



Short communication

Natural wood derived robust carbon sheets with perpendicular channels as gas diffusion layers in air-breathing proton exchange membrane fuel cells (PEMFCs)

Dai Dang^a, Ranjie Zeng^a, Xingwei Chen^a, Xiaohui Su^a, Xu Yang^{a,*}, Huaneng Su^b, Chuande Wu^a, Lei Zhang^c

^a School of Chemical Engineering and Light Industry, Guangdong University of Technology, Guangzhou 510006, PR China

^b Institute for Energy Research, Jiangsu University, Zhenjiang 212013, China

^c Energy, Mining & Environment, National Research Council, Canada



ARTICLE INFO

Keywords:

Air-breathing
PEMFC
Natural wood
Perpendicular channels
Gas diffusion layer

ABSTRACT

Herein, a novel natural wood derived macroporous carbon sheet with three-dimension inter-connected perpendicular-channels was engineered as gas diffusion layers (w-GDLs) for air-breathing proton exchange membrane fuel cells (PEMFCs). Beneficial to the unique accessible perpendicular channels and the presence of a microporous layer, the current density reached 0.139 A/cm² (at 0.6 V) and the maximum power density elevated up to 0.102 W/cm² (at 0.43 V), which are comparable to the best results reported for the air-breathing PEMFCs with high Pt loadings. Furthermore, it exhibited excellent durability during preliminary constant discharge operation, demonstrating the feasibility of this w-GDL for practical applications.

1. Introduction

Currently, the air-breathing proton exchange membrane fuel cells (PEMFCs), which could effectively conduct the gas convection and diffusion through the open cathodes without the help of auxiliary machinery, have gained great attention. As the miniaturized products of fuel cells, the air-breathing PEMFCs feature flexibility, lightweight, and easy operation, which are expected to be competitors of lithium-ion batteries [1–7].

Although air-breathing PEMFCs share the same reaction mechanism as conventional PEMFCs, there are two key issues involved with gas diffusion layers (GDLs) that should be solved. Firstly, the air-breathing PEMFCs are conducted at the open cathodes, in which the reactant oxygen is provided through the GDLs via air convection and diffusion. This inevitably results in the concentration polarization and voltage drop due to the sluggish kinetics of passive gas delivery [8]. Secondly, appropriate water management is critical for the GDLs to obtain stable cell performance. Specifically, there exists a delicate water transport balance at the reactive three-phase boundary of catalyst, electrolyte and reactant gas in PEMFC operation: water is necessary for the hydration of proton-conducting membrane, while excessive water can block the pores of

the electrodes and result in flooding, causing a serious problem on the passive air diffusion [9]. Therefore, the improvement of water transportation and gas diffusion is important at GDLs to ensure the cell performance and stability of air-breathing PEMFCs.

Conventionally, the GDLs are constructed by carbon fiber paper or carbon cloth with a hydrophobic polymer binders such as PTFE for mass transfer and water management. However, the microstructure of such GDLs is mainly dominated by disordered pores, which cannot provide continuous channels for gas, electron and water molecules transport (Fig. 1). With the continuous operation of the cell, the water generated by the cathode will accumulate in the pores of the GDL, hindering the mass transfer, and therefore harm the performance and stability of the cell. Although the last decade has witnessed great progress in tuning the structure of the GDL to achieve high performance of air-breathing PEMFC [8–12], there are still some vital issues left to be solved for future practical applications. Thus, exploring novel structures of GDLs with 3D vertically aligned microchannels that have better mass transfer and water management is a meaningful line of inquire.

Natural wood is one of the most abundant biomass on earth. The multi-layered pore structure inside the wood guarantees the normal transport and distribution of air, moisture and nutrients in the tree.

* Corresponding author.

E-mail address: yangxu@gdut.edu.cn (X. Yang).

<https://doi.org/10.1016/j.catcom.2021.106351>

Received 4 June 2021; Received in revised form 19 August 2021; Accepted 20 August 2021

Available online 22 August 2021

1566-7367/© 2021 The Authors.

Published by Elsevier B.V. This is an open access article under the CC BY-NC-ND license

(<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

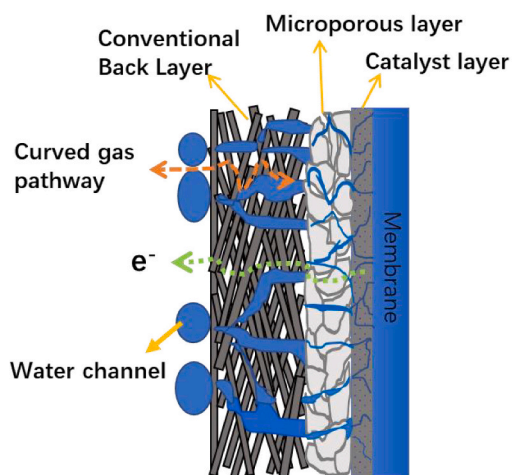


Fig. 1. Schematic diagram of conventional GDL microstructure.

