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Measurement of thermal conductivity and heat pipe effect in hydrophilic and hydrophobic carbon papers

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ABSTRACT

In this paper, we present an experimental study on measurement of the thermal conductivity and heat pipe effect in both hydrophilic and hydrophobic (Toray TGP-H60) carbon papers (around 200 μm thickness) with/out liquid water. An experimental setup is developed for measuring thermal conductance at different liquid water contents and temperatures without dissembling the testing device for water addition. Theoretical analysis is also performed to evaluate the apparent conductance of heat pipe effect. We found that liquid water presence inside these materials increases the overall thermal conductivity. At high temperature around 80 °C, the heat pipe effect is evident for the hydrophilic paper; while for the hydrophobic one, the heat pipe effect is found to be smaller. The distinction is likely due to the different patterns of the capillary liquid flow in the two media. For the hydrophobic paper, liquid water flows back to the evaporation side when the breakthrough pressure is reached and flow is through preferred routes of small flow resistance. As a result, heat pipe effect is active only in part of the medium, therefore smaller than that in the hydrophilic one. The results are important for understanding the heat transfer phenomena occurring in porous media and effects of material surface property.

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1. Introduction

Carbon papers are porous materials that are widely applied in a variety of engineering applications, such as batteries, heat transfer devices, fuel cells, multi-stage filters, high-temperature thermal insulators, and friction/wear applications. Carbon papers are nonwoven fibrous media, based on carbon fibers, and are commercially available. Carbon fibers possess excellent properties of stiffness, strength, conductivity, and light weight, and can be made from polyacrylonitrile (PAN), heavy fractions of oils or coals, cellulose, or others. In fabrication, the raw carbon fibers are carbonized at high temperature in inert environment to reduce other elements (such as nitrogen, oxygen, and hydrogen), yielding fibers with high carbon content and strong mechanical strength. In the papermaking process, the chopped carbon fibers are dispersed in water with binders, such as polyvinyl alcohol, to produce carbon paper roll. Binder content in carbon papers usually ranges from 5% to 15% by weight. The paper's porosity can be 80% or higher, e.g. Toray carbon papers. This type of carbon papers has a mean pore size of about 20 µm. Their pore network provides passages for fluid flow or species transport, while the solid matrix enables electric current conductance or heat transfer [1,2].

Carbon paper's surface property plays an important role in two-phase flow occurring inside the medium. This property can be modified through adding Polytetrafluoroethylene (PTFE), nanostructures, or other chemical agents. Applying PTFE loading is widely adopted in PEM fuel cell development. In this method, carbon papers are dipped into aqueous PTFE suspensions or by spraying. The wet papers are then placed in an oven for drying to remove the residual solvent. High temperature above 300 °C will sinter PTFE and fix it to the fiber surface. Higher PTFE content yields a more hydrophobic property. Adding PTFE, however, reduces the media's porosity and possibly the mean pore size.

Thermal conductivity is an important property of carbon papers, particularly in thermal and power applications. Though carbon fibers are highly conductive, the effective conductivity can be low when the porosity is high. The effective conductivity is also dependent on compression, PTFE loading, solid matrix's tortuosity, and the fluid materials in the void space. Various experimental methods have been employed to measure thermal conductivity [3], and can be extended to carbon papers. Khandelwal and Mench [4] measured carbon papers and Nafion membranes for PEM fuel cell. They reported a thermal conductivity of $0.22 \pm 0.04 \, \text{W/m} \,^{\circ}\text{C}$ for Sigracet® 20 wt.% PTFE carbon paper and $1.80 \pm 0.27 \, \text{W/m} \,^{\circ}\text{C}$ for Toray papers. Zamel et al. [5] measured the through-plane thermal conductivity of dry Toray papers in a range of 50– $120 \,^{\circ}\text{C}$. They reported about 0.8– $1.8 \, \text{W/m} \,^{\circ}\text{C}$ conductivity at high deformation and 0.2– $0.4 \, \text{W/m} \,^{\circ}\text{C}$ at low deformation. Burheim et al. [6]

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Nomenclature C concentration (mol/m³) density (kg/m³) ρ D diffusivity (m²/s) mobility λ the latent heat (I/kg) h tortuosity k thermal conductivity (W/m °C) Κ permeability (m²) Subscript or superscript Μ molecular weight (kg/mol) capillary С р pressure (pa) eff effective S saturation fg phase change и velocity (m/s) gas hр heat pipe liquid Greek symbols surface tension (N/m) sat saturation porosity w water 3 θ contact angle (deg) reference point viscosity (kg/m-s) μ

measured SolviCore porous media at various compaction pressures. For dry media under 4.6, 9.3 and 13.9 bar, the measured conductivity was 0.27, 0.36 and 0.40 W/m $^{\circ}$ C, respectively. They reported an increase of around 0.17 W/m $^{\circ}$ C in thermal conductivity when about 25% liquid saturation is present inside the media. Burheim et al. [7] reported the through-plane thermal conductivities of the several widely used carbon porous transport layers (PTLs). They found that the thermal conductivity of dry PTLs decreases with increasing PTFE content and increases with residual water.

