



Research Paper

Transient investigation of passive alkaline membrane direct methanol fuel cell

Bowen Wang^a, Yibo Zhou^b, Qing Du^a, Yan Yin^a, Kui Jiao^{a,*}^a State key Laboratory of Engines, Tianjin University, 92 Weijin Road, Tianjin, China, 300072^b China Automotive Technology & Research Center, Tianjin, China, 300300

HIGHLIGHTS

- A transient model for passive AAEM-DMFC is developed.
- Effects of current, voltage and methanol feed concentration are investigated.
- Optimal energy density is achieved at a moderate current or voltage.
- Anode and cathode MPL can both increase fuel efficiency.
- MPL is critical on energy density under different operation conditions.

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ABSTRACT

Passive direct methanol fuel cell (DMFC) is considered as an attractive power source for portable devices. In this study, a transient multiphase model for passive alkaline anion exchange membrane DMFC (AAEM-DMFC) is developed. The results show that the power density decreases during the operation and is more difficult to maintain stable with higher current density or lower voltage. The fuel efficiency increases with the increment of current density or the decrement of voltage. With a certain concentration, the optimal energy density is achieved at a moderate current density or voltage. Increasing the initial methanol concentration in reservoir can improve the energy density but decrease the fuel efficiency. Anode micro-porous layer (MPL) can improve the fuel efficiency and increase the energy density with high initial concentration (or low current density) but cause negative effect on the energy density with low initial concentration (or high current density). Cathode MPL shows the similar but less significant effect. Generally, whether to use MPL should be determined by different operating conditions and the use demand.

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1. Introduction

In recent years, with the popularization and development of portable devices, such as smart phones and laptop computers, the demand for efficient, enduring and reliable micro power sources is growing [1,2]. Direct methanol fuel cells (DMFCs) have been considered as desired portable power source due to its advantage of high energy density and design flexibility [2–4]. Passive DMFC is one basic type of DMFC in which methanol diffuses by concentration gradient into anode from reservoir and oxygen diffuses passively into cathode from the ambient air [4]. Compared to active DMFCs, passive DMFCs operate without fuel pump and air fan which benefit minimizing the cell volume and reducing the energy loss of driving these accessory devices.

Methanol crossover (methanol crossing the membrane from anode to cathode) is one major problem for DMFC which reduces fuel utilization and cell performance [5]. Using dilute methanol solution as fuel is a simple method to weaken this adverse phenomenon but also lower the energy density. As to the passive DMFC, due to the lack of those auxiliary devices, the operational condition such as methanol flow rate or methanol concentration is hard to control, therefore, a passive DMFC shows a lower performance than an active DMFC. In the previous studies, a plenty of researches about optimizing the water management [6,7] and membrane electrode assembly (MEA) design [8–16] have been done to reduce the methanol crossover and improve the performance for DMFC.

Micro-porous layer (MPL) which is thin porous media inserted between gas diffusion layer (GDL) and catalyst layer (CL) has been considered effective to reduce methanol crossover in recent studies [9–11]. For example, Shaffer and Wang [10] found that the use of MPL can reduce the water crossover from anode to cathode which leads to lower methanol crossover. Further, the anode MPL

* Corresponding author. Tel.: +86 22 27404460; fax: +86 22 27383362.
E-mail address: kjiao@tju.edu.cn (K. Jiao).

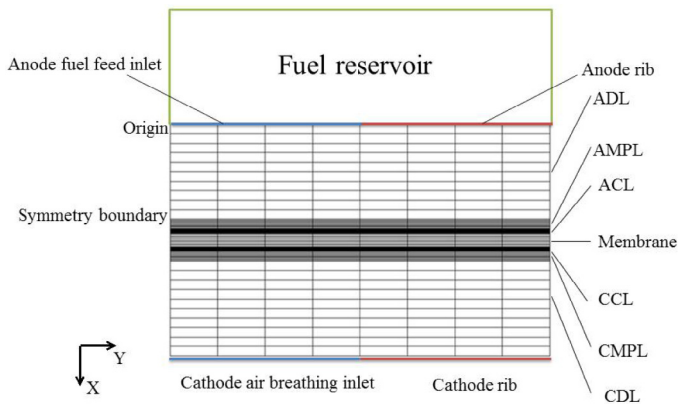


Fig. 1. Computational domain and mesh.

presents more significant effect than cathode MPL. Zago et al. [11] indicated that the anode MPL is critical to maintain an adequate methanol concentration in anode due to the descent of methanol crossover and the increase of the mass transport loss. Besides MPL, alkaline anion exchange membrane (AAEM) is considered as a desired membrane, comparing by conventional proton exchange membrane (PEM), which is beneficial for reducing the methanol crossover and optimizing the water management [13–16]. Moreover, the electrochemical kinetics is faster in alkaline media than in acid media [17]. It is expected that the combination of MPL and AAEM in DMFC can effectively control methanol crossover and improve cell performance [18].

