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Experimental study on the effect of temperature and water content on the thermal conductivity of gas diffusion layers in proton exchange membrane fuel cell

Lei Chen*, Yi-Fan Wang, Wen-Quan Tao

Key Laboratory of Thermo-Fluid Science and Engineering, Ministry of Education, School of Energy & Power Engineering, Xi'an Jiaotong University, Xi'an, Shanxi 710049, People's Republic of China

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ABSTRACT

The gas diffusion layer is one of the key components of proton exchange membrane fuel cell. Temperature and water content of gas diffusion layers have a significant impact on its thermal conductivity. The thermal conductivity of the dry gas diffusion layer shows an upward trend with increasing temperature, but the change is small. The thermal conductivity of the gas diffusion layer shows a linear increase with the increase of water content. When the water content of the gas diffusion layer is 77%, its thermal conductivity increases with the temperature. Its thermal conductivity increased from 0.3592 W/(m·K) at 25 °C to 0.7871 W/(m·K) at 85 °C.

1. Introduction

The Gas Diffusion Layer [1] (GDL) is one of the key components of proton exchange membrane fuel cell. It is located between the catalytic layer and the bipolar plate. It is usually composed of porous conductive fiber materials, such as carbon paper and carbon cloth (Figs. 1 and 2). The fiber diameter is on the order of 10 μm. The thickness of the gas diffusion layer is generally 10–400 μm. The gas diffusion layer has three important functions in the fuel cell: conductive gas, discharge reaction products, collecting current. Firstly, the gas diffusion layer makes the reaction gas (hydrogen or oxygen) reach the catalytic layer uniformly from the flow channel. However, for some gas diffusion layers, because the pores of carbon paper or carbon cloth are relatively large, such as carbon paper Toray TGP-H-060, which accounts for about 90% of the large pores with a pore diameter greater than 20 μm, so a microporous layer (MPL) must be added between the catalytic layer and the carbon paper[2–4]. was added in the meantime. The gas diffusion layer has a double-layer structure: the base layer and the microporous layer. The carbon paper or carbon cloth layer is called a substrate layer (SL). Secondly, the gas diffusion layer is responsible for discharging the water generated by the electrochemical reaction of the cathode catalytic layer into the flow channel [4]. In order to prevent the reaction gas from being blocked (due to the transfer of liquid water in the gas diffusion layer), the carbon paper or carbon cloth used for the gas diffusion layer is generally hydrophobized [5] to build a hydrophobic gas channel. Thirdly, the gas diffusion layer is responsible for providing the electrical contact between the electrode and the external bipolar plate to collect the generated current. The factors affecting the performance of the gas diffusion layer are mainly the type of carbon black, the material of the base layer, the preparation process and the content of the component [6]. Jordan [7] and Passalacqua et al. [8] believe that a larger pore volume is conducive to gas transmission, so it is more conducive to battery output performance. However, Neergat et al. [9] thought that the output performance of the battery produced by MPL with high specific surface Ketjen Black was better.

Because there are both solids and liquids in the gas diffusion layer, and the holes formed by the fiber interleaving are random, the heat transfer process of GDL is very complicated, including heat conduction, convective heat transfer and radiative heat transfer. When studying the heat transfer of non-uniform porous GDL, thermal conductivity is one of the most important material properties. Zhou et al. [10] and Omrani et al. [11] found that gas diffusion layer and MPL thermal conductivity are the most critical parameters to improve fuel cell performance. At present, only a few literatures have described the experimental determination of GDL thermal conductivity. Vie et al. [12] calculated the thermal conductivity of the gas diffusion layer and membrane based on the measured internal temperature distribution of the single fuel cell. Teertstra et al. [13] measured the thermal conductivity of GDL using a steady state measurement method and studied the effect of PTFE content on its thermal conductivity: the higher the PTFE content, the lower the thermal conductivity. Ramousse et al. [14], Khandelwal et al. [15], Nitta et al. [16], Karimi et al. [17], also measured the thermal

* Corresponding author.
E-mail address: chenlei@mail.xjtu.edu.cn (L. Chen).

