Hydrogen Production from the Air

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Hydrogen Production from the Air

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Abstract

Hydrogen gas (H₂) produced by water splitting using renewable energy, namely green hydrogen, is the most promising energy carrier of the low-carbon economy¹⁻⁶. However, the geographic mismatch between renewables distribution and freshwater availability poses a significant challenge to green hydrogen production⁷⁻⁹. Here, we demonstrate a method of directly producing H₂ from the air, namely, capturing freshwater from the atmosphere using hygroscopic electrolyte and converting it to H₂ by electrolysis powered by solar energy. A prototype H₂ generator has been successfully established and operated for 12 consecutive days with a stable performance at an average Faradaic efficiency around 95%. This so-called direct air electrolysis (DAE) module can work under low relative humidity (20%) environment, overcoming water supply issues and producing green hydrogen sustainably with minimal impact to the environment. The DAE modules can be easily scaled to provide H₂ to remote, arid/semi-arid, and scattered areas.

Main

Hydrogen is the ultimate clean energy. Despite being the most abundant element in the universe, hydrogen exists on the earth mainly in compounds like water. H_2 produced by water electrolysis using renewable energy, namely, the green hydrogen, represents the most promising energy carrier of the low-carbon economy¹⁻³. H_2 can also be used as a medium of energy storage for intermittent energies such as solar, wind, and tidal⁴⁻⁶.

The deployment of water electrolyzer is geographically constrained by the availability of freshwater, which, however, can be a scarce commodity. More than one-third of the earth's land surface is arid or semi-arid, supporting 20% of the world's population, where freshwater is extremely difficult to access for daily life, let alone electrolysis^{7,8}. In the meanwhile, water scarcity has been exacerbated by pollution, industrial consumption, and global warming. Desalination may be used to facilitate water electrolysis in coastal areas, however, substantially increasing the cost and complexity of hydrogen production. On the other hand, areas rich in renewable energies are commonly short in water supply. Fig.1 shows a distinctive geographic match between the availability of solar power and the shortage of freshwater in the majority of the continents, such as North Africa, West and Central Asia, Midwest of Oceania, and southwest of North America⁹.

Fig.1 Superimposed atlas of water risk and solar energy potential. Separate maps available in Fig. S1.

Few studies have been trying to mitigate the water shortage for electrolysis. Direct saline splitting can produce

hydrogen, which, however, faces a serious challenge of handling chlorine byproduct^{10,11}. Some proton/anion exchange membrane electrolyzers can use vapor carried by inert gases as a feed; however, the electrolyzers must operate in an air-free atmosphere¹²⁻¹⁶.

In this work, we corroborate that moisture in the air can directly be used for hydrogen production via electrolysis, owing to its universal availability and natural inexhaustibility¹⁷⁻²¹ – there are 13 trillion tons of water in air which is in a dynamic equilibrium with the aqua-sphere. For example, even in the Sahel desert, the average relative humidity (R.H.) is about 20 %²², and the average daytime R.H. at Uluru (Ayers Rock) in the central desert of Australia is 21%²³. Considering deliquescent materials such as potassium hydroxide, sulfuric acid, propylene glycol^{24,25} can absorb water vapor from the air at an R.H. below 20%, here, we demonstrate a method to produce hydrogen by electrolyzing hygroscopic solutions exposed to air. The electrolyzer operates steadily under a wide range of R.H., as low as 20%, while producing high purity hydrogen with a Faradaic efficiency over 95% for more than 12 consecutive days, without any input of liquid water. A solar-driven prototype with five parallel electrolyzers has been devised to work in the open air, achieving an average hydrogen generation rate of 745 L H₂/day/m² cathode, opening up a sustainable pathway to produce green hydrogen without consuming liquid water.

Results

Design of the Direct Air Electrolysis (DAE) module for hydrogen production

Hydrogen production from the air was realized through our DAE module. As shown in the sandwich structure in Fig. 2, this module consists of a water harvesting unit in the middle and electrodes on both sides paired with gas collectors, powered by an external supply, for example, a solar panel. Importantly, the water harvesting unit also serves as the reservoir to hold the electrolyte. Porous medium such as sintered glass foam is soaked with deliquescent ionic substance to absorb moisture from the air via the exposed surfaces. The captured water in liquid phase is transferred to the surfaces of the electrodes via diffusion and subsequently split into hydrogen and oxygen *in situ*. Quartz wool is tightly packed in between the glass foam and the electrodes to ensure the connectivity of the aqueous phase (Fig. S2-S4). The glass foam filled with ionic solutions forms a physical barrier that effectively isolates hydrogen and oxygen from mixing.

