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Research article

Use of wastewater treatment plant biogas for the operation of Solid Oxide Fuel Cells (SOFCs)

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ABSTRACT

Solid Oxide Fuel Cells (SOFCs) perform well on light hydrocarbon fuels, and the use of biogas derived from the anaerobic digestion (AD) of municipal wastewater sludges could provide an opportunity for the CH_4 produced to be used as a renewable fuel. Greenhouse gas (GHG), NO_x, SO_x, and hydrocarbon pollutant emissions would also be reduced. In this study, SOFCs were operated on AD derived biogas. Initially, different H_2 dilutions were tested (N_2, Ar, CO_2) to examine the performance of tubular SOFCs. With inert gases as diluents, a decrease in cell performance was observed, however, the use of CO_2 led to a higher decrease in performance as it promoted the reverse water-gas shift (WGS) reaction, reducing the H_2 partial pressure in the gas mixture. A model was developed to predict system efficiency and GHG emissions. A higher electrical system efficiency was noted for a steam:carbon ratio of 2 compared to 1 due to the increased H_2 partial pressure in the reformate resulting from higher H_2O concentration. Reductions in GHG emissions were estimated at 2400 tonnes CO_2, 60 kg CH_4 and 18 kg N_2O. SOFCs were also tested using a simulated biogas reformate mixture (66.7% H_2, 16.1% CO, 16.5% CO_2, 0.7% N_2, humidified to 2.3 or 20 mol% H_2O). Higher humidification yielded better performance as the WGS reaction produced more H_2 with additional H_2O. It was concluded that AD-derived biogas, when cleaned to remove H_2S, Si compounds, halides and other contaminants, could be reformed to provide a clean, renewable fuel for SOFCs.

1. Introduction

The on-site, cogeneration of electrical power and heat can significantly reduce the cost of wastewater treatment facility operations yielding a process that is more economical for smaller Canadian municipalities. However, at current North American electrical prices, waste-to-energy projects are only economical using conventional engine or turbine technologies for generating capacities greater than ~200 kW because high maintenance costs must be offset by revenue from the power generated (Wheeldon et al., 2007). Fuel cells can beneficially produce more electric energy from one unit of input energy than conventional electric generation systems (Galvagno et al., 2013; Bocci et al., 2014). They require lower maintenance and are inherently modular such that systems can be configured to a number of different applications depending on the required capacity allowing for more flexibility in the scaling of these systems. Anaerobic digestion-derived biogas produced at wastewater treatment plants (WWTPs) can be used as a renewable fuel for power systems that use high temperature fuel cells such as molten carbonate fuel cell (MCFC) and solid oxide fuel cell SOFC. Typically, biogas from AD at a WWTP is comprised of approximately 50–80% CH_4 and 30–50% CO_2 with trace concentrations of H_2S, O_2, N_2, halogenated hydrocarbons, volatile organic compounds (VOCs), NH_3 and siloxane compounds (Wheeldon et al., 2007; Farhad et al., 2010; Alves et al., 2013; Galvagno et al., 2013; Bocci et al., 2014). High temperature fuel cell systems are better suited for integrated internal reforming and tolerance to biogas contaminants (Alves et al., 2013; Galvagno et al., 2013; Bocci et al., 2014), while maintaining electrical efficiencies near 50% (Trendewicz and Braun, 2013; Bocci et al., 2014).

By utilizing the exhaust heat from the high temperature fuel cell stack to convert methane-rich biogas to a hydrogen-rich gas mixture, which can directly generate electricity in the fuel cell

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system, a significant reduction in GHG emissions (Alves et al., 2013), and improvements in air quality can be realized The use of biogas as the fuel for fuel cell power systems would also reduce the need for flaring which generates compounds such as NO_x, SO_x, and hydrocarbon pollutants in the combustion process. Wheeldon et al. (2007) modelled a SOFC system operating on AD biogas from various wastewater treatment plants in Ontario and noted that if all the wastewater sludges in Ontario were processed through AD, the biogas produced could theoretically provide approximately 1.51 GWh/d and reduce carbon dioxide emissions by 432 tonnes/d based on displacing fossil-fueled conventional electrical power generation capacity in use in Ontario at that time.