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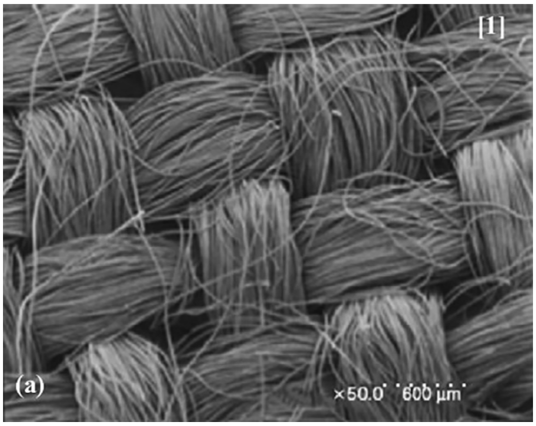

Fig. 1. SEM scan of carbon cloth [4].

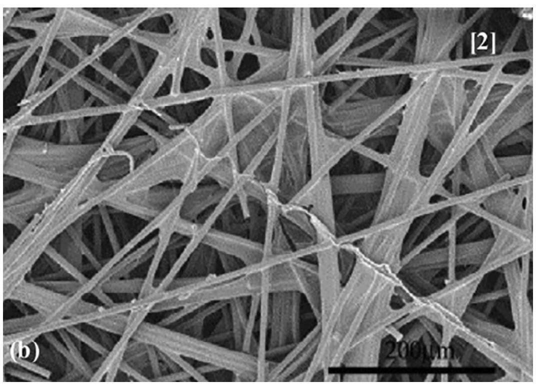

Fig. 2. SEM scan of carbon paper [4].

conductivity of the gas diffusion layer using the same method. Some of these studies specifically considered the effect of pressure on GDL thickness. Chen et al. [18] studied study the effect of PTFE content and external load on the thermal conductivity of GDL materials by sensing technology. Table 1 is the thermal conductivity results of GDL and PEM reported in the literature [17], which shows that the test results are very inconsistent. At the same time, it can be found that these tests rarely study the influence of temperature and water content on the thermal conductivity. Therefore, this paper mainly focuses on the effects of temperature and water content on the thermal conductivity of gas diffusion layers produced in China.

2. Experimental principle and sample preparation

The experimental test system includes a Hotdisk thermal conductivity meter, a computer terminal installed with supporting test software, and a test fixture device, shown in Fig. 3. Its basic principle is the transient planar heat source method, which is a technology developed on basis of the hot wire method and has become an ISO standard for thermal conductivity measurement [19]. A thin layer disc-shaped resistance wire was used as the sample probe. The probe is a thin sheet of double-helix structure formed by etching of metallic nickel. The double helix structure is covered with polyimide or mica insulation protection layer. During the test, the probe is placed between two

Fig. 3. Experimental test system.

identical samples to be tested, and the sandwich structure is formed by close contact. The test sample should be of sufficient thickness and diameter to ensure that the temperature rise of the probe is not transmitted to the boundary during the test, so that the semi-infinite medium assumption is satisfied. The effect of thickness on its thermal coefficient determination is very important. GDL is sandwiched between two steel plates of known thickness, so that the thickness of the entire sandwich structure under a fixed pressure can be read directly through a digital micrometer. Since the steel plates on both sides hardly deform when subjected to a small pressure, its thickness can be measured more accurately. When measuring these values, multiple measurements are used to average the values to obtain a more accurate GDL thickness.

The samples used in this experiment were purchased from Institute of New Energy Wuhan Company. The size is 7cm×7cm. The material object is shown in Fig. 4a, and its microstructure is shown in Fig. 4b.

Since the working temperature of a proton exchange membrane fuel cell is 25–90 °C and the working pressure is atmospheric pressure, the temperature range measured in this paper is 25–90 °C, and the pressure is standard atmospheric pressure. After the experimental equipment system was turned on, the system was stable and the test was started. We still need to wait an hour for the next test to stabilize the system. The sample measurement under the same working condition is generally performed more than three times. When the characteristic temperature is less than 10^{-4} K, the measurement is considered accurate. The input parameters are: the thermal conductivity of the probe at the corresponding temperature, the number of tests, and the thickness at standard atmospheric pressure. The thermal conductivity can be calculated by Hotdisk computer software.

