



Theoretical design strategies of bipolar membrane fuel cell with enhanced self-humidification behavior



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HIGHLIGHTS

- 2-D model for bipolar membrane (BPM) fuel cell with membrane reaction interface.
- Theoretical design strategies of BPM affecting self-humidification are found.
- Enhanced self-humidification behavior obtained in wider work condition.

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ABSTRACT

The bipolar membrane fuel cells (BPMFCs), which have a unique acid-alkaline jointed membrane electrode assembly (MEA) structure, have demonstrated their great potential for self-humidification during operation. Although the self-humidification ability of such bipolar membranes (BPMs) has recently been validated by a one-dimensional BPM model, the transport mechanism and the formation of self-humidification in the MEAs are not well understood. In the present study, a two-dimensional cross-channel MEA model is developed to elucidate the mechanisms and enhancement of water transport on self-humidification with comprehensive consideration of the three electrochemical reaction zones. The water-formation interface model has been successfully investigated by theoretical and experimental interface reaction kinetics, streamlines of water flux present the formation process and mechanism of self-humidification. A critical current (voltage) value, beyond which self-humidification is initiated, is identified. It is also found that such critical current (voltage) can be adjusted by changing the membrane thickness and the water uptake property of the ionomer. It is concluded that fabricating BPMs with proper membrane thickness and water uptake property are effective strategies to enhance the water management and cell performance in BPMFCs.

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1. Introduction

Bipolar membrane fuel cells (BPMFCs) has attracted much interest in recent years because of their potential function of being self-humidifying [1–9]. The heart of a BPMFC is a novel membrane electrode assembly (MEA), which is composed of an acidic catalyst layer (CL), an alkaline catalyst layer, and a bipolar membrane (BPM) as the separator and electrolyte. A typical structure of BPMFC's MEA

is illustrated in Fig. 1. The BPM itself is composed of a proton exchange membrane (PEM) and an anion exchange membrane (AEM) in which the mobile carriers are protons and hydroxide ions, respectively. Inside the BPM, water is produced at the PEM|AEM interface and diffuses toward each electrode with a sufficient concentration gradient as the driving force; half the stoichiometric quantity of the water produced at this interface is consumed by the cathodic oxygen reduction reaction (ORR). Electro-osmotic drag (EOD) [10], i.e., movement of water induced by moving ions, tends to cause accumulation of water on both sides of the PEM|AEM interface. Once the water build-up at this interface is sufficient to establish a steady water flow towards each electrode, the water

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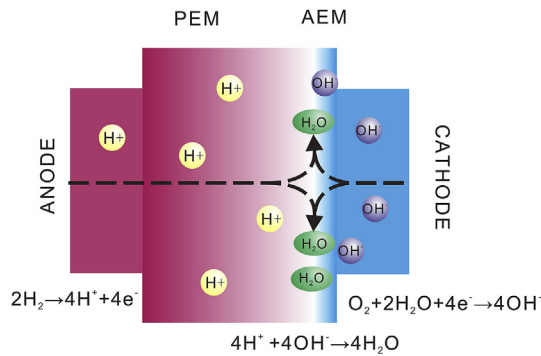


Fig. 1. Typical structure and principle mechanism of a BPMFC.

consumed by the cathode ORR included, self-humidification can be achieved in the BPMFC [5]. Since the conductivity of BPM primarily relies on the membrane water content, uneven water distribution in MEA and membrane dry-out would result in significant increase of ohmic loss. The self-humidification feature of bipolar MEA thus has a potential of simplifying water management that may eventually eliminate the need of external gas humidifiers.

To investigate the self-humidification behavior of bipolar membranes, a series of studies were carried out by a few research groups. A hybrid MEA employing Nafion 212 and a thin layer of AEM (~1.9 μm) was first reported by Ünlü et al. [2]. Their cell produced a higher power in completely dry operation (0% RH in both gas streams) than that at 100% RH. The self-hydrating behavior was attributed to the water produced at the PEM/AEM interface. However, their results contradict with a report by Shen et al. using a hybrid MEA with a BPM that consisted of Nafion membrane and a 35 μm thick AEM [8]. Shen et al. reported higher cell performance when the cell was operated at 100% RH, implying that self-humidification did not occur. To this end, the principle and mechanism of self-humidification in BPMFC remains unclear, therefore, the relationship between BPM mode, reaction at PEM/AEM interface, and self-humidification behavior requires further investigation.