Among them, “transport” is mainly generated by parallel channels along the growth direction, resulting in a vertical material flow. Between these parallel channels, there are still some finer holes that allow the two to exchange substance with each other, thereby forming a “distribution” result at the same level. This material can be carbonized especially to retain most of the original channels, demonstrating the potential to be used as gas diffusion electrodes in energy storage and energy conversion devices [10–25]. The natural wood material used in this work is composed of large numbers of macropores and its pore size distribution across the 3D carbon framework is relatively uniform. The pore size of w-GDL is slightly larger than that of the conventional GDL carbon materials (such as carbon paper), which can better meet the working requirements in air-breathing PEMFCs. The ordered, interconnected and low-tortuosity microchannels in w-GDL will shorten the gas diffusion pathway and improve air exchange efficiency. Meanwhile, the water management would be simplified since water can easily drain away from the aligned tubes, which can avoid flooding at the cathode and enlarge the three-phase boundary among catalysts, oxygen, and electrolyte. It should be mentioned that the well-aligned microchannels will enable

charge transport within the 3D matrix. After screening a large number of possibilities, we found that the natural wood-derived 3D carbon sheets might be good candidates as GDL materials in air-breathing PEMFCs.

Herein, the natural wood that cut perpendicularly to the growth direction is carefully carbonized to retain its inherent 3D macroporous perpendicular channels with a thickness of $\sim 300\ \mu\text{m}$. The commonly used polymer PTFE in conventional GDL is also selected in this work as a binder and hydrophobic additive. By adjusting the proportion of PTFE to the treated natural wood, the diffusion of gas and water can be properly balanced. The hydrophobicity of w-GDL has been studied and some interesting results are obtained. In a single cell test, the air-breathing PEMFC using the optimal w-GDL sample exhibits the maximum current density of $318\ \text{mA cm}^{-2}$ and power density of $102\ \text{mW cm}^{-2}$ at $0.43\ \text{V}$, which is superior to the conventional GDL (c-GDL) ($280\ \text{mA cm}^{-2}$, $88\ \text{mW cm}^{-2}$ at $0.43\ \text{V}$). Furthermore, the wood derived carbon sheets can be served as self-supporting electrode for hydrogen evolution reaction (HER) because it would not only facilitate the diffusion of generated hydrogen bubbles through the 3D connected perpendicular channels but it would also provide a large number of active sites for electrolyte ions, which would lead to the improvement of electrochemical reaction rate of HER. This novel material takes advantage of its unique characteristics, resulting in a promising candidate for the next generation of state-of-the-art low-cost electrode.

2. Experimental

2.1. Synthesis of 3D-connected carbon sheet with perpendicular channels

The 3D-connected macroporous wood derived carbon sheets (w-Carbon) were prepared according to the method reported [20]. Natural wood of pine trees was cut perpendicular to the wood growth direction to obtain wood pieces. The obtained wood pieces were then carbonized in a tubular furnace at $1000\ ^\circ\text{C}$ for 2 h in a nitrogen gas atmosphere. After carbonization, the w-Carbon pieces were carefully trimmed to form the final carbon sheets with a size of $1.0\ \text{cm} \times 1.0\ \text{cm} \times 300\ \mu\text{m}$ (see Fig. S1 in Supporting Information).

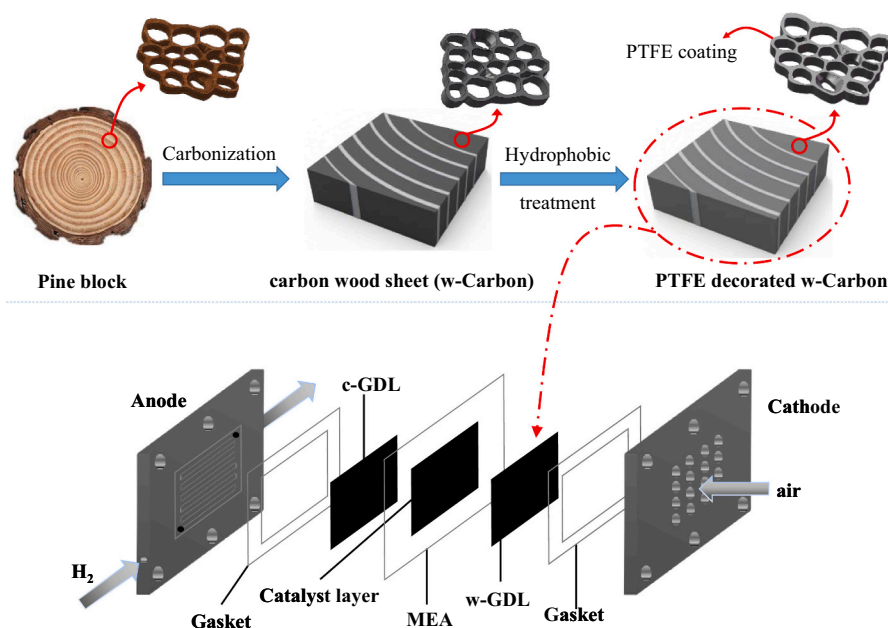


Fig. 2. Schematic diagram for the preparation route of carbon sheet (w-Carbon) with 3D connected perpendicular channels (upper), and configuration of air-breathing PEMFCs (lower).

