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Co-designed dual-ambient energy harvester hydrogel for hydrogen production and electricity generation



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ABSTRACT

In the quest to scavenge and convert environmental energy into useable green power, the development of materials that can efficiently harvest from multiple ambient energy sources remains challenging. Essentially, the reconciliation of differing material and structural requirements necessary for distinct functionalities is still a significant obstacle. In this work, a co-design approach has been taken to tailor a hydrogel capable of meeting the specific demands of two energy conversion processes; solar-induced hydrogen generation and evaporationinduced electricity generation. Specifically, Cu-doped ZnS photocatalysts integrated into a chitosan matrix, featuring internally aligned channels, not only facilitate mass transport but also significantly enhances the hydrogel's photochemical and electrokinetic properties. Dual functionality is attainted through the harmonious combination of light absorption, charge separation, and fluid transport features within a unified material system, embodying a balance between the photophysical reactions for hydrogen production and the capillary flow essential for electricity generation through the streaming potential phenomenon. Finally, the real-world environmental demonstration achieved a hydrogen generation rate of 17.3 mmol m⁻² h⁻¹ under natural sunlight irradiation and a potential output of 192.8 mV. Overall, this study demonstrates strategies applicable to designing other ambient energy harvesters with the potential to convert disparate renewable energy sources for sustainable energy production.

1. Introduction

Ever-increasing global energy demands and rising concerns about global climate issues have brought forth a need to search for renewable energy solutions. Remarkably, the earth's natural environment contains an abundance of a vast range of energies, for example, the daily solar irradiance on the earth far exceeds that of the current annual energy consumption by humankind [1]. At the same time, water covers approximately 71 % of earth's surface and stores an enormous amount of energy in its hydrological cycle [2,3]. Consequently, ambient energy harvesting, which refers to the scavenging and conversion of these environmental energies into usable green electricity or fuels, holds tremendous potential as a promising strategy to alleviate global energy needs.

In particular, photocatalytic H_2 generation has been increasingly recognized as a green pathway for the conversion of solar energy into useful chemical fuel [4–8]. Typically, suspension-type systems in

photocatalytic reactions are more prevalent [9–11]. However, photocatalytic suspensions suffer from certain drawbacks, such as light shielding and scattering effects, aggregation of catalysts particles, the need for mechanical stirring and difficulty in catalyst collection, separation, and recycling [12–14]. To mitigate these issues, researchers have proposed using immobilized-type catalyst, such examples include thin films [15], sheets [16], flat panels [17], photocatalytic gels [18,19] and others [20]. In general, immobilized-type catalyst are believed to enhance performance by reducing light scattering, improve scalability and eliminating the need for continuous agitation [21]. However, further refinement of such immobilized-type catalysts is still required, since they pose a new set of challenges related to their matrix selection, light delivery, gas separation and mass transfer requirements [22].

Additionally, a prominent shortcoming of all devices reliant on solar power is the intermittent nature of the diurnal solar cycle, where generation is limited by the absence of sunshine at night and is also substantially reduced during cloudy or rainy weather [23]. In contrast, the

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ubiquitous nature of the natural hydrological cycle has sparked growing interest in novel hydrovoltaic devices, since these devices can operate regardless of geographical location or weather conditions [3,24]. With the development of water-enabled electricity generation technologies, energy from water droplets [25,26], moisture [27–30], evaporation [31, 32], ambient humidity [33,34], and fluid motion [35,36], can be readily converted into green electricity. Intriguingly, the hybridization of such technologies to a photochemically driven water-splitting device could be a worthwhile pursuit, potentially enabling a secondary means for round-the-clock, on-demand electricity generation. Though admittedly, the conjoining of such systems will likely prove difficult, in part due to the disparate requirements from the standpoint of material selection, structural design and device assembly.