It is well known that water transport plays an important role in the operation and durability of low temperature fuel cells. In the past two decades, numerous papers focused on the issue of water transport through the polymer electrolyte membrane, mostly based for PEMFCs, have been reported, e.g. Springer et al. [10–13], Sui and Djilali [14], Kotaka et al. [15], and Owejan et al. [16], among many others. Studies on the water transport of AEMFCs were not reported until recently when their performance had reached to a certain level, e.g. Zhao et al. [17], Yang et al. [18]. Similar to AEMFCs, the water transport in BPMFC was not significant until more recently when their performance finally increased to a higher record, e.g. Xiang et al. [5–7].

A unique feature of the BPM in terms of water transport is the PEM/AEM interface where protons and hydroxide ions meet and form water. In our previous work, a zero-dimensional reaction kinetic model of the BPM reaction interface was proposed based on the p–n junction theory and chemical reaction kinetics [5]. It was demonstrated that water formation reaction kinetics is distinctly faster relative to the water dissociation reaction. The PEM/AEM interface with water formation may strongly contribute to the self-humidification of the cell. Subsequently, a one-dimensional (1-D) steady state water transport model for BPM was proposed to investigate water distribution in BPM [7] where simulations were carried out by solving the water transport equations of each membrane with a point water source at the PEM/AEM interface

with prescribed RH conditions at the membrane boundaries. The 1-D simulation suggested that diffusion and EOD separately drives the product water from the PEM/AEM interface to each membrane's boundary. The water consumption required for ORR could also be compensated by water from this source as well. An important finding of [7] is that the possibility of self-humidification within the BPM was confirmed. Nevertheless, it is felt that the water management in the BPM MEA is more complicated than the situation modeled in the 1-D BPM because of the fact that the water source/sink on the PEM/AEM interface [7] and the CL are multi-dimensional [19] and water transport may be related to local reaction rate. Therefore, to fully explore the self-humidification strategies in MEA, the transport in the CL and gas diffusion layer (GDL) as well as the electrochemical reactions should both be taken into consideration.

In the present work, a steady state MEA model for BPMFC is developed in a two-dimensional (2-D) cross-channel configuration involving three electrochemical reaction zones (anode CL, cathode CL and PEM/AEM interface). With the model capturing conditions close to reality, water transportation is fully investigated and analyzed throughout the entire cell. The model is also validated by experimental results. The effects of membrane thickness and water uptake were evaluated and projected for the formation of self-humidification in BPMFC. Critical current voltage values could be optimized by carefully modulating the membrane thickness and water uptake properties during BPM fabrication.

2. Mathematical formulation and numerical solution

2.1. Model assumptions and computational domain

The main assumptions of the model are summarized as follows:

1. GDLs, MPLs, CLs, PEM and AEM are homogeneous materials.
2. Cell temperature is constant and uniform (isothermal model).
3. Pressure is constant and uniform (isobaric model).
4. No liquid water is present in the MEA.
5. Gas phases $\text{H}_2/\text{H}_2\text{O}$ in the anode and $\text{O}_2/\text{H}_2\text{O}$ in the cathode are considered to be ideal gases.
6. PEM and AEM are impermeable to reactant gases.
7. The cell is operating at steady state conditions.
8. Protons can only transport through the acid electrolyte. Hydroxide ions can only transport through the alkaline electrolyte.
9. The following phenomena are considered in the electrical process of the BPMFC.
 - a Three-phase reaction interface in the catalyst layer (CL);
 - b EOD by hydroxide in the anion exchange ionomer;
 - c EOD by proton in the proton exchange ionomer.

2.2. Computational domain

Computational sub-domains including the GDLs, MPLs, CLs, PEM and AEM of the 2-D model are shown in Fig. 2. The PEM/AEM interface is modeled as a space charge region (SCR) consisting of two components in PEM (width of d_p) and AEM (width of d_a), respectively [5]. The width of the SCR ($d = d_p + d_a$) is determined by junction potential E_j (V), which is influenced by the cell's operating conditions.

2.3. Governing equations

2.3.1. Electrical equations

A model is developed to simulate the BPMFC with water formation reaction (WFR). The PEM/AEM interface introduces an

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