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Short communication

In operando synchrotron X-ray radiography studies of polymer electrolyte membrane water electrolyzers

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ABSTRACT

Polymer electrolyte membrane water electrolysis (PEL) cells are studied in-operando by synchrotron X-ray radiography. Two-phase flow phenomena associated with the evolution of oxygen and hydrogen in the surrounding water are investigated on a running electrolyzer cell. We examine the gas bubble discharge from the porous transport layer (PTL) into the flow channel and discuss the transport of bubbles in the flow channel. The transport of gas inside the PTL and the number of gas bubble discharge sites is examined and correlated with current density.

1. Introduction

Polymer electrolyte membrane water electrolysis (PEL) is increasingly attracting interest as a means to store electricity generated from intermittent renewable sources like wind or solar [1]. Electrolysis is currently considered one of the few available pathways to achieve a 100% renewable electricity supply [2]. PEL can accommodate higher current densities [1] and a wider range of operation compared to alkaline water electrolysis [3]. In PEL, water is typically supplied via flow channels and distributed across the catalyst layer through a porous transport layer (PTL). Gas evolution occurs at the catalyst layer, and the gas is transported back through the PTL and discharged into the flow channels, where it is transported out of the cell as a two-phase mixture with the feed water. For energy storage applications, higher current densities are desired [3], yet the resulting amount of gas produced could lower the overall cell performance due to mass transport restrictions induced by the reduced supply of water to the catalyst sites.

In the past, in-situ synchrotron X-ray radiography (SR) has been a valuable tool for visualizing processes inside operating electrochemical cells without substantial modification of cell layout or disruption of the electrochemical reactions. This method was first applied to running polymer electrolyte membrane fuel cells (PEFCs) by Manke et al., which elucidated the formation and transport of liquid water for the verification of modeling approaches [4]. Further studies described the transport of water through the porous gas diffusion media [5] and eruptive water transport from the gas diffusion media to the flow channels [6]. SR was also applied to direct methanol fuel cells [7] and high-temperature PEFCs [8,9].

Currently, only few studies have investigated two-phase flow phenomena in PEL. Dedigama et al. [10] examined the two-phase flow in the flow channel using a transparent window made of Perspex (poly(methyl methacrylate) or PMMA). Yet differences in contact angles and electrical conductivity of PMMA compared to typical flow channel materials like titanium [1] could introduce perturbations of the flow regime and disturb the current distribution within the cell. Selamet et al. combined neutron radiography with optical imaging to study the evolution of water thicknesses in a PEL cell over time [11]. However, the spatial resolution was limited to 15 μm for optical and 250 μm for neutron imaging. Selamet et al. also applied X-ray imaging to a regenerative fuel cell operated in the electrolysis mode using an X-ray microscope based on an X-ray tube, without reporting a spatial resolution [12].

On the contrary, spatial resolutions of <5 μm can easily be achieved with SR [13]. These resolutions are necessary to study the PTL of a running PEL cell, for which the mean pore diameter can be as low as 8 μm [14]. In this study, we report for the first time in-operando SR imaging of in-plane and through-plane two-phase flow inside a PEL.

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2. Experimental

2.1. Cell preparation and design

Two PEL cells were used for the SR measurements, one with an active area of 11.9 cm^2 (#1) and one of 17.6 cm^2 (#2). Both cells utilize a Nafion N117 membrane (DuPont) coated with a cathode catalyst layer consisting of platinum on carbon (HISPEC 9100 with 60 wt.% Pt by Johnson Matthey) and Nafion (DuPont), and an anode catalyst layer containing iridium oxide (IrO_2 by Alfa Aesar) and Nafion (DuPont). On the anode side, a PTL composed of sintered titanium powder (GKN Sinter Metals) was employed while conventional Toray carbon paper (TGP-H-120 by Toray) directly obtained from Toray Industries (not treated with PTFE) was used on the cathode side. Graphite separator plates with a machined flow field structure were employed with either a meander-shaped single channel on anode and cathode (#1) or equally-spaced parallel channels on the cathode and a meander-shaped single channel on the anode (#2), as shown in Fig. 1. The channels were 1.4 mm wide and 0.4 mm deep.

The cells were operated at 80 °C and ambient pressure. Both the anode side and the cathode side were continuously supplied with water. Water flow rates ranged from 0.6 to 1.5 ml/(min cm^2) while the current density was varied in the range of 10–200 mA/cm^2.