Herein, this work demonstrates a judiciously designed dual-ambient energy harvester hydrogel capable of producing hydrogen and generating electricity from ambient sunlight and natural evaporation. To address the aforementioned challenges, the disparate requirements are reconciled by specifically co-designing the material and structural aspects of the hydrogel to fulfill distinct dual-functional roles for both energy conversion needs. Cu-doped ZnS (Cu-ZnS) photocatalysts are integrated into a chitosan matrix to form a photoactive hydrogel for hydrogen generation, while inherently aligned internal channels are created to facilitate mass-transport pathways. The Cu doping enhances photochemical reactions by improving light absorption and charge separation. Concurrently, natural surface evaporation drives fluid flow through the internal aligned channels, and when coupled with the favorable surface potential of the inorganic Cu-ZnS channel walls, allows for an evaporatively driven electricity generation through the streaming potential phenomenon. Consequently, the feasibility of the dual-ambient energy harvester in real-world environmental conditions is demonstrated, achieving a hydrogen generation rate of 17.3 mmol $m^{-2} h^{-1}$ under natural sunlight irradiation and a potential output of 192.8 mV. Overall, this work represents a promising approach that demonstrates strategies employable in the designing of other multifunctional ambient energy harvesters for the conversion and generation of renewable energy sources into sustainable energy products.

2. Results and discussion

Fig. 1 schematically illustrates the design strategy underlying the

conceptualization of the dual-ambient energy harvesting hydrogel. The hydrogel was co-designed with the versatility to harness both ambient solar and thermal energy in the forms of sunlight and naturally driven evaporation, for the purpose of hydrogen and electricity generation. For the effective implementation of a photocatalytic hydrogel, one important consideration is the mass flow of reactants (water and sacrificial reagents) and products (hydrogen) within the hydrogel. Accordingly, vertically aligned internal channels have been intentionally created to provide the transport pathway necessary for the quick delivery of reactants to the photocatalytic active sites, while simultaneously allowing the prompt release of gaseous products. Furthermore, through the rational selection of catalytic active material, photo-active Cu-doped ZnS has been utilized, as elemental doping can realize photochemical enhancements such as improved charge separation and enhanced light absorption that would be beneficial for boosting photocatalytic performance.

Interestingly, these structural features also create a duality that allows the harnessing of electricity from evaporation driven by the ambient environment. Here, the same vertically aligned internal channels can be used to promote fluid flow in the hydrogel, induced by the evaporation from the top surface and the rapid capillary wicking of the channels that draws liquid from the bottom reservoir upwards, akin to the natural process of transpiration. This fluid flow interacts with the selected inorganic catalyst layer which possesses a high surface charge, resulting in the accumulation of charges along the flow direction through the streaming potential phenomenon [3,37]. Thereafter, through capitalizing on the generated ion concentration difference, the streaming potential can be exploited for electricity generation.

Ultimately, the hydrogel has been judiciously co-designed as a dualfunctional material, capable of both solar-induced hydrogen generation through photocatalysis and evaporation-induced electricity generation via the streaming potential. With this in mind, the sample preparation method used is imperative since it would determine the formation of the hydrogel's internal structure. Consequently, a directional freeze casting technique was deliberately chosen to synthesize the Cu-ZnS nanosheets incorporated chitosan mixed hydrogel (Fig. 2a). By unidirectionally freezing a dispersion of Cu-ZnS and chitosan in an anisotropic manner along an artificially created temperature gradient, uniaxially aligned ice crystals templates are formed with the Cu-ZnS and chitosan distributed in the spaces between the ice crystals [38,39]. Ice crystals are



Fig. 1. Schematic illustration showing the design and functionality of the Cu-ZnS hydrogel.



Fig. 2. (a) Schematic illustration for the synthesis of Cu-ZnS hydrogel. (b) Digital photograph of Cu-ZnS hydrogel. SEM images of Cu-ZnS hydrogel from the (c,d) topdown and (e,f) cross-section views. (g) SEM image and (h,i) TEM images of Cu-ZnS nanosheets.

subsequently sublimated by freeze drying to obtain a porous structure with a dominant alignment perpendicular to the freezing plane.