3. Results and analysis

Fig. 5 shows the thermal conductivity of the dry gas diffusion layer filled with air in the pores as a function of temperature. It can be seen from the figure that as the temperature increases, the thermal conductivity of the gas diffusion layer shows an upward trend, but the change is not large.

Because the proton exchange membrane in a proton exchange membrane fuel cell must work under a certain water content, a dry gas diffusion layer does not actually exist in a proton exchange membrane fuel cell. Therefore, water must be contained in the pores of the gas diffusion layer. So the thermal conductivity of the gas diffusion layer under a certain water content is measured in this paper. Fig. 6 shows

Table 1
Thermal conductivity of GDL reported in the literature.

Author	Maggio et al.	Wohr et al.	Gurau et al.	Argyropoulos et al.	Toray Indusries Inc.	Nguyen et al.	Hwang et al.
Thermal conductivity of GDL $W{\cdot}m^{-1}{\cdot}K^{-1}$	15.56	65	21.5	0.15	1.6	1.3	1.7

Fig. 4. a) GDL entity picture, b) SEM scan of GDL.

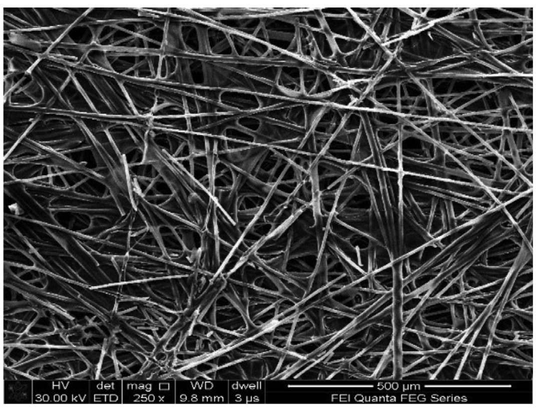

Fig. 4. (*continued*)

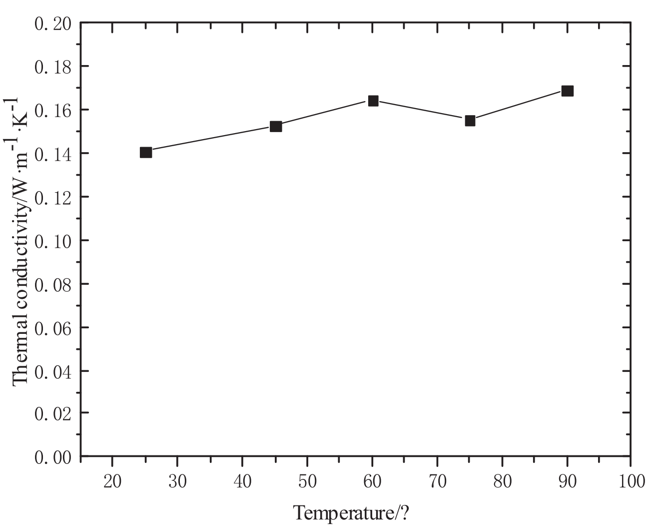

Fig. 5. Thermal conductivity curve of GDL at different temperatures.

the relationship between the thermal conductivity of the gas diffusion layer and its water content. Obviously, the thermal conductivity increases linearly with the increase of the water content. The relationship equation of the thermal conductivity of the gas diffusion layer with the water content can be obtained by the least squares fitting:

$$\lambda_{GDL} = 0.029h_{water} + 0.1454$$

In fact, the temperature of a proton exchange membrane fuel cell changes from startup to normal operation. Therefore, this paper also measured the thermal conductivity of the gas diffusion layer at 77% water content, shown in Fig. 7.

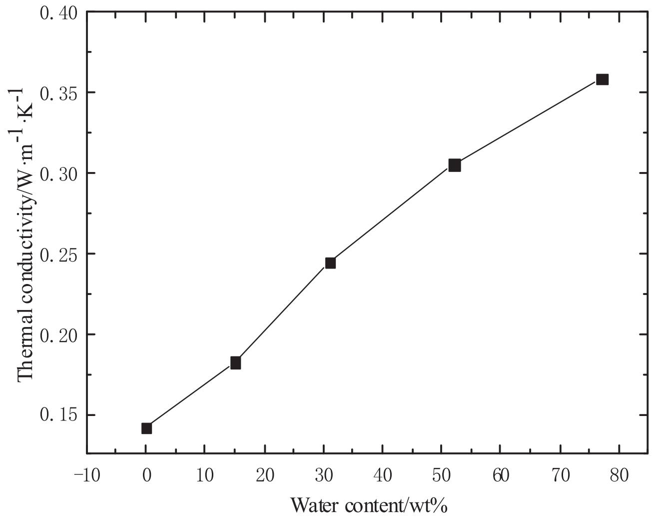

Fig. 6. Thermal conductivity curve of GDL at different water contents.

