

A sandwich structured membrane for direct methanol fuel cells operating with neat methanol



Q.X. Wu, T.S. Zhao^{*}, R. Chen, L. An

Department of Mechanical Engineering, The Hong Kong University of Science and Technology, Clear Water Bay, Kowloon, Hong Kong Special Administrative Region, China

HIGHLIGHTS

- ▶ A sandwich structured membrane for DMFCs operating with neat methanol is proposed.
- ▶ The membrane offers better water management for DMFCs operating with neat methanol.
- ▶ The sandwich structured membrane enables improvements in cell performance.

ARTICLE INFO

Article history:

Received 31 October 2012
 Received in revised form 4 December 2012
 Accepted 3 January 2013
 Available online 27 February 2013

Keywords:

Fuel cell
 Direct methanol fuel cell
 Neat methanol
 Sandwich structured membrane
 Water content
 PtRu

ABSTRACT

Water starvation at the anode represents a challenging issue in the development of direct methanol fuel cells (DMFCs) operating with neat methanol. To tackle the issue, a multi-layered membrane, consisting of an ultra-thin reaction layer sandwiched between two thin membranes, is proposed and developed. The reaction layer is composed of well-dispersed PtRu catalysts, SiO₂ nanoparticles and Nafion ionomers. During the fuel cell operation, the methanol permeated from the anode catalyst layer and the oxygen permeated from the cathode catalyst layer meet and react in the reaction layer of the sandwich structured membrane to form water and CO₂. The produced water is then maintained at a relatively high level by the hygroscopic SiO₂ nanoparticles in the sandwich structured membrane. As a result, such a created water source at a high concentration level can supply the water required not only for the anode methanol oxidation reaction but also for membrane hydration. The performance characterization demonstrates that the DMFC with the sandwich structured membrane results in much higher performance than that with a single layer Nafion membrane does.

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1. Introduction

The rapid development of consumable electronic devices in recent years calls for innovation in portable power sources with high specific energy. Among existing alternative technologies, direct methanol fuel cells (DMFCs) have attracted the most attention, mainly due to their unique features including relatively simple and compact system design, low noise and operating cost, high energy-conversion efficiency and specific energy, ease in fuel handling, environmental friendliness and low-temperature operation [1–12]. These advantages make this type of fuel cell become a leading candidate to replace batteries in portable applications including notebooks, mobile phones and personal digital assistants. However, the widespread commercialization of DMFC technology is still obstructed by several challenging technical issues such as sluggish kinetics of methanol oxidation reaction (MOR) [13] in

the anode electrode, methanol crossover [14] from the anode to the cathode and the water flooding problem in the cathode electrode [15,16]. To reduce the methanol crossover and the resulting mixed-potential at the cathode catalyst layer (CCL), operating a DMFC with diluted methanol solution (typically lower than 4 M) is the most common way in practice. Such an operation, however, inevitably sacrifices the original advantage of high specific energy ($\sim 4900 \text{ W h L}^{-1}$) [11]. Moreover, as a significant amount of water is introduced to the anode in the diluted methanol operation, the large rate of water crossover from the anode to cathode will aggravate the water flooding problem at the cathode, lowering the cell performance [17].

Recently, in order to increase the specific energy, researchers [18–25] have turned their attention to the development of novel strategies that can suppress methanol crossover under the concentrated methanol operation. These strategies include adding a compact microporous layer (MPL) [18], employing an additional porous layer with a high transfer resistance between the fuel reservoir and the anode flow field [19,20], developing an innovative microfluidic anode flow field [21] and introducing a pervaporation membrane

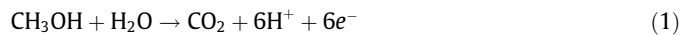
^{*} Corresponding author. Tel.: +852 2358 8647; fax: +852 2358 1543.
 E-mail address: metzhao@ust.hk (T.S. Zhao).