Scanning electron microscope (SEM) imaging was used to examine the structural morphology of the 3D organic–inorganic hierarchical photocatalytic hydrogel (Fig. 2b). The planar top-down view shows the presence of multiple open pores on the surface of the hydrogel (Fig. 2c, d), with a deliberately broken portion revealing the vertical channel walls of these pores (Fig. 2c encircled). This orientation is further evident in the vertical cross-section imaging, illustrating well oriented walls along the axial direction, forming directionally aligned channels (Fig. 2e,f). More importantly, the channel walls are decorated with leaflike Cu-ZnS nanosheets, well dispersed along the cross-linked chitosan matrix (Fig. 2f). The long chain chitosan polymer serves to provide the skeletal framework necessary for catalyst immobilization and formation of the hydrogel structure (Fig. S1)

The individual freestanding Cu-ZnS nanosheets have a well-defined leaf-like morphology that are thin and porous in nature (Fig. 2g,h), which is beneficial for catalytic reactions due to the increased effective surface area for facilitating interactions between the catalyst and reactants. Using a MOF mediated strategy, these Cu-ZnS nanosheets were synthesized and doped for subsequent incorporation into the hydrogel. Zeolitic imidazolate framework (ZIF-L) was first synthesized with a portion (5 %) of Zn ions replaced with Cu ions. The resultant MOF has a consistent morphology and crystal phase that matches well to that of the original Zn ZIF-L (Fig. S2). This is possibly due to the ionic radius of Cu²⁺ (71 pm) being similar to Zn²⁺ (74 pm), allowing its substitution into the framework [40], and providing a route towards incorporating foreign Cu atoms into the subsequent MOF-derived sulfide, Cu-ZnS.

Resultantly, both undoped ZnS and Cu-ZnS share similar morphological features (Fig. 2g,h and Fig. S3a,b), with distinct lattice fringes of 0.31 nm observable under the high-resolution transmission electron microscope (HRTEM) (Fig. 2i, S3c), corresponding to the (111) facet of sphalerite ZnS [41,42]. However, in doped Cu-ZnS, the elemental presence of Cu, together with Zn and S can be clearly identified by TEM-Energy-dispersive X-ray (TEM-EDX) mapping (Fig. 3a) with the elements evenly distributed throughout the nanosheet. Additionally, the spatial distribution of Cu was further studied through higher magnification EDX elemental mapping using a high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM), and reveals the even distribution of Cu even at nanoscale ranges (Fig. S4). This suggests that there is no aggregation of Cu species into small clusters or nanoparticles, but rather are doped uniformly throughout the nanostructure.

To gain insights into the crystal phases of the synthesized samples, powder X-ray diffraction (XRD) analysis was examined. As shown in Fig. 3b, the prepared ZnS consists of peaks that can be well indexed to the (1 1 1), (2 2 0) and (3 1 1) planes of sphalerite ZnS (PDF #05–0566) [41,42]. The diffraction pattern remains consistent as Zn ions were gradually replaced with Cu ions ranging from 1 % to 20 %. Even at increased Cu amounts of 20 %, no additional peaks were observed, thereby excluding the existence of CuS or Cu₂S crystallites. However, it could be noted that with the increase of Cu amount, the samples showed reduced crystallinity along with a slight shift of diffraction peaks to higher angles (Fig. 3b,c). Owing to the Cu cation being smaller compared to that of Zn, the substitution of Cu into the ZnS lattice results in a decreased lattice parameter and thus causes the observed positive peak shift [43].

Subsequently, X-ray photoemission spectroscopy (XPS) analysis was also conducted to understand the chemical state of the sample. The scan results of Cu-ZnS further confirmed the presence of Cu, along with Zn and S. High-resolution scan in the Zn 2p region (Fig. 3d) reveals a doublet peak at 1022.4 and 1045.6 eV that can be attributed to Zn $2p_{3/2}$



Fig. 3. (a) EDX mapping of Cu-ZnS. (Scale bar: 1 μm) (b) XRD spectrum of ZnS and Cu-ZnS at varied % Cu concentrations and (c) enlarged portion of region marked in (b). XPS high resolution scans of (d) Zn 2p (e) S 2p and (f) Cu 2p of ZnS and Cu-ZnS. (g) UV–vis absorption spectrum and (h) hydrogen generation rates of Cu-ZnS at varied % Cu concentration. (i) Hydrogen generation cycling of Cu-ZnS.

and Zn $2p_{1/2}$ peaks of Zn^{2+} , respectively [44,45]. Similarly, the high-resolution spectra of S 2p (Fig. 3e) exhibited two characteristic peaks at 161.7 and 163.0 eV that can be attributed to S $2p_{3/2}$ and S $2p_{1/2}$ peaks of divalent S^{2-} , respectively [45,46]. Furthermore, the Cu 2p high-resolution spectra (Fig. 3f) showed a doublet peak that can be assigned to the bivalent Cu^{2+} species [47]. In comparison with undoped ZnS, it can be observed that the Zn²⁺ doublet peaks of Cu-ZnS experienced a shift of approximately 0.5 eV towards higher binding energies, and this can be attributed to a doping induced shift in chemical potential, resulting from the higher electronegativity of the Cu dopant relative to Zn, thus corroborating the introduction of Cu dopants into ZnS [48, 49].