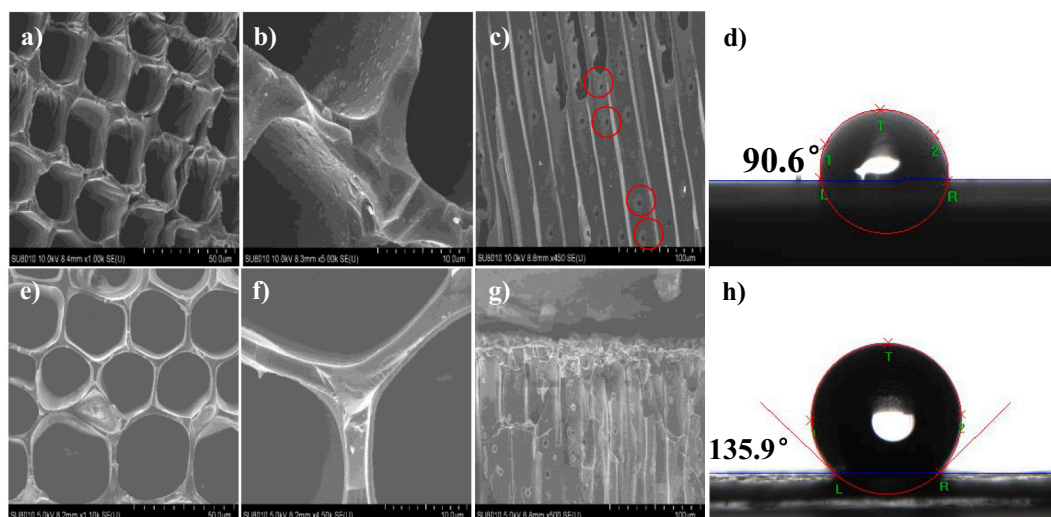


Fig. 3. SEM images and contact angles for the pristine w-Carbon (a–d) and PTFE loaded w-Carbon (e–h).

2.2. Structure characterization

The microscopic morphologies and structure were characterized by a field-emission SEM (Zeiss, Oberkochen, Germany). The hydrophilic properties of the w-GDL were evaluated using the contact angle method on an OCA 40 Video-Based Automatic Contact Angle Meter (DataPhysics, Germany). Raman measurements were carried out using a Raman spectroscope (HORIBA Jobin Yvon LabRAM HR, France) to discern the carbon species.

2.3. Electrochemical characterization

Electrochemical impedance spectroscopy (EIS) was employed to study the polarization resistance of the MEAs by an Ivium electrochemical workstation (Ivium, Netherlands).

2.4. Gas diffusion layer (GDL) preparation

Before the single cell assembly, the GDL was engineered using the w-Carbon in advance. Firstly, the obtained w-Carbon sheets were hydrophobically treated with Teflon dispersion (60 wt%, Dupont), followed by a heat-treatment at 400 °C for 30 min to uniformly disperse the PTFE on the carbon sheets. The final loading of PTFE was in the range of 15–30 wt%. For reference, the c-Carbon paper (TCP-H-060, Toray, Japan) with a PTFE loading (20 wt%) was processed through the same procedure to construct the c-GDL.

2.5. Air-breathing PEMFCs assembly

The assembly of air-breathing PEMFCs is shown in Fig. 2. In the anode side, a flow-field plate with serpentine layouts was employed to introduce the dry hydrogen into the c-GDL made from commercial carbon paper (TCP-H-060, Toray, Japan). For the cathode side, the plate with open windows matrix accessed to the w-GDL leads to direct diffusion of ambient air. Between the anode and cathode GDLs, the catalyst-coated membranes (CCMs) with an active area of 2.25 cm² were prepared using a direct catalyst spraying technique developed in our laboratory [26]. The catalyst (Pt/C, 40 wt% JM4100, Johnson Matthey) loadings at the anode and cathode were 0.1 and 0.2 mg_{Pt} cm⁻², respectively. The Nafion solution (5 wt%) was used as a binder, and the Nafion amount in both the anode and cathode catalyst layers was adjusted to 25 wt%. The membrane type is Nafion® 212 (Dupont, 50 μm).

2.6. PEMFC performance test

The PEMFC performance was measured in a standard 2.25 cm² single cell using a Fuel Cell Testing System (Arbin Instruments, USA). Pure hydrogen without any humidification was fed into the anode at a flow rate of 100 mL min⁻¹, while the natural circulating air was diffused into the cathode in the manner of “air-breathing”. Before each test, the cell was activated in a chronoamperometry mode at 0.8 V until a stable performance was obtained.