To date, demonstrations of biogas-fueled SOFC systems are limited to small-scale proof of concept efforts. Sulzer-Hexis in Switzerland demonstrated 5500 h of operation of a 1 kW SOFC on fermentation gas, for which a maximum electrical efficiency of 33% was reported Jenne (2003). Omosun et al. (2004) modelled system efficiencies and associated costs of a hypothetical biomass-fueled SOFC system. They also compared using hot-gas conditioning versus a cold gas conditioning sub-system and noted that the hot-gas conditioning system was more efficient but required a higher capital cost. Van Herle et al. (2004) modelled a 100 kW SOFC system fueled by wastewater treatment plant AD-derived biogas with an external reformer and reported that steam reformation of the biogas yielded a net electrical efficiency (49%). In a case study of the AD system at the Kingston, Ontario Ravensview WWTP, modelling efforts by Wheeldon et al. (2007) predicted 117 kW of electrical output and 132 kW of net heat output, with a combined heat and power efficiency of 55%. In these simulations (Van Herle et al., 2004; Wheeldon et al., 2007), H_2S removal using activated carbon was included, but gas conditioning components for other impurities and biogas compositional variability were not considered. Papadias et al. (2012) conducted a detailed analysis of landfill and anaerobic digester biogas impurities, along with a sensitivity analysis of electricity costs of a fuel cell system incorporating appropriate biogas cleaning. Trendewicz and Braun (2013) assessed the performance and life cycle costs of biogas-fueled SOFC systems for combined heat and power (CHP) applications at WWTPs with electrical power capacities of 300 kWe–6 MWe. The SOFC system model included anode gas recirculation, a biogas pretreatment system and a waste heat recovery unit. Their model predicted a net electrical efficiency of 51.6% LHV and net CHP efficiency of 87.5% LHV. At an estimated 0.05–0.08 $/kWh, they concluded that the cost of SOFC-based electricity was competitive with that of grid pricing (Trendewicz and Braun, 2013). In another study, Tjaden et al. (2014) investigated the thermodynamic performance of a small-scale (25 kWe) SOFC model fueled with biogas from various feedstocks. Their system analysis, which modelled steam reforming of biogas and included a comprehensive electrochemical SOFC stack model, yielded an electric efficiency of 56.55% LHV (Tjaden et al., 2014).

In this paper, an experimental study of the performance of an SOFC operated on AD-derived biogas is reported. The effect of biogas variability on cell performance was investigated using H_2 partial pressures of 65% and 25% and 10% of total pressure. Various diluents were studied including inert gases (N_2, Ar) and CO_2. The feed gas was humidified with 2.3 mol% steam. An empirical model which predicted the variation in cell performance with H_2 concentration was fit to these data. This empirical model was then incorporated into a SOFC system simulation that was developed using the process modelling software package UniSIM™. This process model predicted the overall performance of a SOFC system fueled with AD biogas which was converted to a H_2-rich reformate by steam reforming. In the next phase of the experimental study, a simulated biogas dry reformate mixture was prepared with the following composition: 66.7% H_2, 16.1% CO, 16.5% CO_2 and 0.7% N_2. This composition is based on a Gibbs Energy equilibrium calculation that predicted the equilibrium composition resulting from the steam reforming reaction at typical conditions. This method of using premixed simulated reformate for testing SOFC performance when operated using steam reformer product gas has been frequently used and has the benefit of eliminating variability that can arise due to reformer control stability (Hedström et al., 2009; Tanaka et al., 2011). Simulated reformate based on this gas mixture was fed to the lab scale SOFC stack at humidification levels of 2.3 and 20 mol% H_2O. The two levels of humidification reflect the difference of drying the reformate before feeding to the cell versus using the reformer product directly including the unreacted excess steam that would be present for a steam to carbon (S/C) ratio of 3.

2. Materials and methods

All experiments were conducted using commercially available tubular anode-supported SOFCs with YSZ electrolyte, a Ni-YSZ anode and Sr-doped $LaMnO_3$ YSZ cathode. The cells used in testing had an active area of approximately 135 cm^2. The cells were mounted in a test stand modified to accommodate mixed gas fuels and the addition of H_2O vapour to the system. The test stand was maintained at 800 °C and 1 atm for all tests conducted, and fuel flow rates were set to maintain approximately 80% fuel utilization.

All polarization curves were collected of a current density range from 0 to 500 mA/cm^2, or to a minimum voltage of 0.25 V, whichever occurred first. Below current densities of 100 mA/cm^2, the cells were unable to maintain stable voltages for the specified air and fuel utilization, as such, the flow rate was held constant such that the effective air and fuel utilization was less that that above 100 mA/cm^2. This sensitivity to flow was likely due to the stability of the mass flow controllers at fuel flow rates less than 5% of full scale. The fuel flow rate was controlled to maintain 80% fuel utilization at current densities greater than 100 mA/cm^2

2.1. Testing of anode supported tubular Solid Oxide Fuel Cells on dilute hydrogen gas

Dilute H_2 tests were conducted using three different diluents: N_2, Ar and CO_2. It was assumed that N_2 and Ar would be completely chemically inert in the SOFC and that CO_2 would react via the reverse water-gas shift (WGS) reaction (Equation (1)) to produce CO that could potentially affect the performance of the cell during the steam reforming reaction (Equation (2)).

$$CO + H_2O \rightleftarrows CO_2 + H_2 \tag{1}$$

$$CH_4 + H_2O \rightarrow CO + 3H_2 \tag{2}$$

Diluent tests were conducted at 25% and 65% H_2, 2.3% H_2O (as vapour), with the remainder the diluent gas. The H_2 partial pressures were selected to simulate those that would be expected as a result of variations in AD-derived biogas conditioning leading to fluctuations in H_2 partial pressures in the fuel gas mixture. Polarization curves were recorded at current densities ranging between 0 and 500 mA/cm^2, or until a minimum voltage of 0.25 V was reached. 0.25 V was the setting at which the SOFC test stand entered its E-Stop mode. Experiments were conducted in duplicate.

2.2. System efficiency and greenhouse gas emissions modelling

A simulation was developed in UniSIM to model a SOFC system, estimating output gas composition (and estimate GHG emissions), stack efficiency and stack performance. The system simulation was