Given that the incorporated photoactive material is the principal constituent that governs the catalytic ability of the hydrogel, necessitates its well-selection, optimization and understanding. Material doping in photocatalyst is known to effectuate several benefits that include, among others, enhanced light absorption, improved charge separation, increased catalytic sites, and altered surface properties [50–53]. Accordingly, the deliberate introduction of Cu dopants into ZnS, was specifically used to effect improvements in its hydrogen generation performance. The resultant optical properties of ZnS doped with varying concentrations of Cu can be observed through UV-Vis absorption spectroscopy (Fig. 3g). Pristine ZnS shows an absorption primarily in the UV-range, while an observed red shift in the absorption edge could be noted upon increasing copper dopant concentration from 0 % to 20 %. The observed red shift signifies that Cu doping into ZnS improves optical properties by extending the light absorption range. Additionally, the absence of secondary absorption peaks even with substantially increased dopant concentration, further substantiates the lack of CuS crystallite formation. Subsequently, the H₂ generation performance of ZnS and Cu-ZnS synthesized with varied levels of Cu doping was investigated. As shown in Fig. 3h, pristine undoped ZnS had a H₂ production rate of 2.11 mmol g⁻¹ h⁻¹. Interestingly, a mild doping of Cu at 1 % results in a substantial increase of the H₂ generation rate, with the optimum doping percentage found at 5 % corresponding to a production rate of 21.95 mmol g⁻¹ h⁻¹, an enhancement of more than 10 times over pristine ZnS. Additionally, repeated testing of the optimized Cu-ZnS sample also displayed excellent stability, maintaining a steady production rate even after extended testing for multiple cycles (Fig. 3i). These results demonstrate the effectiveness of Cu doping as a strategy for improving H₂ evolution performance.

To elucidate the role Cu dopants plays during photocatalysis, the photoluminescence (PL) spectra of Cu-ZnS and ZnS were measured. Since PL emission results from the recombination of photogenerated electron-hole pairs, lower PL intensities suggest reduced recombination and thereby higher charge separation efficiencies [54]. In comparison to pristine ZnS, the PL emission of Cu-ZnS has effectively been quenched following Cu doping (Fig. 4a), signifying effective charge transport occurring between ZnS and Cu. This can be ascribed to the presence of additional Cu states that aid in significantly reducing charge recombination in ZnS [55]. Correspondingly, this leads to an increased number of electrons available for participation in photocatalytic reactions, which has also been observed in the transient on-off photocurrent response when comparing Cu-ZnS and ZnS (Fig. 4b). Compared to pristine ZnS, the photocurrent response of Cu-ZnS shows a higher increase during the light-on state that can be attributed to the efficient

electron-hole pairs generation and separation. Moreover, characterization through electrochemical impedance spectroscopy (EIS) analysed for the hydrogen evolution reaction (HER) also corroborates these findings. As shown in Fig. 4c, Cu-ZnS has a smaller semi-circle diameter as compared to ZnS on its Nyquist plots, which is indicative of a more facile charge transfer interaction at the electrode-electrolyte interface due to the higher separation efficiency of photogenerated electron-hole pairs [56,57].

To further investigate the dopant-induced changes between Cu-ZnS and undoped ZnS, Kelvin Probe Force Microscopy (KPFM) fitted with a UV light source was used to measure light-induced variations in surface potential [58]. The topography image of Cu-ZnS surface and corresponding surface potential mappings in both dark and illuminated conditions is shown in Fig. 4d. Under UV-light irradiation, Cu-ZnS shows a substantial increase in surface potential of approximately 40 mV compared to its dark conditions (Fig. 4e, S5). This increase in surface potential can be attributed to the photogeneration of electron-hole pairs by Cu-ZnS. Considering the samples were dispersed on a grounded Pt-coated substrate during the KPFM measurements, photogenerated electrons are removed through grounding and the accumulation of holes causes the measured positive shift in surface potential. On the other hand, the surface potential of pristine undoped ZnS measured in both dark and under light illumination shows only a relatively minor change (Fig. S6), suggesting a reduced amount of photogenerated charges. Taken together, these results further corroborate the notion that the inclusion of Cu dopants can increase the number of available photogenerated electrons, and can be attributed to the improved light



