



## Alkaline anion exchange membrane water electrolysis: Effects of electrolyte feed method and electrode binder content

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### H I G H L I G H T S

- Catholyte feed method and anode binder content in an MEA for AEMWE were examined.
- Dry cathode operation was beneficial for long-term performance.
- Optimal PTFE binder content in the anode was 20 wt% for dry cathode operation.

### A R T I C L E I N F O

#### Keywords:

Alkaline anion exchange membrane water electrolysis  
Electrolyte feed method  
Membrane electrode assembly  
Binder content

### A B S T R A C T

Herein, we investigate the effects of catholyte feed method and anode binder content on the characteristics of anion exchange membrane water electrolysis (AEMWE) to construct a high-performance electrolyzer, revealing that the initial AEMWE performance is significantly improved by pre-feeding 0.5 M aqueous KOH to the cathode. The highest long-term activity during repeated voltage cycling is observed for AEMWE operation in the dry cathode mode, for which the best long-term performance among membrane electrode assemblies (MEAs) featuring polytetrafluoroethylene (PTFE) binder-impregnated (5–20 wt%) anodes is detected for a PTFE content of 20 wt%. MEAs with low PTFE content (5 and 9 wt%) demonstrate high initial performance, rapid performance decay, and significant catalyst loss from the electrode during long-term operation, whereas the MEA with 20 wt % PTFE allows stable water electrolysis for over 1600 voltage cycles. Optimization of cell operating conditions (i.e., operation in dry cathode mode at an optimum anode binder content following an initial solution feed) achieves an enhanced water splitting current density ( $1.07 \text{ A cm}^{-2}$  at 1.8 V) and stable long-term AEMWE performance (0.01% current density reduction per voltage cycle).

### 1. Introduction

The increasing fossil fuel consumption and greenhouse gas emission are a matter of growing concern, requiring the development of renewable energy systems [1]. In this regard, hydrogen is considered to be a sustainable alternative to fossil fuels, exhibiting the advantages of high power conversion efficiency (up to 60%) and zero-carbon operation of fuel cells [2,3]. However, the current methods of hydrogen production (mainly steam reforming of natural gas) result in the emission of greenhouse gases such as carbon dioxide or hydrocarbons [4], necessitating the search for alternative processes to achieve a clean

energy cycle from hydrogen production to power generation in fuel cells, e.g., water electrolysis powered by renewable energy sources [5,6]. In addition, the benefits of electrolytic hydrogen production include increased yields of high-purity hydrogen, relatively modest space requirements for on-site fueling stations, and the reusability of the existing water and electricity infrastructure [7].

Compared to conventional alkaline water electrolysis using alkaline electrolytes and porous diaphragm separators, alkaline anion exchange membrane water electrolysis (AEMWE) features the advantages of safety, efficiency, and gaseous product separation due to employing membrane-based systems [8,9]. Moreover, the operation of AEMWE

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systems at high pH allows the oxygen evolution reaction (OER) to be catalyzed by cost-effective non-noble metal (e.g., Co, Ni) oxides [10,11], whereas water electrolysis systems based on proton exchange membranes require expensive noble metal catalysts (e.g., Ir and Ru oxides) to ensure OER activity and operational durability in acidic environments [12]. In view of these advantages, numerous studies have focused on the development of durable anion exchange membranes [13–23] and electrocatalysts [24–34] for improving the ionic conductivity and catalytic activity of AEMWE systems, respectively, to attain high performance under high pH operating conditions. However, the performance of the developed materials needs to be validated in full-cell configuration due to being dependent on the conditions of electrode and membrane electrode assembly (MEA) fabrication and cell operation. In other words, given that active and durable components are developed for AEMWE, the optimization of electrode and MEA fabrication and cell operation are important for enhancing the properties of the above materials and their performance in full-cell systems [35].

The importance of MEA fabrication and cell operation conditions has been largely demonstrated for applications employing the membrane electrode, e.g., for alkaline or proton exchange membrane (PEM) water electrolysis and fuel cell development. Thus, Dunnill et al. modified the cell design and operation conditions of alkaline water electrolysis to achieve enhanced performance using a zero-gap electrode and an anion exchange membrane [36]. Masel et al. reported the effects of ionomer and membrane composition of the electrode on AEMWE performance, realizing a current density of  $6 \text{ A cm}^{-2}$  at 2.05 V after MEA optimization [37,38]. Jensen et al. also reported the relationship between electrolyte type and AEMWE performance using an *m*-PBI (polybenzimidazole)-based ion-solvating polymer electrolyte [39]. In addition, extensive research has also been performed on the optimization of MEA fabrication for PEM fuel cells (PEMFC, similarly to the case of AEMWE systems discussed above) under various operating conditions by varying electrode structure [40–44] and pressing conditions [45–49]. Jeon et al. showed that the electrode structure can be controlled by varying the Nafion ionomer contents (10–40 wt%) of electrodes and studied its effect on low-temperature (65 °C) PEM fuel cell performance at various relative humidities (RHs) [42], demonstrating that at low RH, the best results were obtained at high ionomer contents. In particular, the MEA with an ionomer content of 30 wt% exhibited the highest performance at 40–95% RH, with that having a 40 wt% ionomer content showing the best performance at 25% RH [42]. Cho et al. also studied the performance of PEM fuel cell MEAs by varying their ionomer content (20–40 wt%) at low relative humidity ( $\leq 35\%$  RH) and intermediate temperature (120 °C) [43], revealing that the MEA with 40 wt% ionomer content performed best under dehydrated conditions, i.e., when the RH of the supplied gas and the water production rate were low [43]. The results of previous MEA electrode structure optimizations demonstrate that device performance is highly dependent on cell operating conditions, which should be taken in consideration when constructing effective electrode structures.

