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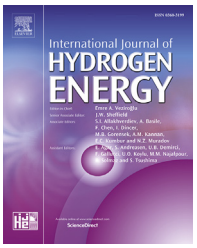

In-situ investigation of bubble dynamics and two-phase flow in proton exchange membrane electrolyzer cells

Yifan Li, Zhenye Kang, Jingke Mo, Gaoqiang Yang, Shule Yu, Derrick A. Talley, Bo Han, Feng-Yuan Zhang *

Nanodynamics and High-Efficiency Lab for Propulsion and Power, Department of Mechanical, Aerospace & Biomedical Engineering, UT Space Institute, University of Tennessee, Knoxville (UTK), USA

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ABSTRACT

Gas bubble dynamics and two-phase flow have a significant impact on the performance and efficiency of proton exchange membrane electrolyzer cells (PEMECs). It has been strongly desired to develop an effective experimental method for *in-situ* observing the high-speed/micro-scale oxygen bubble dynamics and two-phase flow in an operating PEMEC. In this study, the micro oxygen bubble dynamic behavior and two-phase flow are *in-situ* visualized through a high-speed camera coupled with a specific designed transparent PEMEC, which uses a novel thin liquid/gas diffusion layer (LGDL) with straight-through pores. The effects of different operating conditions on oxygen bubble dynamics, including nucleation, growth, and detachment, and two-phase flow have been comprehensively investigated. The results show that temperature and current density have great effects on bubble growth rate and reaction sites while the influence of flow rate is very limited. The number, growth rate, nucleation site, and slug flow regime of oxygen gas bubbles increase as temperature and/or current density increases, which indicates that an increase in temperature and/or current density can enhance the oxygen production efficiency. Further, a mathematical model for the bubble growth is developed to evaluate the effects of temperature and current density on the bubble dynamics. A mathematical model has been established and shows a good correlation with the experimental results. The studies on two-phase flow and high-speed micro bubble dynamics in the microchannel will help to discover the true electrochemical reaction at micro-scale in an operating PEMEC.

Introduction

With the growing development of various energy production and use industries, such as aerospace, public transportation and domestic use, the traditional energy production and storage methods can no longer meet the public demands because of its global-warming potential, low energy density, risk of explosion, continuous growing price, and carbon monoxide inhalation [1,2]. With this background, water electrolysis, for example, uses electricity to split water into its

* *Corresponding author.*
E-mail address: fzhang@utk.edu (F.-Y. Zhang).

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constituent oxygen and hydrogen atoms, offering the most promising method currently known for oxygen and hydrogen production [3]. Electrolysis can take place quickly at different scales ranging from micro to macro. The advanced proton exchange membrane electrolyzer cells (PEMECs) used for this purpose are widely considered to offer an efficient way of producing oxygen and hydrogen from renewable energy. What's more, PEMECs offer a range of benefits, including heightened effectiveness, quick response times, higher levels of gas purity, and ease of handling [4–9]. However, they present certain challenges of cost, durability, and efficiency [10–13].

In an operating PEMEC, the oxygen evolution reaction (OER) occurs at anode side and must meet the triple-phase boundary requirements involving electrons, catalysts (IrRuOx), proton carriers, and pathways for water and oxygen. Water is circulated during the operation at the anode side of the PEMECs to the interface between the catalyst layer and the liquid/gas diffusion layer (LGDL) where it reacts with the catalyst to be split into oxygen, protons, and electrons. The oxygen is carried with the incoming water flow to the outlet at the anode. The protons are transferred from the anode side to the cathode side through the membrane in the middle of the cell and then react with the electrons from outside to form hydrogen gas.

