



# A self-humidifying acidic–alkaline bipolar membrane fuel cell



Sikan Peng<sup>a,1</sup>, Xin Xu<sup>a,1</sup>, Shanfu Lu<sup>a</sup>, Pang-Chieh Sui<sup>b</sup>, Ned Djilali<sup>b,\*</sup>, Yan Xiang<sup>a,\*</sup>

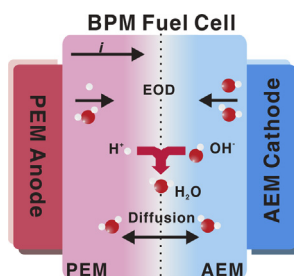
<sup>a</sup> Beijing Key Laboratory of Bio-inspired Energy Materials and Devices, Beihang University, Beijing 100191, PR China

<sup>b</sup> Institute for Integrated Energy Systems, and Department of Mechanical Engineering, University of Victoria, Victoria, BC V8W 3P6, Canada

## HIGHLIGHTS

- Bipolar membrane fuel cells fabricated with a composite acidic–alkaline membrane.
- Self humidifying operation demonstrated with completely dry reactant gas feed.
- Faster kinetics with alkaline cathode allows operation with non-Pt catalyst.
- Theoretical analysis of factors controlling water transport and self-humidification.

## GRAPHICAL ABSTRACT



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## ABSTRACT

To maintain membrane hydration and operate effectively, polymer electrolyte membrane fuel cells (PEMFCs) require elaborate water management, which significantly increases the complexity and cost of the fuel cell system. Here we propose a novel and entirely different approach to membrane hydration by exploiting the concept of bipolar membranes. Bipolar membrane (BPM) fuel cells utilize a composite membrane consisting of an acidic polymer electrolyte membrane (PEM) on the anode side and an alkaline electrolyte membrane (AEM) on the cathode side. We present a novel membrane electrode assembly (MEA) fabrication method and demonstrate experimentally and theoretically that BPM fuel cells can (a) self-humidify to ensure high ionic conductivity; and (b) allow use of non-platinum catalysts due to inherently faster oxygen reduction kinetics on an alkaline cathode. Our Pt-based BPM fuel cell achieves a two orders of magnitude gain in power density of  $327 \text{ mW cm}^{-2}$  at 323 K under dry gas feed, the highest power output achieved under anhydrous operation conditions. A theoretical analysis and *in situ* measurements are presented to characterize the unique interfacial water generation and transport behavior that make self-humidification possible during operation. Further optimization of these features and advances in fabricating bipolar MEAs would open the way for a new generation of self-humidifying and water-management-free PEMFCs.

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## 1. Introduction

The main barriers to large scale adoption of Polymer electrolyte membrane (PEM) fuel cells as a mainstream electrical power source are cost and durability, both of which are largely determined by (i) the perfluorosulfonic acid (PFSA) membranes that ensure ionic transport, and (ii) the catalyst layers that facilitate the

\* Corresponding authors.

E-mail addresses: [xiangy@buaa.edu.cn](mailto:xiangy@buaa.edu.cn) (Y. Xiang), [ndjilali@uvic.ca](mailto:ndjilali@uvic.ca) (N. Djilali).

<sup>1</sup> These authors contributed equally to this work.

electrochemical reactions. Operation of a PEM fuel cell depends critically on good ionic conductivity, and this requires the PFSA membrane remains well hydrated. This is typically achieved by active external humidification [1] using elaborate water and temperature management strategies that increase system complexity and cost. Water management is not only critical for performance, but also impacts durability [2]. In addition, the relatively low operating temperatures ( $<100\text{ }^{\circ}\text{C}$ ) limit the reaction rates, particularly for the cathode oxygen reduction which is sluggish in acidic systems. This is compensated for by using relatively high platinum loadings that increase cost.

The distribution of water in the membrane is primarily determined by the balance between electro-osmotic drag (EOD) and diffusion. Depending on the coupling between membrane properties, local relative humidity (RH) and electric potential, net water transport through the membrane may be directed towards either cathode or anode side of the membrane [3]. Much effort has focused on predicting transport within PFSA type membranes [4] and on optimizing external humidification schemes. Alternatives to conventional humidification that simplify operation without compromising performance have been explored, including for example self-humidifying fuel cells prepared by incorporating the Pt based catalysts into the membrane matrix to catalyze the  $\text{H}_2$  and  $\text{O}_2$  into water [5]; however these cells suffer from the possible short-circuits and lack of stability.

Bipolar membrane (BPM) fuel cells provide another avenue for water management. Unlike monolithic PEM fuel cells, a BPM fuel cell is formed by combining an acidic–alkaline composite membrane with an acidic PEM on the anode side and an alkaline anion exchange membrane (AEM) on the cathode side [6–9]. BPMs have features that open new avenues in two critical areas: (a) water generation takes place at the PEM/AEM interface providing the possibility to devise *passive self-humidification* over the entire cell thereby ensuring high ionic conductivity and lower ohmic losses; (b) alkaline cathode with inherently faster kinetics that allows use of lower catalyst loading and/or less expensive non-platinum catalysts [10–12]. The underlying mechanisms for water transport in a BPM are illustrated in Fig. 1: water diffusion from the PEM/AEM interface drives product water to both electrodes, and the net water transport is determined by the balance between the EOD and diffusion in the BPM. It is possible to control the water fluxes by modifying the BPM pattern and fabrication procedure.