3. Results and discussion

3.1. Structure characterization

The morphology of the pristine w-Carbon was observed by SEM, as shown in Fig. 3a–c. The regular macropores array in the size of 20–30 μm are seen distinctly in the w-Carbon, and certain smaller pores of 2–5 μm (indicated by a red circle) are distributed in line on the channel wall, suggesting the 3D connected porosity (Fig. 3a–c). The contact angle of 90.6° indicates its hydrophilic surface of the natural derived w-Carbon (Fig. 3d). After PTFE modification, most parts of macroporous channels remain accessible and unimpeded (Fig. 3e–f); from the magnified region, the previously smoothed inner surface becomes relatively rough, verifying the evenly dispersed PTFE aggregation. The corresponding contact angle of 135.9° suggests its hydrophobic properties, because of the PTFE decoration (Fig. 3h). In contrast, the commercial carbon paper displays randomly and horizontally stacked brittle carbon fibers (with 10–15 μm diameter) bounded into a web-like matrix, and several small inter-fiber pores seem susceptible to be stuck by PTFE aggregation (Fig. S2, Supporting Information), which may cause an obstacle for the water/gas delivery. These SEM observations demonstrate that the structure of w-Carbon may possess better permeability than the commercial carbon, which would influence significantly the mass transfer of water and air diffusion in the cathode.

The I_D/I_G value in the Raman spectra can be used to characterize the graphitization degree of various carbon materials, and the smaller I_D/I_G value generally corresponds to the higher degree of graphitization [28,29]. As shown in Fig. S3, typical D (~1320 cm⁻¹) and G (~1580 cm⁻¹) bands were observed for all three samples, i.e., w-GDL (20 wt% PTFE), w-GDL without PTFE, and c-GDL (20 wt% PTFE), which indicates that the formation of graphitic carbon was obtained after pyrolysis for all samples. And the calculated I_G/I_D values of all the carbon materials show an increasing order: w-GDL > c-GDL (20 wt% PTFE) > w-GDL (20 wt% PTFE), implying a relatively high graphitized degree in w-GDL (20 wt%

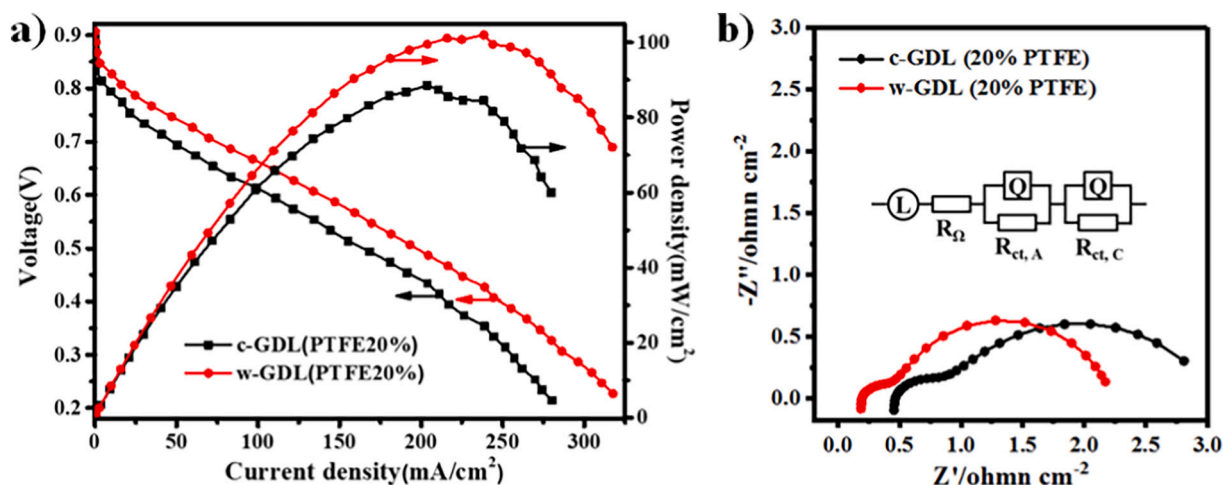


Fig. 4. Polarization and power density curves (a), and *in situ* impedance curves (b) for the air-breathing PEMFC assembled by different GDLs.

Table 1

Structure and electrochemical properties of the air-breathing PEMFCs using different GDLs.

GDLs	Max. Current density (A/cm ²)	Peak power density (W/cm ²)	R_{Ω} (Ω/cm ²)	$R_{ct, A}$ (Ω/cm ²)	$R_{ct, C}$ (Ω/cm ²)
c-GDL	0.280	0.089	0.4484	0.4019	2.235
w-GDL	0.318	0.102	0.1857	0.276	1.767

% PTFE), which could pave a highway for transferring electron along the 3D connected perpendicular-channels to the active sites in the carbon framework.