Fig. 4. (a) Photoluminescence spectrum, (b) photocurrent and (c) EIS measurements comparing ZnS and Cu-ZnS. (d) Topography and surface potential of Cu-ZnS in dark and under light irradiation. (e) Histogram of surface potential of Cu-ZnS in dark and under light irradiation. (f) UV–vis absorption spectrum of chitosan hydrogel and Cu-ZnS hydrogel. (g) Hydrogen generation rates of chitosan hydrogel, Cu-ZnS hydrogel and Cu-ZnS suspension. (h) Hydrogen generation cycling of Cu-ZnS hydrogel with aligned channels and random pores.

absorption and increased charge-transfer capabilities of Cu-ZnS.

Although photocatalytic processes are commonly conducted in suspension form, suspension-based systems possess certain disadvantages, such as light shielding by the uppermost suspension layer, light scattering, sedimentation over time and the difficulty in collection and separation for reuse [13,14]. With this in mind, the Cu-ZnS nanosheets are incorporated into a chitosan matrix to form an immobilized-type photocatalytically active hydrogel following the design principles as outlined in Fig. 1. Light absorption properties were thereafter measured through UV-Vis absorption spectroscopy (Fig. 4f). Firstly, it could be noted that the pristine chitosan hydrogel has low photo-absorption. However, such poor photo-absorption is beneficial for its role as a matrix material, since it reduces the possibility of potential light shielding effects that could compromise the effectiveness of the photocatalyst material. Hence, following the addition of Cu-ZnS the photo-absorption properties of the hydrogel were significantly improved.

To maximize the light exposure of the hydrogel, hydrogen generation performances were tested by positioning it horizontally with only the bottom surface of the hydrogel touching the solution (Fig. 4g). Expectedly, the chitosan hydrogel showed negligible hydrogen generation performance, due to the lack of photocatalyst. In contrast, after Cu-ZnS incorporation, the Cu-ZnS hydrogel showed an elevated hydrogen generation rate of 95.7 mmol $m^{-2} h^{-1}$, which is also substantially higher when compared to a suspension system of freestanding Cu-ZnS with an equivalent catalyst mass (Fig. 4g). Furthermore, the hydrogen production rate of Cu-ZnS hydrogel is also able to remain relatively constant after repeated cycling, affirming its stability for hydrogen generation (Fig. 4h). The results suggest that Cu-ZnS can be assembled into a photoactive hydrogel without compromising its light absorption properties, hydrogen generation performance and stability. At the same time, the immobilization of the catalyst in the gel matrix, imparts benefits to prevent its aggregation and dissolution. In comparison, hydrogel synthesized without directional freeze casting, resulting in a random pore structure (Fig. S7), shows similar initial hydrogen generation rates since the hydrogels were fully wetted before measurement (Fig. 4h). However, upon repeated cycling, the performance of the hydrogel with random pore structure falls rapidly in subsequent cycles (Fig. 4h). This can be attributed to the effectiveness of the constructed directionally aligned channels for fluid transport, which has also been demonstrated through water wicking experiments. With aligned pores, rapid and complete wetting of the top surface of the hydrogel can happen within the first 30 s (Fig. S8). In contrast, the hydrogel with random pore structure (Fig. S7) fails to wick water efficiently, leaving its top surface

dry even after 3 mins (Fig. S8). The rapid wetting of the Cu-ZnS hydrogel can be credited to the provision of the internal aligned channels, resulting in lower tortuosity and stronger capillary action [59], which is necessary for facilitating mass flow of reactants to the active sites in the hydrogel.