Fig.2 The concept of direct air electrolysis (DAE) for hydrogen production. a) A schematic diagram of the DAE module with a water harvesting unit made of porous medium soaked with hygroscopic ionic solution. **b)** Equilibrium water uptakes of hygroscopic solutions at different R.H.^{24,25}.

Deliquescent substances characterized with a strong affinity with water tend to extract moisture from the

atmosphere at exposure, absorbing sufficient water to form an aqueous solution which is hygroscopic in nature. When the chemical potential (μ) of water vapor in the atmosphere is greater than that in a hygroscopic solution, i.e. $\mu_{\text{air}} > \mu_{\text{solution}}$, the solution will continue absorbing water vapor and being diluted until the vapor-liquid equilibrium is reached at $\mu_{\text{air}} = \mu_{\text{solution}}$ ²⁶, making the concentration of the solution C equal to the equilibrium one C^* . In this study, we tested several deliquescent materials, including KOH, CH₃COOK, and H₂SO₄, representing a base, a salt, and an acid, respectively. All three materials spontaneously absorb moisture and form ionic electrolytes. It was found that the direct air electrolysis modules using the respective electrolytes were able to produce hydrogen gases successfully for an extended period with a continual supply of air and power. However, the hydrogen evolution performance of the DAE module with KOH started to decline after 72 hr and stopped at 96 hr. It was observed that the voltage of the DAE module increased from 2.30 V to 2.40 V due to the gradual conversion of KOH into K₂CO₃ and eventually KHCO₃ at exposure to CO₂ in the air. KHCO₃ is less soluble in water hence less conductive as an electrolyte, and critically it is non-deliquescent. For CH₃COOK based DAE module, the voltage was as high as 3.70 V due to the large size of acetate anions and substantial ethane byproduct found along with O₂ at the anode (Fig. S5).

Sulfuric acid was identified as one of the best hygroscopic materials that can absorb water from the air down to concentrations of 10 wt% or below²⁷. Meanwhile, the sulfuric acid solutions are high in conductivity (0.61 S cm⁻¹ at 50.0 wt%)^{28,29}, non-volatile, and it is non-toxic to the environment. In this regard, the following studies were carried out with sulfuric acid electrolyte equipped with platinum (Pt) mesh electrodes (Fig. S6). It is also interesting to note that in the concentration range of sulfuric acid of this work, the corresponding freezing point is below -30 °C³⁰, implying potential working temperature under icing environment.

Performance of the DAE module

The DAE module's performance was investigated using current density (J) and voltage (V) characteristic experiments conducted at 25 °C. The effect of relative humidity ranging from 20% to 80%, as well as the pore size and thickness of the sintered glass foams, were studied. Sintered glass foams were labelled as G1, G2 and G3 corresponding to the pore size of 50–70, 30–50, 16–30 μm , respectively. Also, a series of experiments with extended time durations of 288 hr was conducted to investigate the stability of the DAE module.

The effect of the different pore size of sintered glass foams on the J-V behavior is shown in Fig. 3a, using 62.0 wt% H_2SO_4 solution as electrolytes. Current density is negligible ($<1 \text{ mA cm}^{-2}$) at a voltage below 2.0 V due to the overpotential of the Pt mesh. As long as the capillary force still holds the electrolyte, the current density increases with the use of larger pored sintered glass foams, indicating higher conductivity and energy efficiency for overall water splitting due to better mobility of electrolyte in larger pores. At 3.0 V, a current density of 27.1 mA cm^{-2} was achieved using G3 sintered glass foam and it increased to 36.5 mA cm^{-2} using G1 sintered glass foam. Therefore, the G1 sintered glass foam was chosen for further study of the foam thickness, owing to the high electrical conductivity, low resistance, and high energy efficiency it brings to the DAE module.

