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Review article

Liquid water transport characteristics of porous diffusion media in polymer electrolyte membrane fuel cells: A review

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HIGHLIGHTS

- Researches on liquid water transport properties of GDMs in last decade are reviewed.
- A reliable Leverett-type correlation for GDMs is hard to be established for modeling.
- A comprehensive data set of liquid water transport properties for GDMs is needed.

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ABSTRACT

Fundamental understanding of liquid water transport in gas diffusion media (GDM) is important to improve the material and structure design of polymer electrolyte membrane (PEM) fuel cells. Continuum methods of two-phase flow modeling facilitate to give more details of relevant information. The proper empirical correlations of liquid water transport properties, such as capillary characteristics, water relative permeability and effective contact angle, are crucial to two phase flow modeling and cell performance prediction. In this work, researches on these properties in the last decade are reviewed. Various efforts have been devoted to determine the water transport properties for GDMs. However, most of the experimental studies are ex-situ measurements. In-situ measurements for GDMs and extending techniques available to study the catalyst layer and the microporous layer will be further challenges. Using the Leverett–Udell correlation is not recommended for quantitative modeling. The reliable Leverett-type correlation for GDMs, with the inclusion of the cosine of effective contact angle, is desirable but hard to be established for modeling two-phase flow in GDMs. A comprehensive data set of liquid water transport properties is needed for various GDM materials under different PEM fuel cell operating conditions.

1. Introduction

Water management has been a crucial problem for efficient operation of PEM fuel cells for twenty years. The existence of liquid water in gas channel (GC), gas diffusion layer (GDL) and catalyst layer (CL) increases the reactant transport resistance to electrode catalyst surface, causing severe performance loss. It is indispensable for obtaining reliable and high cell performance to keep water balance between membrane hydration and the avoidance of cathode flooding. Some water management strategies were proposed for individual fuel cell or a fuel cell stack, such as novel flow field designs [1–4], porous water transport plates [5–7], microporous layer structures [8–10], and so on. Due to the interaction of electrochemistry, two-phase flow and heat transfer, the complex transport phenomena inside porous GDL and CL is not yet fully understood. In addition, the transport behavior is complicated by the microscale and the anisotropic nature of thin-film GDL.

In the last decade, many efforts have been made to visualize the liquid transport phenomena in PEM fuel cells. Bazylak [11] reviewed novel techniques developed for visualizing the water accumulation in the GC, GDL or membrane of the PEM fuel cell. The liquid water in PEM fuel cell has been visualized by using optical microscopy [12–16], neutron imaging [17–24], X-ray radiography [25–27], and nuclear magnetic resonance (NMR) [28]. These visualization techniques have provided detailed insight in GDMs with regard to the nature of water transport inside PEM fuel cells.

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However, it is still challenging to obtain the detail distributions of water content in the GDL or membrane of an operating PEM fuel cell due to the limitations of the above methods.

In addition, numerous PEM fuel cell models have been developed to attempt to provide an accurate prediction of liquid water distribution in GDMs. For modeling the two-phase flow in GDMs, there are two major approaches based on the continuum method, i.e. two-fluid model [29–38] and multiphase mixture model [39–49]. The two-fluid model is mainly attributed to the work of Nguyen and co-workers [29–33], Djilali and co-workers [34–36], and Li and co-workers [37,38]; and the mixture model was mainly developed by Wang and co-workers [39–46], You and Liu [47,48], and Mazumber and Cole [49]. As indicated in the review [50], compared to the mixture model, only one extra liquid water transport conversation equation is solved in the two-fluid model and non-equilibrium water phase change can be accounted for by setting different source terms in the conservation equations. However, with both of continuum methods, the accurate prediction is dependent on the correct liquid transport properties, like capillary relationship, relative permeability and so on. In recent years, other approaches, such as Lattice Boltzmann (LB) method [51–57], full morphology (FM) models [58–60] and pore-network (PN) models [51–64], have been utilized to investigate liquid water behaviors in GDMs and shed some light on liquid water transport in PEM fuel cells.