As discussed above, MEA optimization and proper cell operation are important for improving cell performance. Specifically, AEMWE MEAs feature a hydroxide-conducting polymer electrolyte membrane with two electrodes (comprising a mixture of catalyst and polymeric binder) on both sides, similarly to the case of PEMFCs. During water electrolysis, hydrogen gas and hydroxide ions are produced by the hydrogen evolution reaction occurring at the cathode, and oxygen gas, water, and electrons are produced by water (or hydroxide) oxidation at the anode given that hydroxide is transported through the anion exchange membrane [50]. So far, only few MEA optimization studies have been conducted for AEMWE applications, as discussed above [13,51–53], i.e., electrode optimization via catalyst loading [51] or electrode binder content variation [52] and modification of electrode [13,53] or MEA fabrication processes [52]. Moreover, the dependence of cell performance on operating conditions such as cell temperature [51],

electrolyte solution type/concentration [13,51], and feed configurations [50] has been underexplored. As discussed above, the optimum MEA structure varies with cell operating conditions and therefore, the analysis of performance-influencing factors is important to maximize cell efficiency.

Herein, we analyze the dependence of single cell performance on the type of catholyte-supplying procedure and electrolyte basicity to maximize the AEMWE water splitting current, subsequently varying the polymer binder content (5–20 wt%) of the anode under the optimized operating conditions to construct a high-performance MEA for AEMWE. The physicochemical properties of AEMWE electrodes are evaluated by electrochemical analyses (linear sweep voltammetry and impedance spectroscopy) and physical assays (mercury porosimetry and inductively coupled plasma mass spectroscopy) to elucidate the relationship between operation conditions, MEA structure, and cell performance.

## 2. Experimental

### 2.1. MEA preparation and cell operation

Hydrogen and oxygen evolution electrodes were prepared by the catalyst coated substrate method using Pt/C (Pt 46.5 wt%, Tanaka K. K.) and IrO<sub>2</sub> (Premion<sup>®</sup>, Alfa Aesar) powders, respectively. Although IrO<sub>2</sub> was used as a model catalyst in this study, certain OER catalysts are significantly more effective in basic aqueous solution, e.g., pyrochlore catalysts feature a 5–10 times higher activity in 0.1 M KOH [54]. Catalyst ink slurries containing polytetrafluoroethylene (PTFE) binder (60 wt% PTFE dispersion in H<sub>2</sub>O, Aldrich) were prepared using a mixture of isopropyl alcohol (A.C.S. grade, Burdick & Jackson) and deionized water (DI water, 18 M $\Omega$ ) solution. For oxygen evolution electrodes, PTFE contents of 5, 9, 15, and 20 wt% (with respect to the total solid weight) were used, and 250- $\mu\text{m}$ -thick titanium papers were utilized as diffusion layers. An anion exchange membrane (A201, Tokuyama) was used as a solid electrolyte.

For single cell tests, a titanium plate with a parallel flow field and a graphite plate with a serpentine flow field (2.5 cm  $\times$  2.5 cm) were utilized as anode and cathode current collectors, respectively. The cell was operated at 50 °C, and four different catholyte feed methods were tested, i.e., supplying i) DI water as initial and operating feed (F1); ii) DI water as initial feed only, with no feed supplied during operation (F2), iii) 0.5 M KOH solution as both initial and operating feed (F3), and iv) 0.5 M KOH solution as initial feed only, with no feed supplied during operation (F4). The catholyte was supplied at a flow rate of  $3 \text{ mL min}^{-1}$  when required, while 0.5 M KOH was constantly introduced to the anode in all four cases at a rate of  $1 \text{ mL min}^{-1}$ . The use of a basic aqueous electrolyte at the anode enabled facile OER kinetics and allowed one to investigate the effect of catholyte feed method on AEMWE performance. Prior to starting the experiments, initial feeds were supplied for 10 min. The electrolyte feed and electrode fabrication conditions are summarized in Table 1.

### 2.2. Electrochemical analyses

For electrochemical analyses, a high-current potentiostat (HCP-803, Bio-Logic) was used to control cell voltage and measure impedance. The AEMWE current density was measured for repeated voltage cycles from 1.5 to 2.2 V at a scan rate of  $20 \text{ mV s}^{-1}$ . The current density measured in each cycle was insignificantly affected by the scan rate in the range of  $1\text{--}20 \text{ mV s}^{-1}$ , but a performance change was observed upon repeated cycling as described below. Electrochemical impedance spectroscopy (EIS) was employed to determine the cell performance-affecting resistances for different operating and electrode fabrication conditions, with the corresponding analyses performed at 1.8 V, an alternating voltage amplitude of 10 mV in the AC frequency range of 15 kHz–30 mHz.

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