The sintered glass foam's thickness also plays a role in the J-V behavior. As shown in Fig. 3b, the J-V curve shifts upwards with decreasing glass foam thickness. At 3.0 V, the current densities are 17.5 and 36.5 mA cm^{-2} while using 2.5 and 1.5 cm thickness G1 sintered glass foams, respectively. According to Pouillet's law³¹, the resistance is proportional to the distance between the electrodes, suggesting that a large distance between the cathode and anode contributed to high resistance for overall water splitting. Hence, under specific current density, the gap between two electrodes should be as small as possible to maintain relatively high energy efficiency. However, the mass transfer area for water absorption is proportional to the sintered glass foam's thickness. There is a trade-off between the water absorption area and conductivity. Considering both factors,

we chose the G1 sintered glass foam with 1.5 cm total thickness for further investigation, given that it could provide sufficient mass transfer area for air-electrolyte contact while maintaining moderate energy efficiency. The observed experimental concentration of sulfuric acid C is constantly above its equilibrium concentration C^* during the direct air electrolysis process. This difference represents the driving force for the mass transfer of water from the vapor phase into electrolyte solution and then onto the electrochemical reaction sites at the electrodes. Fig. 3c shows that at $J=15.0 \text{ mA cm}^{-2}$, the experimental concentration in the DAE module is approximately 5wt% higher than the equilibrium at steady state, which means a stable in-situ H_2SO_4 concentration over 8 hours under a constant current density, where the rate of water absorption from air equals the rate of water consumption by electrolysis. Likewise, such steady-state mass transfer driving force can be established at fixed air relative humidity. As shown in Fig.3c inset, the driving force increases proportionally with the increase of current density, which means the rate of water absorbed by the DAE module rises when the water electrolysis rate is turned up. For instance, at R.H. = 80%, if a minimal current density is applied, the sulfuric acid concentration in the module is close to the equilibrium $C_{\text{H}_2\text{SO}_4}^* = 26.8 \text{ wt}\%$, and the mass transfer driving force of water absorption is nearly zero. If we increase the current density J to 70 mA cm^{-2} , the steady-state concentration of sulfuric acid is increased to 46.7 wt%, 75 % higher than the equilibrium one $C_{\text{H}_2\text{SO}_4}^* = 26.8 \text{ wt}\%$. Therefore, our DAE module is intrinsically self-converged, compatible with a broad range of air humidity and current density.

The DAE module's J-V behavior has also been studied under different H_2SO_4 concentrations (Fig. 3d). With the decrease of H_2SO_4 concentration from 62.5 wt% to 32.6 wt%, the current density for the electrolysis reaction increases significantly from 37.5 mA cm^{-2} to 97.0 mA cm^{-2} , under a constant voltage of 3.0 V. Such change can be attributed to the improved electrical conductivity of diluted H_2SO_4 (Fig. S7)^{28,29}. Also, the viscosity of the electrolyte decreases as the acid is being diluted (Fig. S8), resulting in higher electrocatalytic activity and reduced electrochemical polarization^{32,33}.

Fig.3 DAE module performance at 25 °C.

a) J–V curves for modules using various porous substrate with different pore sizes. **b)** Effect of foam thickness on J–V performance.

c) The experimental steady state concentration of sulfuric acid at $J=15.0 \text{ mA cm}^{-2}$ (red) VS equilibrium concentration $C_{\text{H}_2\text{SO}_4}^*$

(black) under different R.H.. The inset shows the effect of current density on steady state concentration under 80% R.H. where

$C_{\text{H}_2\text{SO}_4}^* = 26.8 \text{ wt\%}$ (red dashed line). **d)** The effect of electrolyte concentration on J–V performance. **e)** Example recording of cell

voltage (black symbol), H_2SO_4 concentration (red symbol) and $C_{\text{H}_2\text{SO}_4}^* = 47.7 \text{ wt\%}$ (red dashed line) for DAE modules at constant

current density of 15.0 mA cm^{-2} for 288 hr at 40 % R.H.. **f)** Current density collected under different voltage for 288 hr at 40 % R.H.. Fig. 3c to 3f were operated with G1 sintered glass foam and 1.5 cm gap between the two electrodes.

The DAE module was found stable during continual electrolysis. Performance of the electrolysis cell at various voltage, energy efficiency, and air R.H. are shown in Table S1 and Fig. S9. After a minor fluctuation initially, the J-V behavior stabilize for a 48 hr run. For further laboratory test, we chose 40% R.H. air as the gas atmosphere condition. As shown in Fig. 3e, the concentration of H_2SO_4 fed to the module was 55.0 wt% initially, and it converged to 51.1 wt% over the first 120 hr. In the following 168 hr, the electrolyte concentration, the DAE module's voltage, and the mass transfer driving force for moisture absorption ($\Delta C = C_{\text{exp}}[51.1 \text{ wt\%}] - C^*[47.7 \text{ wt\%}] = 3.4 \text{ wt\%}$) all stabilized. Accordingly, the corresponding current densities collected under specific voltages (2.4, 2.7, 3.0 V) also reached steady state in this 12 day continual operation (Fig. 3f). This result indicates excellent adaptability and long-term stability of the DAE modules under different air R.H., cell voltage, and electrolyte concentrations.