3.2. Single cell test

We first conducted the cell performance for the PEMFC assembled by w-GDL with various PTFE loadings (Fig. S4). We found that by increasing the PTFE loading (< 20 wt%) can significantly improve the cell performance, while excessive PTFE loading (> 20 wt%) can deteriorate the cell performance. This could be ascribed to the fact that at proper PTFE loading, the hydrophobic treated w-GDL can effectively eliminate the water produced from the cathode, while excessive PTFE could result in clogging access paths and led to poor mass transfer. Thus, we fixed the PTFE loading at 20 wt% for the w-GDL if not noted elsewhere.

Fig. 4 compares the cell performance for the PEMFC assembled by w-GDL and c-GDL. In low current density region (<0.1 A/cm²), the negligible discrepancy between these two samples is observed, owing to the dominant charge transfer-controlled region [30]. However, this difference becomes prominent at the high current density region due to the limitation of mass transport, suggesting the advantageous structure feature of w-GDL on the mass transfer. Specifically, w-GDL shows a higher maximum current density of 0.318 mA/cm², which is 10% higher than that of the c-GDL does. The peak power density reaches 102 mW/cm² at 0.43 V, comparable to the best ones reported for air-breathing PEMFCs with higher Pt loadings [9,31]. This excellent performance can be attributed to the merits of the 3D connected perpendicular channels of w-GDL during mass transfer: *i*) the excessive produced water can be effectively discharged through these macroporous channels by vaporization [32]; *ii*) the oxygen-containing air can be diffused unimpededly to the catalyst layer.

The resistances of these two different GDL-derived PEMFCs were analyzed in the single cell fixture at 0.8 V by *in situ* impedance measurements, as shown in Fig. 4b. The cell ohmic resistances (R_{Ω}), anode and cathode charge transfer resistances ($R_{ct, A}$ and $R_{ct, C}$) were obtained by fitting the data with a Randels model equivalent circuit, and values

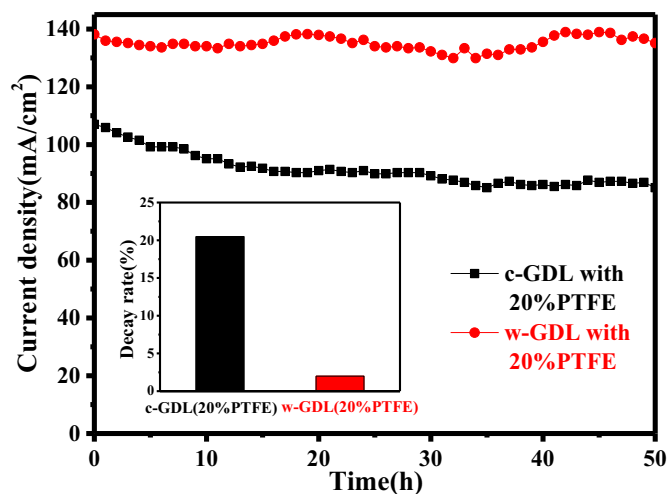


Fig. 5. Long-term durability test for PEMFC assembled by w-GDL.

are listed in Table 1. Explicitly, the PEMFCs assembled by the w-GDL display lower charge transfer resistance compared to that of c-GDL, implying that the w-Carbon structure could create abundant reaction sites, leading to a more efficient electrochemical process in the electrodes. Especially, the much low ohmic resistance is obtained in w-GDL compared with the c-GDL, indicating that the monolithic carbon skeleton can effectively decrease the contact resistance, which is consistent with the polar curve analysis mentioned above.

The long-term operation stability is an important criterion for the PEMFC operating system in practical applications. Generally, the flooding issue at the cathode plays a key role in air-breathing PEMFC performance degradation. Appropriate water management in the GDL is needed to balance the water flooding and self-humidification. To verify the stability of the single cell, a primary durability test (after activation process) was performed by continuously operating the resultant PEMFCs assembled by various w-GDLs and c-GDLs at 0.6 V, as shown in Fig. 5.