2.2. Synchrotron X-ray radiography

The radiographic measurements were performed at the synchrotron radiography station of the Helmholtz-Zentrum Berlin (BAMline at BESSY II). A monochromatic X-ray beam with an energy of 25 keV was chosen to ensure sufficiently high transmission through all cell components. An optical setup with a (4008 × 2672) $pixel^2$ CCD camera (PCO 4000 with a $CdWO_4$ scintillator screen) was used to capture images with a pixel size of 2.15 μm. The temporal resolution (including exposure time and readout time) was 5 s for the settings chosen.

3. Results & discussion

The X-ray transmission images were normalized to preceding images. An increase in transmission (lighter area in the images) indicates an increase in the gas presence, as the X-ray attenuation coefficient of gases is orders of magnitude smaller than that of the surrounding liquid water. A decrease in transmission (darker area) indicates an increase of water in areas where gas was previously present. These transmission variations allow for analyzing the transport of gas bubbles inside the cell.

In-plane SR was used to visualize gas bubble formation in an operating PEL cell at the PTL/flow channel interface. Fig. 2a illustrates an image series taken on the cathode side interface at different times during the growth of a hydrogen bubble. Such growth proceeds until a critical diameter is reached at which the bubbles are discharged into the flow channel and new bubbles can be formed. The temporal evolution of the bubble diameter and the subsequent bubble discharge, at one specific location at the PTL/flow channel interface, is given in Fig. 2b for different current densities.

In the case of the discharge site shown in Fig. 2a, the average diameter at discharge, the periodicity of bubble formation and the moles n of gas ejected to the flow channel at the critical discharge diameter d are listed in Table 1 for current densities of 10, 75 and

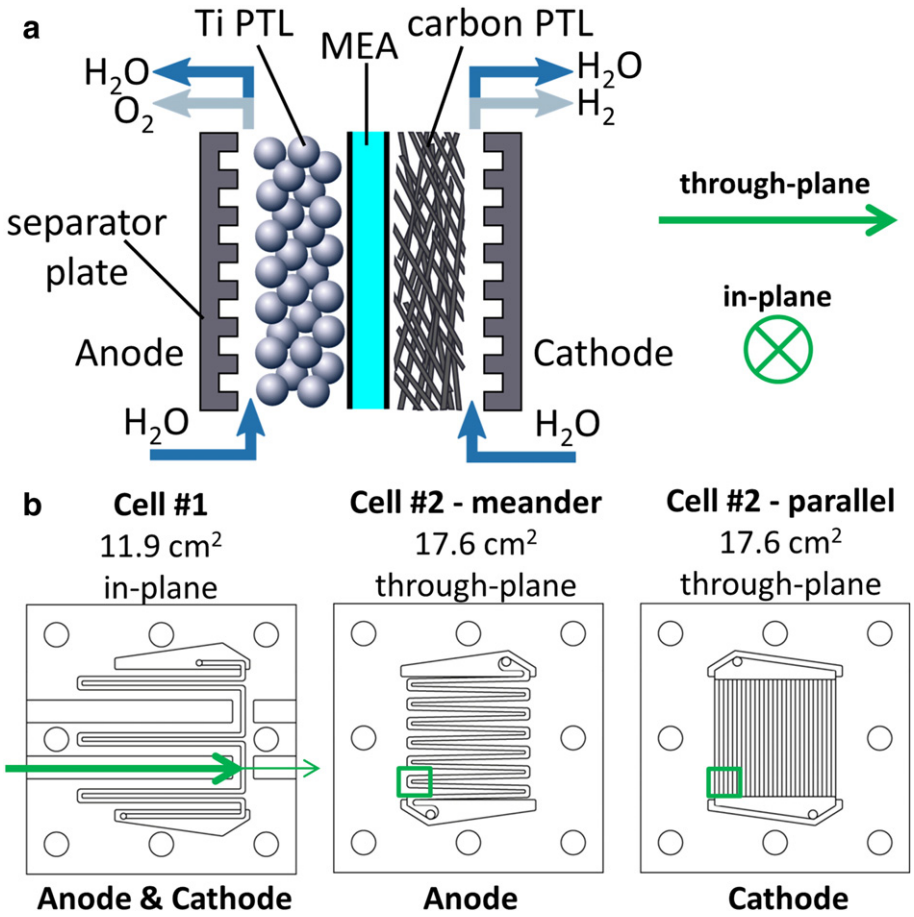

Fig. 1. (a) Sketch of the PEL cell showing the components separator plates, titanium PTL, MEA, carbon PTL with indication of beam direction for in-plane and through-plane measurements. (b) Flow field structures used for in-plane and through-plane measurements.