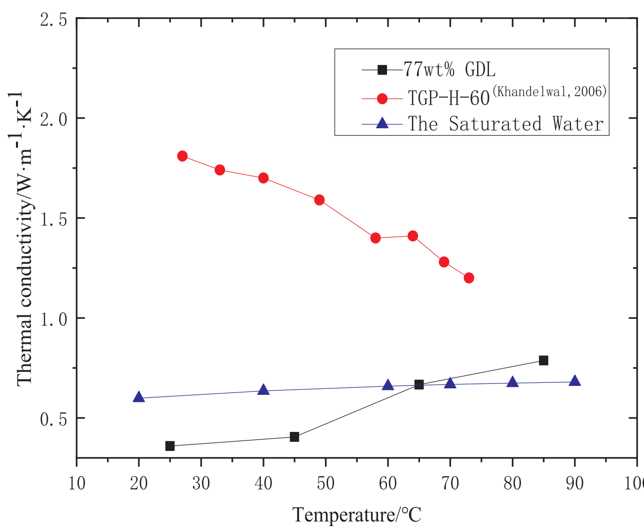

Fig. 7. Thermal conductivity curve of GDL with 77% water content at different temperatures.

It can be seen from Fig. 7 that the thermal conductivity increases with temperature, from 0.3592 W/(m·K) at 25 °C to 0.7871 W/(m·K) at 85 °C. The figure also shows the thermal conductivity of Toray carbon paper (TGP-H-060) measured by Khandelwal et al. [15]: it decreased from 1.80 W/(m·K) at 26 °C to 1.24 W/(m·K) at 73 °C. Comparing with the experimental results measured by Khandelwal et al., the two trends are completely different. The main reasons are as follows:

1) The structure of the test materials is different. Khandelwal et al. [15] measured TGP-H-060, and the measurement material in this chapter was GMPL, which was further processed based on Toray TGP-H-060 carbon paper.
2) Water content is different. Khandelwal et al. did not mention water content in the text. If it contains water, the thermal conductivity of water increases rapidly with increasing temperature, so the thermal conductivity of the gas diffusion layer should not decrease so quickly. It is concluded that it may be the measurement result in the dry state. Even in the dry state, it is inconsistent with the result that the thermal conductivity of the dry GMPL is basically unchanged. In this paper, GMPL with a water content of 77 wt% was measured, and its thermal conductivity increased with increasing temperature. This result is reasonable.

3) Different test methods are used. Khandelwal et al. used a steady-state thermal conductivity test method. The influence of thermal resistance needs to be considered, which will inevitably bring certain errors. This article adopts the transient method without considering the contact thermal resistance. It's more accurate.

4. Conclusion

In this paper, the thermal conductivity of the gas diffusion layer was measured using a Hotdisk thermal conductivity meter, and the effects of water content and temperature on its thermal conductivity were studied. The thermal conductivity of the dry gas diffusion layer shows an upward trend with increasing temperature, but it does not change much. The thermal conductivity of the gas diffusion layer under a certain water content shows a linear increase with the increase of the water content, and the relationship equation between the thermal conductivity and the water content is obtained by the least square method fitting. The temperature of a proton exchange membrane fuel cell changes from startup to normal operation. Therefore, when the water content of the gas diffusion layer was 77%, the change of the thermal conductivity of the gas diffusion layer under the influence of temperature was also studied. The thermal conductivity showed an upward trend with increasing temperature. It increases from 0.3592 W/(m·K) at 25 °C to 0.7871 W/(m·K) at 85 °C.

CRediT authorship contribution statement

Lei Chen: Conceptualization, Methodology, Formal analysis. **Yi-Fan Wang:** Writing - review & editing. **Wen-Quan Tao:** Writing - review & editing.

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.

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