The electrochemical reactions at the anode and cathode of a PEMEC can be given as Equations (1) and (2).

$$\text{Anode}: 2H_2O \xrightarrow{Catalyst} 4H^+ + O_2 + 4e^- \quad (1)$$

$$\text{Cathode}: 4H^+ + 4e^- \xrightarrow{Catalyst} 2H_2 \quad (2)$$

Some experimental and modeling studies have been already conducted to the bubble and two-phase transport dynamics inside a water electrolyzer under various conditions. Mo et al. discovered the inside fluid phenomena by using a high-speed visualization system together with a specifically designed transparent PEMEC that allowed them to gather almost all the information they needed in the microchannels. They observed a very interesting phenomenon that the bubble only generated at the rim of the pores, and that almost no bubble generated in the center [14–16]. Sakuma et al. observed single and multiple oxygen low-speed bubbles evolution phenomena, with the bubble diameter increasing from about 0.02 mm to 0.6 mm within 4.6 s. They found the surface wettability played a major role in bubble growth behaviors. Nucleation sites decreased and bubble sizes became large with increasing surface hydrophobicity. Bubbles were also difficult to detach from hydrophobic electrode. The result was discovered by using the current interrupter method, which could measure the ohmic resistance corresponding to the surface coverage [17]. Lafmejani et al. studied the gas-liquid flow in a PEMEC by using the computational fluid dynamics simulation method and in-situ experimental research, which employed 4 mm width channel, 0.968 m/s inlet liquid velocity and 6.38×10^{-4} m/s inlet gas velocity. Both the experimental and simulation results showed that there were some small bubbles between the long taylor bubbles, and it was also found that the shape of the micro-channel could affect the behavior of taylor bubbles [18]. Ito et al. investigated the influence of water flow in a channel on the gas production efficiency of an operating solid polymer water electrolyzer and the detachment phenomena of hydrogen gas bubbles in a 1000 μm width channel. The study found that production efficiency increased with water flow rate and bubbles were more easily to detach at 120 cc/min and 0.3 MPaG [19]. Dedigama et al. measured and visualized the two-phase flow phenomena relating to the operation potential of a transparent single channel PEMEC (3 mm × 85 mm) with different water flow rates (1, 3, 5 ml/min). They found bubble diameter was changed from 0.87 mm at low potential with a flow rate of 5 ml/min to 2.93 mm at high potential with a flow rate of 1 ml/min, which meant the oxygen bubble size could increase with increasing in potential and decrease with increasing in water flow rate [20]. Chandran et al. observed constant increases in the size of hydrogen bubbles when they were generated at different locations. Bubble sizes varied from 40 to 100 μm at the surface of an electrode. When the bubble was generated at a position of 2 mm away from electrode, it would change from 100 to 130 μm. Bubble growth velocity also varied from 0.3 cm/s to 0.76 cm/s [21]. Bo et al. established a mathematical model to simulate the two-phase flow behaviors in a PEMEC and did a comprehensively investigation of different factors including contact angle, porosity, and pore size, on the capillary flow and the distribution of liquid water in microchannel. The results showed that an increasing in contact angle will raise the cell voltage and decrease cell performance, the increased LGDL porosity will enhance the cell performance. The other factors like the membrane thickness, current density also have a significant influence on the cell performance [22–25].

Many researchers have studied on the dynamics of bubble evolution and two-phase flow in different systems, including a boiling system, a simulator, a fuel cell, and a water electrolyzer cell. Yang et al. used a microscope and a high-speed camera to track the lifetime and size of single hydrogen bubbles on a platinum microelectrode. It is found that the bubble increased from 50 to 250 μm within 1 s [26]. Zhang et al. took advantage of micro visualization system and designed transparent PEM fuel cells. They successfully *in-situ* revealed the micro drop dynamics on the GDL surface and two-phase flow in micro channels [27,28]. For further *in-situ* investigation of the dynamics of microdroplets and electrochemical reactions on the CL surface, including micro drop formation, growth, coalescence, and removal, Zhang et al. developed metallic gas diffusion layer with straight pores and catalyst-visible operational fuel cells. They showed reducing the GDL pore size and increasing catalyst layer hydrophobic property would retain liquid water in the gas phase [29–31]. Kumbur et al. and LaManna et al. investigated multiphase capillary transport through the thin diffusion media, microporous layer, catalyst layer and their interfaces, and proposed a new correlations between capillary pressure–liquid saturation. They also found that any hydrophobicity increase in the DM led to higher capillary pressures and less flooding [32–34]. LaManna et al. further studied interfacial effects for DM|MPL and MPL|CL the diffusion thoroughly Cubaud et al. investigated different types of liquid/gas flow patterns in square microchannels and observed five primary types of flow regimes in hydrophilic channels: bubbly, wedging, slug, annular, and dry flow. Three different flow regimes were also seen in

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