Demonstration of DAE modules stack with solar panel in the open air

To further demonstrate the DAE module's working capability in a practical environment, we designed and constructed a free-standing hydrogen generation tower consisting of five DAE modules stacked in parallel superimposed vertically with a solar panel for power supply. The details of the structure of the tower are shown in Fig. 4a and Fig. S10. One of the advantages of such design is that the footprint of the tower is no more than the solar panel, meaning our DAE will not occupy extra land, which can be costly in some areas. The tower was tested for two days, 8 hr per day, in the open air of a hot-dry summer (Mediterranean climate) in the campus of the University of Melbourne. The setup of the module of the outdoor surroundings is provided in Video S1. The outdoor temperature varied from 20°C to 40°C , and the relative humidity ranged from 20% to 40% over the testing period. Since the solar panel was used as a renewable energy supply, the voltage, and the current of each DAE module were solely determined by solar intensity (Video S2), which varies every

hour. The product hydrogen gas evolved from the cathode was collected in an inverted, water-filled cylinder over water, which was then used to examine the gas production rate (Video S3). The oxygen generated on the anode of the DAE was vented into the air.

The performance of the tower was shown in Fig. 4b and Fig. S11, in the form of the hydrogen generation rate, hydrogen evolution Faradaic efficiency (η_{f,H_2}), the overall current and the voltage. During the open-air demonstration, η_{f,H_2} was over 90% during the daytime, shown as the red line in Fig. 4b. On the first day, when the weather was sunny, the current output was stable around 400 mA and voltage 2.68 V. The hydrogen evolution rate was 186 ml h⁻¹, with the total hydrogen production at 1490 ml in a day, which is equivalent to 745 L H₂/day/m² of the cathode, or 3.7 m³ H₂/day/m² tower.

On the second day, a few hours of good sunlight guaranteed the current output stable at 400 mA from 9:00 to 13:00, with an average hydrogen generation rate of about 179 ml h⁻¹, similar to that of the first day. However, in the early morning from 8:00 to 9:00, the solar intensity was limited, resulting in a relatively lower current output of 270 - 370 mA and a hydrogen generation rate of 140 ml h⁻¹. In the cloudy late afternoon (14:00 to 16:00), the poor weather conditions reduced the solar panel's current output to as low as 50 mA, and hence, the hydrogen generation rate dropped to 21 ml h⁻¹. On the whole, under non-ideal weather conditions, the total hydrogen production could still reach 1188 ml on the second day.

The gas product collected from the cathode has been analyzed with gas chromatography (GC.), suggesting pure hydrogen. The gas produced from the anode has also been measured (Fig. S12) with a GC. showing it is a high purity oxygen. The Faradaic efficiency of oxygen η_{f,O_2} at the anode is over 91.0%. Since the Faradaic efficiency of both H₂ and O₂ measured and calculated by energy and mass balances are comparable, we can confirm the overall electrolysis is a water-splitting process.

Fig.4 Open-air demonstration of the hydrogen generation tower in Dec 2020 Melbourne. a) Photo of the designed hydrogen generation tower featuring scalable DAE modules with minimized footprint. b) Hydrogen generation rate and Faradaic efficiency on an hourly basis at corresponding air humidity and temperature for two days under different weather conditions.

Discussion

In summary, to overcome the water shortage problem in the case of widespread deployment of hydrogen production, we have demonstrated a method of producing high purity hydrogen from the air by using hygroscopic electrolyte soaked in a porous medium as the moisture absorbent. Such direct air electrolysis (DAE) module can operate under a broad range of air humidity, electrolyte concentration, and current density, for more than 12 days continuously without any decay and without any attendance, performing at a hydrogen Faradaic efficiency of around 95%. This DAE module can be easily scaled up and integrated with renewable power. In our demonstration, a prototype of standalone hydrogen generation tower with five DAE modules stacked in parallel superimposed vertically under a solar panel (with minimum footprint) was constructed and tested outdoor in a hot-dry summer. The prototype achieved automated steady hydrogen production depending on the solar intensity under varying weather conditions. On a warm sunny day, the hydrogen production rate

can reach $3.7 \text{ m}^3 \text{ H}_2/\text{day}/\text{m}^2$ tower. Such DAE farms, if further upscaled, can generate abundant hydrogen in arid and semi-arid areas with negligible change to the regional air humidity and minimal impact to the environment (Supplementary Note 1).