The heat pipe effect will be promoted in carbon papers when there exists two-phase flow under a nonisothermal condition. The effect results from the transport and phase change of the working fluid flow, e.g. the vapor-phase diffusion, capillary liquid flow, condensation, and evaporation: at the evaporation side, the liquid work fluid evaporates, absorbing heat. The resulting vapor transports towards the condensation side, where the vapor condenses to liquid, releasing heat. The condensed work fluid then returns back to the evaporation side, restarting the cycle. In this process, heat is transported from the evaporation to the condensation sides. Wang and Wang [8,9] evaluated vapor-phase diffusion and heat pipe conductance, indicating that the former is a significant mechanism in the water management of PEM fuel cell and the latter contributes about 0.3-0.5 W/m °C conductivity at 80 °C. Though the carbon paper's conductivity has been measured by several experiments, little has been done experimentally to investigate the heat pipe effect and the role of liquid in the media. In this study, experiment was designed for precise control of liquid injection into a sealed sample, and measurement of heat pipe effect. By comparing the results of dry and wet carbon papers, the added thermal conductance contributed by liquid water presence, including heat pipe effect, was obtained.

2. Heat transfer and heat pipe effect in carbon papers

In carbon papers, the carbon fibers are randomly aligned in the lateral direction. In the presence of static air or hydrogen gas in the void space, a major portion of heat flow is through the solid matrix of fibers because of its high conductivity. The solid matrix's structure therefore plays an important role in heat transfer through the carbon papers.

2.1. Carbon paper microstructure

Carbon papers consist of randomly aligned carbon fibers bound by carbonized thermoset resin, see Fig. 1(a). The fiber diameter is usually around 5–10 um. The paper's fiber structures can be reconstructed through imaging or stochastic modeling. The former uses imaging techniques, such as the X-ray Tomograph or magnetic resonance. The material is repeatedly sectioned and imaged. The images are used in software to reconstruct the detailed material's 3-D structure. The latter method is based on the knowledge of the material's structure and stochastic theory. It is cost effective and fast, and its resolution is usually limited by the reconstruction mesh. Fig. 1(b) displays the reconstructed solid matrix of a carbon paper [10], showing a highly tortuous solid matrix due to the lateral alignment of its carbon fibers. Direct simulation results revealed that the through-plane heat conduction occurs via a route combining the lateral path along fibers and through-plane one at the contact points, see Fig. 2(a), resulting in a value of about 13 for the tortuosity of the solid matrix [10], and a complex distribution of temperature, see Fig. 3. The effective conductivity can be evaluated by accounting for the volume fraction of individual

$$k^{\text{eff}} = k^{s} (1 - \varepsilon)^{\tau_{s}} + k^{f} \varepsilon^{\tau_{f}} \tag{1}$$

where τ is tortuosity. In the occasion that the fibrous matrix is much more conductive than the fluid in the void, the effective conductivity can be approximated by only accounting for the solid content:

$$k^{\text{eff}} = k^{s} (1 - \varepsilon)^{\tau_{s}} \tag{2}$$

Adding PTFE changes the carbon paper's surface properties, and it also alters the medium's porosity, and possibly its thermal conductivity. Fig. 1(c) and (d) display the carbon papers with 0 and 30 wt.% PTFE loadings, respectively.

2.2. Vapor-phase diffusion and heat pipe effect

The pore size of commercial carbon papers such as Toray papers usually ranges from 10 to 100 μm with the mean size of about 20 μm [2]. Due to the small dimension, phase equilibrium generally holds true between vapor and liquid phases in local pores. Under an isothermal condition, this equilibrium ensures a uniform vapor partial pressure throughout the medium. In a non-isothermal environment, i.e. temperature varies spatially, local vapor-partial pressure differs spatially, leading to vapor-phase diffusion. Using a 1-D case as example, the presence of a temperature gradient will cause the following vapor-phase diffusive flux [8]:

$$-D_{g}^{w,eff}\frac{dC^{w}}{dx} = -D_{g}^{w,eff}\frac{dC_{sat}(T)}{dx} = -D_{g}^{w,eff}\frac{dC_{sat}}{dT}\frac{dT}{dx}$$
 (3)

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