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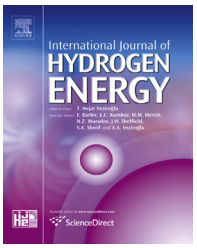

Performance of high temperature PEM fuel cell materials. Part 1: Effects of temperature, pressure and anode dilution

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ABSTRACT

High temperature proton exchange membrane (HT-PEM) fuel cells operate most effectively at temperatures 160 °C or greater and can tolerate high carbon monoxide concentrations in fuel feeds. For practical mobile fuel cell systems, the ability to tolerate high levels of carbon monoxide enables a simplified integrated reformer fuel cell design, where a one-step reforming process can directly feed the fuel cell. This creates the potential for practical fuel cell systems to be designed that are capable of operating using an on-board hydrocarbon fuel. Hydrocarbon reforming processes (i.e., partial oxidation, steam reforming, or autothermal reforming) generate effluent gas compositions that typically contain various amounts of hydrogen, carbon monoxide, carbon dioxide, nitrogen, and water. While there are several studies that have examined the effects of CO dilution on HT-PEM fuel cell performance, there are very few that have examined the effects of other common reformate species, particularly while operating at varying temperatures and pressures. This work aims to fill the research gap in published data through evaluation of the performance exhibited by two HT-PEM membrane-electrode assembly (MEA) types, Advent's TPS©-based MEA and the BASF Celtec series MEA, based on acid-doped polybenzimidazole (PBI). In Part I of this series, the effects of temperature and pressure for various percentages of anode dilution were investigated. Temperatures and pressures were varied from 160 to 200 °C and 101.3–200 kPa, respectively. The effect of hydrogen diluted with nitrogen in the anode feed was examined with nitrogen concentrations up to 70%. Overall, the PBI MEA provided greater performance than the TPS for all tested conditions. Our results show that the performance loss due to high dilution levels can be wholly mitigated through an increase in operating temperature, pressure, or a combination thereof. For example, even at a dilution level containing 70% nitrogen, operating the cell at 200 °C and at 200 kPa provides the same power output as running the cell on pure hydrogen at 160 °C and at atmospheric pressure. This data can be used to model the performance effect of high diluent concentrations in the anode gas feed when developing an integrated reformer/fuel cell system.

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Introduction

System designs integrating an on-board fuel reformer with a conventional PEM fuel cell using Nafion©-based MEAs are very complex, involving multiple fuel reforming and shift reactors to purify the hydrogen stream entering the fuel cell. Most single-step reforming processes tend to generate large amounts of carbon monoxide (CO), which adsorbs strongly onto platinum (Pt) at low temperatures, poisoning the electrode catalyst. For conventional polymer electrolyte membrane (PEM) fuel cells (PEMFCs) that operate around 80 °C, significant performance loss is observed at CO concentrations in the anode feed in excess of 10 ppm [1,2]. To reduce CO concentrations below 10 ppm, multiple reactors such as water gas shift (WGS) [3], preferential oxidation (PROx) [4], and pressure swing adsorption (PSA) [5] are required to clean up the reformate after the initial reforming reaction. Operating several reactors to produce dynamic power for mobile applications is impractically complex, and a CO-tolerant PEMFC technology would greatly simplify the overall system design.

High temperature PEM fuel cells (HT-PEMFC) offer numerous advantages over conventional PEM fuel cells including fuel flexibility and reduction in balance of plant (BoP) equipment required at the system level. Operating at temperatures typically between 160 and 200 °C allows the fuel cell to generate electricity using hydrogen feeds that contain much higher concentrations of contaminants than can be tolerated with conventional Nafion-based PEM fuel cells [2,6,7]. For example, Li et al. [2] considered the effect of CO concentrations up to 16% in a HT-PEM fuel cell. Although the performance of the fuel cell with CO concentrations of 16% was the lowest of all the tested operating conditions, useful amounts of power were generated nonetheless. This demonstrates that high power outputs are achievable with HT-PEM fuel cells, even when operating on very high concentrations of diluents. Additionally, operating at temperatures above the boiling point of water reduces complex water management BoP equipment because water can easily be removed from the cell as a vapor rather than as a liquid. These benefits allow for a much simpler integrated fuel reformer and fuel cell stack design.