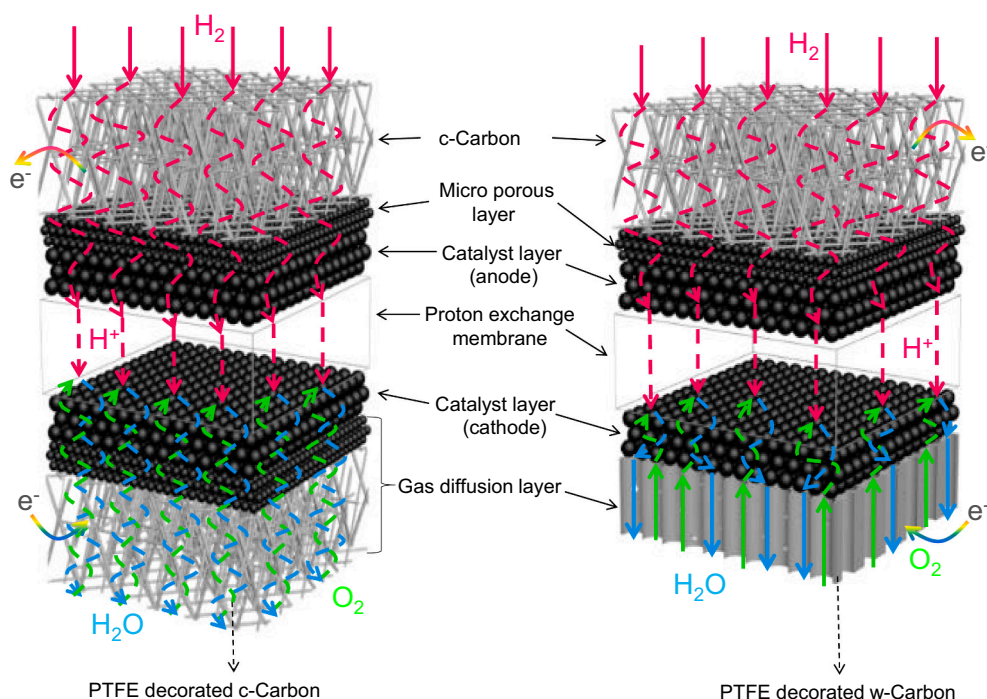


Fig. 6. Schematic diagram for the diffusion kinetics in the PEMFC assembled with c-GDL and w-GDL.

Distinctively, using the w-GDL, the PEMFC shows excellent stability with only 2.5% current density drop after over 50 h operation at high current density around 140 mA/cm². In contrast, the conventional PEMFC using the c-GDL displays 20% current density drop even during the initial 10 h run.

To better illustrate the advantageous structure feature of the w-GDL performed in the air-breathing PEMFC, the schematic diagram for the diffusion kinetic in the assembled cell is presented in Fig. 6. Compared to the conventional carbon paper (c-carbon), this novel wood derived 3D inter-connected macroporous carbon holds several merits that are beneficial to mass transfer, charge transfer and even heat release. The well-distributed macropore array can diminish the clogging induced by the PTFE decoration, facilitating the mass transfer, while maintaining the good hydrophobicity of GDL. The perpendicular channels in the GDL could decrease the transport route distance, which would significantly improve the mass transfer, charge transfer as well as heat release. The w-GDL consists of an entire carbon piece cut from wood, which can avoid the aggregation issues of carbon fibers in conventional carbon paper. This is important for maintaining the reactive active sites during electrochemical reaction at the cathode.

4. Conclusions

Through carefully designed carbonization of the natural wood, we have successfully prepared novel carbon sheets (w-Carbon) with 3D inter-connected perpendicular macroporous channels. With an optimized PTFE loading (20 wt%), the hydrophobic w-Carbon served as cathode GDL to assemble air-breathing PEMFCs. Maximum current density (0.318 A/cm²) and power density (0.102 W/cm²) were achieved in a single cell testing, outperforming those observed in the PEMFCs using commercial carbon paper as GDL (c-GDL). Moreover, after 50 h of long-period operation, the current density declined by merely 2.5%. The improved performance is strongly associated with the unique merits the w-GDL has, which includes the perpendicular accessible channel, the presence of microporosity, and appropriate hydrophobic properties. Owing to these advanced attributions, improved mass transfer and charge transfer, as well as a good balance between GDL hydration and

water drainage are achieved, both responsible for the improved PEMFCs performance. The GDL prepared by this method has the advantages of low cost, well-kept ordered structure and low mass transfer resistance, which provide a new direction for the carbon materials selection and design and improvement of air-breathing PEMFCs.

Declaration of Competing Interest

None.

Acknowledgments

We gratefully acknowledge the financial support from the National Natural Science Foundation of China (21905055), the PhD Start-up Fund of Natural Science Foundation of Guangdong Province (No. 2018A030310514), Educational Commission of Guangdong Province (No. 2017KQNCX064), the start-up funding of Guangdong University of Technology (220413207 and 220418129), and support from Research Fund Program of Key Laboratory of Fuel Cell Technology of Guangdong Province.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.catcom.2021.106351>.

References

- [1] N. Mirrashid, S.M. Rakhtala, M. Ghanbari, Robust control design for air breathing proton exchange membrane fuel cell system via variable gain second-order sliding mode, *Energy Sci. Eng.* 6 (2018) 126–143.
- [2] N. Ul Hassan, B. Tunaboylu, M. Yasin, Development and technological steps for realization of AB-PEM Fuel cell - a step towards clean & sustainable energy solutions, in: 2018 4th International Conference on Power Generation Systems and Renewable Energy Technologies (Pgsret-2018), 2018, pp. 104–109.
- [3] A. Baroutaji, J.G. Carton, J. Stokes, A.G. Olabi, Application of open pore cellular foam for air breathing PEM fuel cell, *Int. J. Hydrogen Energy.* 42 (2017) 25630–25638.