Interestingly, the structural features of the hydrogel combined with an analogous experimental setup could readily support electricity generation induced by its natural environment, which can be facilely harnessed from the potential difference between upper and underside of the hydrogel. When the channel wall carrying a surface charge comes into contact with a liquid, an electrical double layer (EDL) consisting of a bonded layer of counter-ions and a diffused layer of mobile free ions forms at the surface-liquid interface inside the channels. As the hydrogel surface undergoes evaporation, a continuous fluid flow occurs within its internal channels, propelled by rapid capillary wicking that draws the fluid from the bottom reservoir upwards towards the surface. This movement of the liquid causes the mobile free ions in the EDL to deviate from its equilibrium position, moving in the fluid flow direction. The displacement gives rise to an electrokinetic effect that results in charge accumulation regions (Fig. 5a). The resulting ion concentration difference causes an electrical potential difference built up that is known as the streaming potential and can be exploited for electricity generation [3.60.61].

In view of this, KPFM was used to investigate the prospective application of Cu-ZnS hydrogel for electricity generation. The surface topology and potential of the Cu-ZnS hydrogel was first measured (Fig. 5b,c). Subsequently, through comparing the dry and wet states of the hydrogels, the potential is noted to have fallen by 6.49 V, suggesting a large change in surface charge accumulation occurs in its wetted state (Fig. 5d,e). To further elucidate this charge difference, the timedependent variation in surface potential over a fixed area of the hydrogel was sampled under the influence of a fluid flow (Fig. 5f). Upon the initiation of the fluid flow across the gel, an initial drop in potential was recorded with the median value falling by 1.58 V after 75 s and thereafter stabilizing at approximately -6.60 V. This can be interpreted as an indication of a depletion of cations at the monitored region, caused by the preferential flow of cations away from this region under the influence of externally driven fluid flow. Resultantly, the depletion of cations causes the measured negative difference in surface potential.

Having established the feasibility of Cu-ZnS hydrogel to generate an internal ion concentration difference, electrically conductive carbon fiber electrodes were connected to the top and bottom surfaces of the hydrogel to harness the potential difference. As shown in Fig. 6a, an



Fig. 5. (a) Schematic illustration of streaming potential phenomenal in Cu-ZnS hydrogel. (b) Topography and (c) surface potential of Cu-ZnS hydrogel. Histogram of the surface potential of Cu-ZnS hydrogel in (d) dry and (e) wet states. (f) Time-dependent surface potential variation over a fixed region of Cu-ZnS hydrogel in response to fluid flow.



Fig. 6. (a) Open-circuit potential and (b) short-circuit current density of Cu-ZnS hydrogel. (c) Potential, current density, and power density as a function of the external resistance. (d) Open-circuit potential response of Cu-ZnS hydrogel to humidity variation. (e) Zeta potential of Cu-ZnS and chitosan. (f) Outdoor testing of Cu-ZnS hydrogel for hydrogen and electricity generation in natural ambient environment.

average open circuit potential of 185.4 mV was obtained, with a continuous short circuit current density of approximately 20.7 mA m⁻² (Fig. 6b). The power output of Cu-ZnS hydrogel was also tested by connecting a variable resistor through the external circuit. It was noted that Cu-ZnS hydrogel generates a maximum power density of 1.82 mW m⁻² at an external resistance of 0.3 M Ω (Fig. 6c). Furthermore, the output potential shows little attenuation even after extended testing which is a testament to the stability of the hydrogel for electricity generation (Fig. 6a).

To ascertain the origin of this measured potential difference, controlled experiments were carried out by changing the measurement environment of the hydrogel. When both the top and bottom of the hydrogel were exposed to air, the potential difference measured was negligible (Fig. S9a). Additionally, if the hydrogel is completely submerged in electrolyte, only a marginal potential of approximately 4.15 mV was generated (Fig. S9b). These findings suggest the contributions from electrochemical reaction sources, such as galvanic or electrochemical corrosion processes, are likely to be insignificant. Whereas the absence of an evaporative driving force in both scenarios could be the factor for the observed poor performance. Nevertheless, as further validation of the evaporation driven mechanism, changes in the open-circuit potential was recorded over repeated cycling of elevated humidity (Fig. 6d). A humidity rise of approximately 10 %RH is shown to cause a steady decline in the measured potential until the end of the cycle, at which point a decrease in humidity is accompanied by a corresponding steady rise in potential. This inverse relation between the measured potential and change in humidity was consistent across multiple repeated cycling periods. Since increasing humidity slows down the evaporation rate, it in turn reduces the internal flow rate within the gel and ultimately leads to a lower potential difference. In light of these findings, it is evident that evaporation is the driving force behind the induced potential.