Of the novel types of membranes used for HT-PEMFCs, such as those described in Refs. [8–11], only a few have seen some commercial success. One of the commercially available membranes is based on a pyridine containing poly(ether sulfone) [12–14], also known as TPS®, and is currently produced by Advent Technologies Inc [15]. A large focus of researchers using TPS-based membrane electrode assembly (MEAs) has been on developing direct methanol fuel cells (DMFC) such as in Refs. [14,16]. While commercially available, there is, however, limited MEA performance data based on Advent's TPS technology in published literature.

The most widely studied HT-PEMFC membrane is the Celtec-P series developed by BASF based on acid-doped polybenzimidazole (PBI) [17], and it has seen some commercial success [18]. Other commercially available PBI based MEAs are offered by Danish Power System [19], and were investigated in Ref. [20]. The Celtec MEAs were chosen for use in our research as they appear to be more widely used in prior published research and they represent the state-of-the-art in commercially available HT-PEM MEAs.

Numerous performance studies have been conducted using the Celtec series MEAs investigating parameters such as temperature [21,22], CO concentration [17,23,24], relative humidity [25,26], and durability [27–29]. Most performance studies for PBI-based HT-PEMFCs have focused largely on cell performance when operating on pure H_2, or H_2 mixed with various amounts of CO, and at atmospheric pressure. However, if hydrogen is generated from an on-board fuel processor using a catalytic partial oxidation (cPOx), steam reforming (SR), or autothermal reforming (ATR) process, other gases may be produced that will enter into the anode of the fuel cell stack, such as N_2, CO_2, H_2O, CH_4, C_2H_4, and C_2H_6. For example, under optimal reforming operating conditions, propane cPOx will produce effluent gas composed of 28% H_2, 23% CO, and 49% N_2 [30].

Presently, very little data for HT-PEM fuel cells can be found while operating on high concentrations of N_2 in the anode, as well as under high pressures up to 200 kPa. If future integrated reformer HT-PEMFC systems are to be developed, the impact of N_2 dilution on a HT-PEMFCs performance should be investigated, as well as the effect of operating pressure. This article is the first in a series that attempts to fill this gap by investigating the performance of commercial TPS and PBI-based MEAs while varying the temperature, pressure, and different anode dilution concentrations with nitrogen (N_2). It is important to note that the effect of anode N_2 dilution is the focus of this study rather than CO_2, another common fuel reforming effluent gas; this is for two reasons. The first is because some fuel reforming processes generate significant amounts of N_2 in the effluent, and very little if any CO_2 as described in the previous paragraph. The second is that several other works that investigate the effect of CO_2 concentrations in the anode up to 25% versus N_2 concentrations at the same level, show that N_2 and CO_2 result in the same performance loss when operating at the temperatures investigated in this study [2]. However, the performance loss when operating at lower cell temperatures, such as 60 °C, is greater for CO_2 than N_2 at the same concentration levels [31]. In this work, the H_2 concentration was varied from 30 to 100%, the temperature was varied from 160 to 200 °C, and the operating pressure ranged from atmospheric to 200 kPa.

Experimental

All experiments were performed using a modified Hydrogenics fuel cell test stand. Several TPS and PBI-based MEAs were purchased from Advent Technologies with active cell areas of 45.2 cm^2 [15]. BASF has stopped manufacturing the Celtec series MEAs, but has licensed their technology to Advent Technologies who manufactured the MEAs used in our experiments. The MEAs purchased were the Celtec P1100W and Advent's TPS series MEA, with total precious metal loadings of 1.7 and 3.0 mg/cm^2, respectively. The manufacturer did not provide the specific amount of precious metal loading on the anode/cathode, nor the catalyst composition. The MEAs were placed in a test cell, shown in Fig. 1, comprised of two graphite plates with flow channels cut in a quad serpentine

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