- [4] M.H. Cano, M.I.A. Mousli, S. Kelouwani, K. Agbossou, M. Hammoudi, Y. Dube, Improving a free air breathing proton exchange membrane fuel cell through the maximum efficiency point tracking method, *J. Power Sources* 345 (2017) 264–274.
- [5] S.P. Isanaka, T.E. Sparks, F.F. Liou, J.W. Newkirk, Design strategy for reducing manufacturing and assembly complexity of air-breathing proton exchange membrane fuel cells (PEMFC), *J. Manuf. Syst.* 38 (2016) 165–171.
- [6] L. Wang, X.L. Tian, Y. Xu, S. Zaman, K. Qi, H. Liu, B.Y. Xia, Engineering one-dimensional and hierarchical PtFe alloy assemblies towards durable methanol electrooxidation, *J. Mater. Chem. A* 7 (2019) 13090–13095.
- [7] J. Luo, H. Tang, X. Tian, S. Hou, X. Li, L. Du, S. Liao, Highly selective TiN-supported highly dispersed Pt catalyst: ultra active toward hydrogen oxidation and inactive toward oxygen reduction, *ACS Appl. Mater. Interfaces* 10 (2018) 3530–3537.
- [8] W. Jang, J. Choi, Y. Seo, B. Kim, Effect of cathode flow field configuration on air-breathing proton exchange membrane fuel cell, *Int. J. Precis. Eng. Manuf.* 16 (2015) 1129–1134.
- [9] Z.A. Xiang, S.J. Liao, D. Dang, X.L. Tian, S.Y. Hou, F.F. Liu, H.L. Peng, Z.Y. Fu, Enhanced water management in the cathode of an air-breathing PEMFC using a dual catalyst layer and optimizing the gas diffusion and microporous layers, *Int. J. Hydrogen Energ.* 40 (2015) 3961–3967.
- [10] Y.D. Kuang, C.J. Chen, G. Chen, Y. Pei, G. Pastel, C. Jia, J.W. Song, R.Y. Mi, B. Yang, S. Das, L.B. Hu, Bioinspired solar-heated carbon absorbent for efficient cleanup of highly viscous crude oil, *Adv. Funct. Mater.* 29 (2019).
- [11] Y.D. Kuang, C.J. Chen, S.M. He, E.M. Hitz, Y.L. Wang, W.T. Gan, R.Y. Mi, L.B. Hu, A high-performance self-regenerating solar evaporator for continuous water desalination, *Adv. Mater.* 31 (2019).
- [12] Z.Y. Yang, M. Chen, M. Xia, M. Wang, X.D. Wang, An effective and durable interface structure design for oxygen reduction and methanol oxidation electrocatalyst, *Appl. Surf. Sci.* 487 (2019) 655–663.
- [13] M.W. Zhu, Y.J. Li, F.J. Chen, X.Y. Zhu, J.Q. Dai, Y.F. Li, Z. Yang, X.J. Yan, J. W. Song, Y.B. Wang, E. Hitz, W. Luo, M.H. Lu, B. Yang, L.B. Hu, Plasmonic wood for high-efficiency solar steam generation, *Adv. Energy Mater.* 8 (2018).
- [14] T. Li, H. Liu, X.P. Zhao, G. Chen, J.Q. Dai, G. Pastel, C. Jia, C.J. Chen, E. Hitz, D. Siddhartha, R.G. Yang, L.B. Hu, Scalable and highly efficient mesoporous wood-based solar steam generation device: localized heat, rapid water transport, *Adv. Funct. Mater.* 28 (2018).
- [15] M.W. Zhu, Y.J. Li, G. Chen, F. Jiang, Z. Yang, X.G. Luo, Y.B. Wang, S.D. Lacey, J. Q. Dai, C.W. Wang, C. Jia, J.Y. Wan, Y.G. Yao, A. Gong, B. Yang, Z.F. Yu, S. Das, L. B. Hu, Tree-inspired design for high-efficiency water extraction, *Adv. Mater.* 29 (2017).
- [16] C. Jia, Y.J. Li, Z. Yang, G. Chen, Y.G. Yao, F. Jiang, Y.D. Kuang, G. Pastel, H. Xie, B. Yang, S. Das, L.B. Hu, Rich mesostructures derived from natural woods for solar steam generation, *Joule* 1 (2017) 588–599.
- [17] C.J. Chen, S.M. Xu, Y.D. Kuang, W.T. Gan, J.W. Song, G.G. Chen, G. Pastel, B. Y. Liu, Y.J. Li, H. Huang, L.B. Hu, Nature-inspired tri-pathway design enabling high-performance flexible Li-O₂ batteries, *Adv. Energy Mater.* 9 (2019).
- [18] S.M. Xu, C.J. Chen, Y.D. Kuang, J.W. Song, W.T. Gan, B.Y. Liu, E.M. Hitz, J. W. Connell, Y. Lin, L.B. Hu, Flexible lithium-CO₂ battery with ultrahigh capacity and stable cycling, *Energy Environ. Sci.* 11 (2018) 3231–3237.