Apart from the external driving factors, the material and structural features of the hydrogel towards electricity generation were also investigated. As a control, Cu-ZnS hydrogels with randomly aligned pores were synthesized without directional freeze casting (Fig. S7), and its performance of approximately 18.7 mV was significantly lower compared to that of Cu-ZnS hydrogel with directionally aligned pores (Fig. S10). As evidenced by the water wicking experiments (Fig. S8), the lack of distinct aligned channels results in poor capillary wicking, thereby impeding the electrolyte flow in the gel. This obstruction hampers the formation of charge accumulation regions, and resultantly poorer performance. On the other hand, pristine chitosan hydrogels featuring aligned directional channels were also compared (Fig. S1), and only achieved a potential of 48.8 mV (Fig. S11). This lower open circuit potential could be attributed to the difference in surface potential of the channel walls. It was noted that Cu-ZnS has a Zeta potential of $-49.1 \pm$ 6.84 mV which is considerably higher in magnitude compared to chitosan, $-7.24 \pm 3.09 \text{ mV}$ (Fig. 6e). Likewise, the surface potential measured by KPFM shows a similar trend, with a negative shift of 43 mV recorded after a chitosan film was mixed with Cu-ZnS (Fig. S12). Since the generated streaming potential is proportional to the magnitude of the channel wall's surface potential [61,62], it can be effectively augmented by the addition of Cu-ZnS that raises the absolute surface potential. Overall, it can be established that both material and structural features of the Cu-ZnS hydrogel are integral components to enable evaporation-induced electricity generation.

Finally, the feasibility of using the dual-ambient energy harvester hydrogel in outdoor real-world conditions was further assessed through the development of a prototype reactor, constructed to serve as a proofof-concept (Fig. S13). The hydrogen generation performance of the prototype reactor was measured outdoor under natural sunlight. Despite intermittent cloud coverage resulting in an average solar flux of 529.1 Wm⁻², the Cu-ZnS hydrogel showed a steady rise in hydrogen production over time with an overall generation rate of 17.3 mmol m^{-2} h^{-1} (Fig. 6f). Similarly, evaporation driven electricity generation could also be observed in outdoor settings. At an ambient temperature of 27.1 °C and humidity of 88 %RH, the Cu-ZnS hydrogel exhibits stable and continuous electricity generation with an open circuit potential measuring an average output of 192.8 mV (Fig. 6f). Accordingly, Cu-ZnS hydrogel demonstrates it is fully functional in the real-world environment as a versatile dual-ambient energy harvester, capable of harnessing both ambient solar and thermal energy in the forms of sunlight and natural evaporation.

3. Conclusion

In summary, a judiciously co-designed dual-functional hydrogel has been demonstrated for its versatility to harness both ambient sunlight and naturally driven evaporation, for production of hydrogen and electricity generation. The proposed hydrogel design well integrates Cudoped ZnS photocatalysts into a chitosan matrix with inherently formed aligned internal channels that supports the mass flow of reactants and release of gaseous products. Additionally, photochemical enhancement is achieved through Cu doping, which enhances light absorption and improves charge separation. Similarly, the internal aligned channels also provide a path for fluid flow through the hydrogel driven by surface evaporation. Concurrently, coupled with the surface potential of the inorganic Cu-ZnS channel walls allows for evaporatively driven electricity generation through the streaming potential phenomenal. Cumulatively, real-world environmental usage of the dual-ambient energy harvester has been demonstrated via a prototype reactor, which exhibits an overall hydrogen generation rate of 17.3 mmol $m^{-2} h^{-1}$ under natural sunlight irradiation and an average potential output of 192.8 mV. In general, the strategies employed in this study demonstrates the potential in designing other ambient energy harvesters with the ability to convert disparate renewable energy sources for sustainable energy production.

CRediT authorship contribution statement

Kane Jian Hong Lim: Writing – original draft, Methodology, Investigation, Formal analysis, Conceptualization. Wanheng Lu: Investigation, Formal analysis. Serene Wen Ling Ng: Methodology, Investigation. Minmin Gao: Methodology. Ghim Wei Ho: Supervision, Funding acquisition, Conceptualization.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper

Data availability

Data will be made available on request.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.nanoen.2024.109850.

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