Many reviews about PEM fuel cell technology progress and fundamental research [50,65–69] have been published in the recent years. Peighambardoust et al. [65] gave a review of past and present research in the field of development of proton exchange membranes for fuel cells to achieve better performance, higher durability and lower cost. Wang et al. [66] reviewed the latest status of PEM fuel cell technology development and applications and outlined major challenges in fuel cell technology development and the needs for fundamental research for the near future and prior to fuel cell commercialization. Jiao and Li [50] presented a comprehensive review of previous researches related to water transport in PEM fuel cell, including the experimental investigations, modeling works, and researches related to cold start (startup from subzero temperatures) and high temperature PEM fuel cell (operating at the temperatures higher than 100 °C) with less water management needed. Khan et al. [67] reviewed two-phase modeling approaches to study various transport processes and reactions in PEM fuel cells along with some experimental work. Tsushima and Hirai [68] examine the state of art regarding in situ diagnostics, particularly visualization techniques, for probing the behavior of water in PEMFCs, with attention to neutron radiography, X-ray imaging, magnetic resonance imaging, and optical visualization techniques. Zamel and Li [69] gave a comprehensive review of the experimental, numerical and theoretical work dedicated to the estimation of the transport properties in the various components of PEM fuel cells, including gas diffusion layer, microporous layer catalyst layer and the membrane electrolyte layer.

As indicated in most of above reviews [50,66,67], fundamental understanding of liquid-water behaviors in GDMs is required to predict its effect on cell performance. The proper empirical correlations of liquid water transport properties are crucial to two phase flow modeling to obtain the accurate prediction. The present work focus on this aspect and the related researches in the last decade are reviewed.

2. Liquid water transport equation and capillary diffusion coefficient

The liquid water transport mechanisms in GDMs can be explained by capillary force, hydraulic permeation and gravity effect, as well as the water condensation and evaporation. The liquid water transport is evidently influenced by the condensation and evaporation in an operating fuel cell. The water phase change rates are strongly affected by the local conditions such as mass and heat transfer in the cell and many factors, including the pressure, temperature, and humidification of inlet gases as well as current density and cell voltage. A comprehensive investigation of the condensation and evaporation process in GDMs is rather complicated and needs to be performed in the vicinity of the liquid/vapor interface on the molecular level. However, for PEMFC modeling on the macroscopic level, it is impractical to incorporate such processes and a revised form of the Hertz-Knudsen-Langmuir equation [29,30,37,70] was commonly employed to take into account the condensation and evaporation. Other methods can be referred to Caulk and Baker's work [71,72]. They proposed a one-dimensional model for heat and two-phase water transport in hydrophobic GDMs and the analysis correlated well with the experimental results by limiting current methods. Since liquid water transport combined with the water phase change in fuel cell are so complex, the accuracy of the mass transfer rate of phase change calculations on the macroscopic level remains debated [50]. In this review, due to the size limitation of the review, the research progress of the water phase change and the role of the heat transfer on liquid water transport would not be discussed.

In GDMs, liquid water transport is strongly affected by the pore walls due to the small pore sizes, and the wall adhesion effect becomes significant in this scenario, where the capillary effect and surface wettability of GDMs therefore play a significant role in liquid water transport. In general, the liquid water transport in GDMs is mainly driven by the capillary force, while body forces, such as gravity, do not significantly affect its momentum. The capillary action is related with the properties of GDMs, e.g. surface tension and internal contact angle to water. With the help of a two-phase generalized form of Darcy's law and a capillary pressure function, the governing equation for liquid water transport in GDMs is derived from the volume averaged continuity equation, taking the form [29].

$$\nabla\cdot\left(\rho_{\mathrm{l}}\frac{\eta_{\mathrm{g}}}{\eta_{\mathrm{l}}}\frac{K_{\mathrm{rl}}}{K_{\mathrm{rg}}}\mathbf{u}_{\mathrm{g}}\right)=\nabla\cdot(\rho_{\mathrm{l}}D_{\mathrm{c}}\nabla s)+S \tag{1}$$

where η_{g} and η_{l} are the viscosities of gas and liquid phases, respectively; K_{rg} and K_{rl} are the relative permeabilities of gas and liquid phases, respectively; $\mathbf{u}_{\mathrm{g}}$ is gas velocity vector; D_{c} is capillary diffusion coefficient; s denotes liquid water saturation; S represents the water source due to electrochemical reaction, membrane desorption and mass transfer during evaporation and condensation.

2.1. Definition of capillary diffusion coefficient

The capillary diffusion coefficient, which is the most significant parameter in the liquid water transport conservation equation and also called as the liquid water diffusivity, has been derived from the continuity equation and Darcy's law [29], expressed as

$$D_{\mathrm{c}}=\frac{KK_{\mathrm{rl}}}{\eta_{\mathrm{l}}}\frac{dp_{\mathrm{c}}}{ds} \tag{2}$$

where K represents the intrinsic permeability of porous materials; p_{c} denotes capillary pressure, which is related with liquid water saturation s, defined as

$$p_{\mathrm{c}}=p_{\mathrm{l}}-p_{\mathrm{g}}=f(s) \tag{3}$$