- [19] H.Y. Song, S.M. Xu, Y.J. Li, J.Q. Dai, A. Gong, M.W. Zhu, C.L. Zhu, C.J. Chen, Y. A. Chen, Y.G. Yao, B.Y. Liu, J.W. Song, G. Pastel, L.B. Hu, Hierarchically porous, ultrathick, "breathable" wood-derived cathode for lithium-oxygen batteries, *Adv. Energy Mater.* 8 (2018).
- [20] Y.J. Li, T.T. Gao, Y.G. Yao, Z.Y. Liu, Y.D. Kuang, C.J. Chen, J.W. Song, S.M. Xu, E. M. Hitz, B.Y. Liu, R.J. Jacob, M.R. Zachariah, G.F. Wang, L.B. Hu, In situ "chainmail catalyst" assembly in low-tortuosity, hierarchical carbon frameworks for efficient and stable hydrogen generation, *Adv. Energy Mater.* 8 (2018).
- [21] Y. Zhang, W. Luo, C.W. Wang, Y.J. Li, C.J. Chen, J.W. Song, J.Q. Dai, E.M. Hitz, S. M. Xu, C.P. Yang, Y.B. Wang, L.B. Hu, High-capacity, low-tortuosity, and channel-guided lithium metal anode, *P. Natl. Acad. Sci. USA* 114 (2017) 3584–3589.
- [22] Y.J. Li, K. Fu, C.J. Chen, W. Luo, T.T. Gao, S.M. Xu, J.Q. Dai, G. Pastel, Y.B. Wang, B.Y. Liu, J.W. Song, Y.N. Chen, C.P. Yang, L.B. Hu, Enabling high-areal-capacity lithium-sulfur batteries: designing anisotropic and low-tortuosity porous architectures, *ACS Nano* 11 (2017) 4801–4807.
- [23] C.J. Chen, Y. Zhang, Y.J. Li, Y.D. Kuang, J.W. Song, W. Luo, Y.B. Wang, Y.G. Yao, G. Pastel, J. Xie, L.B. Hu, Highly conductive, lightweight, low-tortuosity carbon frameworks as ultrathick 3D current collectors, *Adv. Energy Mater.* 7 (2017).
- [24] C.J. Chen, Y. Zhang, Y.J. Li, J.Q. Dai, J.W. Song, Y.G. Yao, Y.H. Gong, I. Kierzewski, J. Xie, L.B. Hu, All-wood, low tortuosity, aqueous, biodegradable supercapacitors with ultra-high capacitance, *Energy Environ. Sci.* 10 (2017) 538–545.
- [25] F. Shen, W. Luo, J.Q. Dai, Y.G. Yao, M.W. Zhu, E. Hitz, Y.F. Tang, Y.F. Chen, V. L. Sprenkle, X.L. Li, L.B. Hu, Ultra-thick, low-tortuosity, and mesoporous wood carbon anode for high-performance sodium-ion batteries, *Adv. Energy Mater.* 6 (2016).
- [26] D. Dang, L. Zhang, X. Zeng, X. Tian, C. Qu, H. Nan, T. Shu, S. Hou, L. Yang, J. Zeng, S. Liao, In situ construction of Ir@Pt/C nanoparticles in the cathode layer of membrane electrode assemblies with ultra-low Pt loading and high Pt exposure, *J. Power Sources* 355 (2017) 83–89.
- [27] Z. Wei, R. Pan, Y. Hou, Y. Yang, Y. Liu, Graphene-supported Pd catalyst for highly selective hydrogenation of resorcinol to 1, 3-cyclohexanedione through giant pi-conjugate interactions, *Sci. Rep.* 5 (2015) 15664.
- [28] S.-G. Kim, O.-K. Park, J.H. Lee, B.-C. Ku, Layer-by-layer assembled graphene oxide films and barrier properties of thermally reduced graphene oxide membranes, *Carbon Lett.* 14 (2013) 247–250.
- [29] H. Su, Q. Xu, J. Chong, H. Li, C. Sita, S. Pasupathi, Eliminating micro-porous layer from gas diffusion electrode for use in high temperature polymer electrolyte membrane fuel cell, *J. Power Sources* 341 (2017) 302–308.
- [30] Z.A. Xiong, S.J. Liao, S.Y. Hou, H.B. Zou, D. Dang, X.L. Tian, H.X. Nan, T. Shu, L. Du, Construction of a high-performance air-breathing cathode using platinum catalyst supported by carbon black and carbon nanotubes, *Int. J. Hydrogen Energ.* 41 (2016) 9191–9196.
- [31] O.A. Obeisun, D.P. Finegan, E. Engebretsen, J.B. Robinson, O.O. Taiwo, G. Hinds, P.R. Shearing, D.J.L. Brett, Ex-situ characterisation of water droplet dynamics on the surface of a fuel cell gas diffusion layer through wettability analysis and thermal characterisation, *Int. J. Hydrogen Energ.* 42 (2017) 4404–4414.