For a passive DMFC, many steady-state models have been developed to study the influence of operating parameters, such as current density, methanol concentration and structure of MEA (especially the role of MPL) on cell performance [19,20]. However, the actual operating process of a passive DMFC is that the methanol concentration at anode inlet keeps decreasing with the fuel consumed and the cell outputs power decreases continuously until the fuel is used up. The steady-state models cannot simulate the whole operating process of a passive DMFC. Therefore, developing a transient model has many advantages which can present the change of cell performance with the operating time and some performance parameters such as energy density and fuel utilization based on the whole operating process. Some transient models were developed in recent years [21,22]. Xiao et al. [21] found that the operating duration is shortened with the decrease of initial feed concentration or the increase of current density. Guo et al. [22] revealed the role

of MPL on the transient performance. However, to the best of the authors' knowledge, the previous transient models are all for PEM-DMFC, and transient investigation for passive AAEM-DMFC is needed.

In this study, a transient multiphase model for passive AAEM-DMFC is developed to investigate the transient transport characteristics and evaluate the performance parameters during the whole working duration under various operating conditions. The role of MPL on the transient performance of passive AAEM-DMFC is also elucidated.

2. Model description

2.1. Physical problem

The computational domain and mesh is shown in Fig. 1. The fuel reservoir stands straight on the top of anode and methanol enters the cell passively without any mechanical device. Oxygen is also supplied passively from the ambient air in cathode inlet. The two-dimensional computational domain shows the typical components of passive DMFC which includes seven layers: anode diffusion layer (ADL), anode MPL (AMPL), anode catalyst layer (ACL), A201 membrane, cathode catalyst layer (CCL), cathode MPL (CMPL), cathode diffusion layer (CDL). The anode, cathode and overall reactions are

Anode oxidation:



Cathode reduction:



Overall reaction:



2.2. Governing equations

In this transient two-dimensional two-phase model, eleven conservation equations are solved in different layers of the computational domain. All the relevant transport properties and source terms involved in the governing equations are listed in Tables 1–4 [15,23–29].

(1) Electric potential (ADL, AMPL, ACL, CCL, CMPL, CDL)

$$\nabla \cdot (\kappa_s^{\text{eff}} \nabla \varphi_s) + S_{Vs} = 0 \quad (4)$$

Table 1
Design and material parameters.

Parameter	Symbol	Value
Diffusion layer thickness	δ_{DL}	2.0×10^{-4} m
Micro-porous layer thickness	δ_{MPL}	2.0×10^{-5} m
Catalyst layer thickness	δ_{CL}	1.0×10^{-5} m
Membrane thickness	δ_m	2.8×10^{-5} m
Channel width	δ_{ch}	5.0×10^{-4} m
Rib width	δ_{rib}	5.0×10^{-4} m
Height of fuel reservoir	h_0	1.0×10^{-3} m
Porosity of DL,MPL,CL	$\varepsilon_{DL}, \varepsilon_{MPL}, \varepsilon_{CL}$	0.6, 0.3, 0.3
Contact angle of DL,MPL,CL	$\theta_{DL}, \theta_{MPL}, \theta_{CL}$	$110^\circ, 120^\circ, 95^\circ$
Permeability of DL, MPL, membrane	K_{DL}, K_{MPL}, K_m	2.0×10^{-12} m ² 5.0×10^{-13} m ² 2.0×10^{-18} m ²
Permeability of ACL and CCL	K_{ACL}, K_{CCL}	5.0×10^{-14} m ² 1.0×10^{-13} m ²
Electrolyte volume fraction in ACL and CCL [15]	ω_a, ω_c	0.34, 0.3
Density of dry A201 membrane [23]	ρ_m	1092.7 kg m ⁻³
Equivalent weight of A201 membrane [23]	EW	0.5882 kg mol ⁻¹

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