Method

DAE module fabrication

A self-designed DAE module was used under each R.H. in this project. More details can be found in the Supplementary document. Pt mesh electrodes (99.99% purity, $20 \times 20 \text{ mm} + 0.5 \times 20 \text{ mm}$, Yueci technology co.) with geometric area 4 cm^2 were attached directly to the quartz wool (99.95% purity, 5-10 μm , Xinhua co.), and then connected with the sintered glass surfaces, with geometric area 7.84 cm^2 (Shundao sintered glass foam co.). The sintered glass foams' thickness was 3 mm, and quartz wool was layered between two foams. For example, for 1.5cm total thickness, three foams and four layers of quartz wool were used stacked. Teflon plate with Pt wireline (99.99% purity, Xudong Co. Ltd.) was used as current feeders and electrolyte distributors. After assembly, the DAE module was put inside the climate test chamber (DHT-100-40-P-SD, Shanghai Doaho Co. Ltd.), keeping a close environment at a constant R.H. and temperature. The DAE module connected directly with a D.C. power supply (Wanptek DPS3010U), which could supply constant current for electrolysis. The cathode's output gas production was bubbled through a water bath, and collected in an inverted, liquid-filled cylinder.

Data collection

Another DC power supply (Nice Power R-SPS605D) was used to connect with the DAE module to collect the current density vs. voltage (J–V) performance curve. The current was measured after 30 seconds under each voltage, using an applied voltage from 1.80 V to 3.00 V with a 100-mV increase. The area of the electrode was 4 cm^2 . Under each R.H., the J-V behavior was tested before putting into the environment oven and after operating over 24 hours and 48 hours. Each J-V behavior was verified by repeated measurements three times, with the current densities variation controlled within 5-10%.

Under each R.H., the DAE module was operated under constant current density, and the weight needed to be checked each 4 hr until it maintained stable over an 8-hr period.

Faradaic efficiency

The gas product flowed into a measuring cylinder (25.0 mL) through a rubber pipeline for volume measurement by a collection of gases in an inverted, water-filled cylinder over water. Gas collected inside the cylinder was drawn out by the syringe and then pushed into the gas chromatography (GC.) system (7890B, Agilent technologies) with a thermal conductivity detector (TCD.) for analyzation. The Faradaic efficiencies η_{f,H_2} and η_{f,O_2} were compared to the gas production with the ideal production rate.

Stability test

55.0 wt% H₂SO₄ (formulated with 98% sulfuric acid and R.O. water) was used as the electrolyte and tested under 40% R.H. and 25 °C in the climate test chamber for 12 days under constant current density 15.0 mA cm⁻².

Open air demonstration with solar panel

Five DAE modules was stacked vertically in parallel, and the hydrogen generation was collected. Supporters were used to keep the distance between each unit. Here, a commercial silicon solar panel was connected in series and put on the top of the units, with an open-circuit voltage of around 6.0 V and a short circuit current around 400 mA under Melbourne's natural sunlight. The gas product was also flowed into a measuring cylinder by collecting gases in an inverted, water-filled cylinder over water.

Data availability

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

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Contributions

G.L. conceived the idea. J.G. and Y.Z. conducted the experiments. J.G., G.L., A.Z. and Y.Z. analyzed the data.

K.C. and Y.G. helped with characterizations. J.G. led the draft of the manuscript with input from G.L., A.Z.,

X.F., G.H. and Y.Z..

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Ethics declarations

Competing interests

The authors declare no competing interests.

Supplementary Information

Supplementary Information

This file contains additional data to support the conclusions of the manuscript including details of DAE modules, prototype, and composition of gas production.

It contains Supplementary Fig. S1 to S12, Supplementary Table S1 to S2, Supplementary Note 1, and Supplementary References.

Supplementary Video 1

Prototype of hydrogen generation tower

Supplementary Video 2

Effect of solar intensity

Supplementary Video 3

Real-time hydrogen generation

Supplementary Files

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