurement Systems). All measurements were carried out at constant temperature of 20 °C and a nitrogen flow of 200 sccm. DAP-derived membranes with a mass of about 10 mg were used for the measurements. The measurements were carried out at 20 °C with RH first reduced to 0% until equilibrium with the mass variation \(< 0.002%\) min⁻¹ and the initial dry mass was recorded. Then, the RH was increased from 5 to 95% with a step size of 10% for the sorption process. The desorption was subsequently performed with a step size reduction of RH of 10%.

Static Water Contact Angle Test: Static water contact angles were measured on a contact angle system (KRUSS) using the sessile droplet method with a dosing volume of 2.0 μL deionized water. The result was acquired by taking the average value from more than three measurements.

Manipulation of Moisture-Induced Lifter: The experiment was performed in a home-made humidity chamber. The membrane was cut into a strip with a dimension of 30 × 5 mm, which was folded into a rectangular groove in the middle. The strip was vertically placed and then a load hung at the bottom. The wet paper was placed close to one side of the membrane, resulting in an asymmetrical structure, which could lift up the object. The bending/unbending process of the membrane was recorded by smartphone.

Fabrication of the Devices with DAP-Derived Membranes: To fabricate the moisture-induced devices, the glass slide was prepared as the substrate. The aluminum foil was fixed on the glass slide with double-sided tape and it worked as one electrode. Then, the membrane was glued to the surface of aluminum foil through a circular conductive tape with a diameter of 1 cm. Subsequently, the protective layer of enameled copper line to be in contact with the conductive layer was removed and installed in the middle via double-sided tape with the dimension of 2.5 × 2.5 mm for electrical measurements.

Electromechanical Test of the Moisture-Induced Electricity: RH inside the home-made chamber was adjusted with saturated aqueous solutions of KCl, NaCl, K₂CO₃, CH₃COOK, LiCl, and LiBr, which yielded RHs of 84.2%, 75.4%, 43.0%, 22.5%, 11.1%, and 6.3%, respectively, under ambient temperature. The SP-300 Potentiostats of BioLogic was used to measure the voltage generated by the membranes.

Fabrication of Miniwindmills Using DAP-MA Membranes: The DAP-MA10 membranes with a dimension of 30 × 30 mm was cut along the predrawn lines, followed by folding them into windmill shape using an origami process. The output of the DAP-MA10 miniwindmills was tested under a simulated high real environmental RH using saturated NaCl solution. The electric generation was recorded on potentiostats.

Supporting Information

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

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

amylopectin, electric generation, membranes, moisture induced

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