to control the delivery rate of highly-concentrated methanol solution [22–25]. Unfortunately, although the methanol-crossover rate can be dramatically reduced by the above proposed approaches, the cell performance at the highly-concentrated methanol operation also suffers from an unexpected reduction as compared to the diluted-methanol operation. It is believed that the water shortage problem in the anode electrode is the main reason [11,26]. In particular, when feeding neat methanol to the anode, one of the reactants for the anode MOR, water, is no longer contained in the fed fuel. Under such a circumstance, the water shortage problem at the anode becomes the worst. To complete the anode MOR at the neat-methanol operation, one of the solutions is that the produced water at the CCL spontaneously diffuses through the membrane to the anode by utilizing the water concentration gradient between the anode and cathode. As a consequence, the water transport flux from the cathode to anode and the water concentration in the anode catalyst layer (ACL) become factors that limit the anode MOR performance at the neat-methanol operation. In addition, it is worth mentioning that the proton transfer resistance of the membrane is also related to water concentration within the membrane electrode assembly (MEA) and the proton conductivity of the membrane increases with the water content. Therefore, elevating the water concentration across the MEA is also critically important for the maximization of the cell performance at the neat-methanol operation by reducing the Ohmic loss. However, only a few efforts [27–34] have been directed to such an important issue of the water management in DMFCs operating with neat methanol. In an attempt to gain understanding of the water transport characteristics through the membrane at the neat-methanol operation, Wu and Zhao [27] experimentally determined the water transport flux from the cathode to anode by changing various structural parameters and found that the water transport flux depended primarily on the designs of the anode gas diffusion layer (GDL), membrane and cathode GDL. To achieve a better water management, Xu et al. [23] introduced a water management layer (WML) between the cathode GDL and the cathode flow field, Masdar et al. [28] proposed adding a hydrophobic air filter next to the cathode flow field, Li and Faghri [29] employed a perforated cover with a low open ratio at the cathode and Park et al. [30] developed

a multilayer MEA structure with hydrophilic and hydrophobic layers in the cathode. All these approaches can increase the mass transfer resistance of water from the CCL to the cathode side, which facilitates the water back diffusion from the cathode to anode. Rather than modification of the cathode structure, Wu et al. [31] added a water retention layer, consisting of SiO_2 particles and Nafion ionomer, onto both sides of the membrane to buildup the produced water within the MEA. Inspired by the original work done by Watanabe et al. [35] who proposed adding Pt catalysts into the Nafion membranes for self-humidification in proton exchange membrane fuel cells (PEMFCs), in this work, a sandwich structured membrane, which is made of an ultra-thin reaction layer sandwiched between two thin membranes, is developed to improve the water management. Such a reaction layer comprises well-dispersed PtRu catalysts, nanosized SiO_2 particles and Nafion ionomer. By virtue of highly-active PtRu catalysts, the permeated methanol from the ACL and the permeated oxygen from the CCL can react with each other to generate water and CO_2 in the reaction layer of the sandwich structured membrane. As the transport distance of water is shortened, the water back diffusion can be enhanced. In addition, the hygroscopic SiO_2 nanoparticles in the sandwich structured membrane enable the produced water to be maintained at a relatively high level. As a result, such a created water source at a high concentration level can supply the water required not only for the anode MOR but also for membrane hydration.

2. Basic idea

Consider a conventional MEA design shown in the left-hand side of Fig. 1. Under the neat-methanol operation, the water required for the anodic MOR:



is transported from the cathode, where water is produced from the oxygen reduction reaction (ORR):

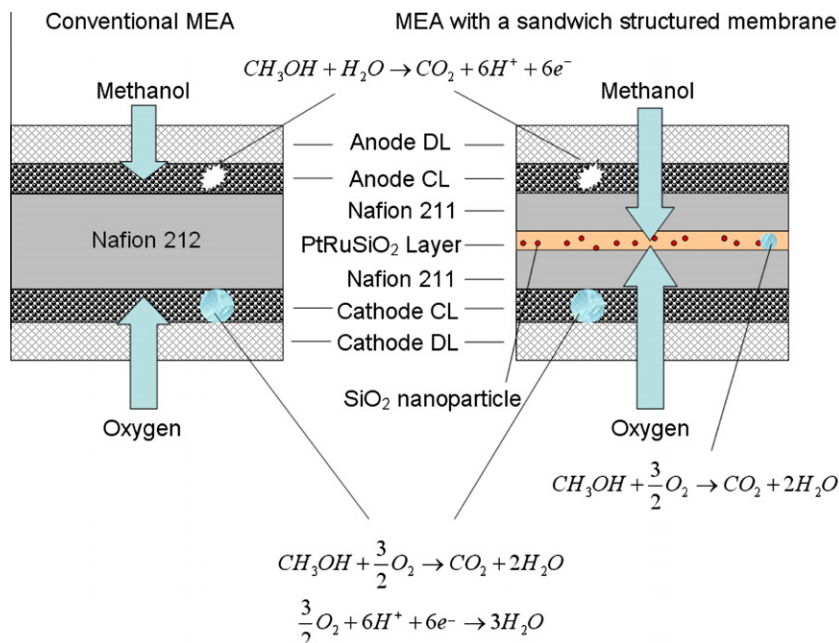


Fig. 1. Design of a sandwich structured membrane.

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