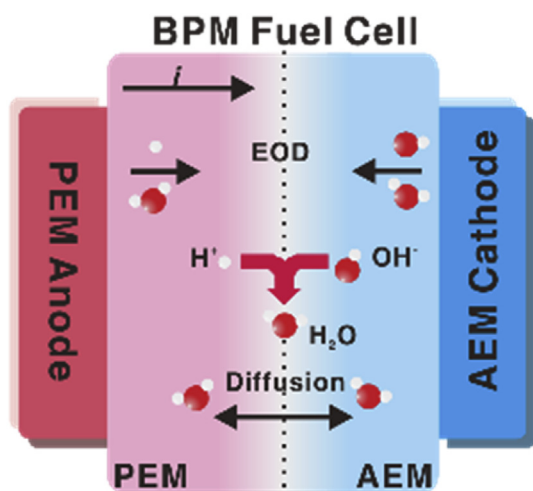


Fig. 1. Schematic of a bipolar membrane (BPM) fuel cell showing directions and driving forces for water transport: EOD and diffusion inside of acidic PEM and alkaline AEM.

Although BPM fuel cells are conceptually promising, the power densities reported to date have been low, e.g.  $0.64\text{ mW cm}^{-2}$  at  $323\text{ K}$  with  $150\text{ }\mu\text{m}$  thick AEM [8]; ca.  $6\text{ mW cm}^{-2}$  at  $333\text{ K}$  with  $35\text{ }\mu\text{m}$  thick AEM [13], and external humidification was needed in all tests. Recently, operation of a hybrid anion–cation membrane fuel cell without external humidification was demonstrated with remarkable power densities [8,14,15]. The key step in this development was hybridization of the electrode with the introduction of an AEM cathode pressed directly onto the PEM. Thus, the hybrid membrane electrode assembly (MEA) consisted of a PEM-ionomer anode, PEM membrane and alkali polymer electrolyte cathode and the hybridization was confined to the electrode structure, as opposed to the hybrid *membrane* concept presented in this work. This suggests that poor performance of previous BPM fuel cells was likely due to the limited success in constructing well integrated composite bipolar membranes. This motivated this study to look into the feasibility of higher performance self-humidifying BPM fuel cells with novel membrane fabrication technique, demonstrate the use of non-Pt catalysts, and perform a theoretical analysis of the self-humidification process.

## 2. Experimental

### 2.1. BPM fabrication

A simple and effective membrane interface fabrication method was developed by first hot-pressing together a pretreated Nafion membrane and a quaternary ammonium polysulfone (QAPS) membrane [16,17] to form a Nafion-QAPS BPM (NQBPM). QAPS is an anion exchange membrane with a relatively simple chemical structure (see Fig. 2) that has been used in several alkaline fuel cell studies [17]. Nafion membranes were pretreated with  $5.0\text{ wt.}\%$   $\text{H}_2\text{O}_2$  and  $0.5\text{ mol L}^{-1}$   $\text{H}_2\text{SO}_4$  following standard procedures [18]. QAPS membranes were cast and dried at  $65\text{ }^{\circ}\text{C}$  using QAPS solution ( $2.0\text{ wt.}\%$ ) in  $N,N$ -dimethylformamide. For example, the thickness of the ionomer solution when casting the membrane was ca.  $330\text{ }\mu\text{m}$ . Following evaporation of the solution and drying, the membrane thickness was  $11\text{ }\mu\text{m}$ . The QAPS membranes were then immersed in aqueous  $1.0\text{ mol L}^{-1}$  KOH solution overnight to exchange  $\text{Cl}^-$  into  $\text{OH}^-$  in the alkaline ionomer. After several rinses in Milli-Q ultrapure water (Millipore,  $18.2\text{ M}\Omega$  at  $25\text{ }^{\circ}\text{C}$ ) and pretreatment in ethanol, the QAPS membranes were hot-pressed with the pretreated Nafion membranes at  $60\text{ }^{\circ}\text{C}$  and  $4\text{ MPa}$  for  $10\text{ min}$  to form the NQBPM.

### 2.2. MEA fabrication

The acidic anode and Ag-based alkaline cathode were made using the conventional thin hydrophilic electrode method, while the Pt-based alkaline cathode was made using the thick hydrophobic electrode method. Pt catalyst inks were prepared by mixing Nafion solution ( $5\text{ wt.}\%$ ), Pt/C catalyst ( $50\text{ wt.}\%$ , E-TEK), isopropyl alcohol, and pure water for the thin hydrophilic acidic anode, whereas polytetrafluoroethylene (PTFE) dispersion ( $15\text{ wt.}\%$ ) was used instead of Nafion solution for the Pt-based thick hydrophobic alkaline electrode. Pt loading for both electrodes was  $0.5\text{ mg cm}^{-2}$ . The Ag-based alkaline cathodic catalyst ink was prepared by mixing QAPS solution ( $2.0\text{ wt.}\%$ ), Ag/C catalyst ( $50\text{ wt.}\%$ ),  $n$ -propyl alcohol and Ag loading was  $1.0\text{ mg cm}^{-2}$ . The inks were dispersed by sonication for  $30\text{ min}$  before casting onto a Toray carbon paper (TGP-H-060) gas diffusion layer and dried at  $40\text{ }^{\circ}\text{C}$ . The Pt-based alkaline cathode was then treated by sintering at  $350\text{ }^{\circ}\text{C}$  for  $30\text{ min}$  in  $\text{N}_2$  atmosphere, followed by casting QAPS solution ( $2.0\text{ wt.}\%$ ) and drying at  $40\text{ }^{\circ}\text{C}$  to obtain thick hydrophobic electrodes. The alkaline cathodes were subsequently immersed in

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