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Fuel cell performance assessment for closed-loop renewable energy systems

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ABSTRACT

Fuel cells and electrolysis are promising candidates for future energy production from renewable energy sources. Usually, polymer electrolyte fuel cell systems run on hydrogen and air, while the most of electrolysis systems vent out oxygen as unused by-product. Replacing air with pure oxygen, fuel cell electrochemical performance, durability and system efficiency can be significantly increased with a further overall system simplification and increased reliability. This work, which represents the initial step for pure H_2/O_2 polymer electrolyte fuel cell operation in closed-loop systems, focuses on performance validation of a single cell operating with pure H_2/O_2 under different relative humidity (RH) levels, reactants stoichiometry conditions and temperature. As a result of this study, the most convenient and appropriate operative conditions for a polymer electrolyte fuel cell stack integrated in a closed loop system were selected.

1. Introduction

A typical polymer electrolyte fuel cell (PEFC) combines hydrogen from a tank and oxygen from the air to produce electricity, with water and heat like its byproducts [1]. A regenerative fuel cell also operates the reverse cycle, using the excess electric power to split water into hydrogen and oxygen, which are stored and fed back to the fuel cell to produce peak power when it needs [2]. In the case of electrolysis, the decision to store oxygen or vent it into the atmosphere strongly depends on the application. Closed-loop systems are designed to recirculate the reactants within the fuel cell, without an external purge. This means that the excess hydrogen at the exit of the anode (or oxygen at the cathode) is fed back to the anode (or cathode) inlet, after the removal of liquid phase water within a tank. The line pressure, after the reactants have been partially consumed by the reaction, is restored by means of a pressure regulator supplying hydrogen (at the anode) or oxygen (at the cathode). A small twin-head reciprocating compressor (one head for each gas) is dedicated to the recirculation of the reactants, the supply of reactants to the fuel cell inlet and water removal from exhausts. Being the compressor placed at the fuel cell outlet, the volume to be recirculated at the anode is usually within the 10%–20% of the stoichiometric flow for the anode, or within 50%–100% at the cathode, respectively. The by-product of the reaction, which is only pure water for hydrogen–oxygen fed fuel cells, is temporarily stored in the phase-separation tanks (one tank for each gas) and periodically purged to another tank. When the fuel cell is not operative, the water stored in this latter tank is then used by an electrolyzer to produce hydrogen and oxygen, which are stored back in the pressurized vessels to feed again the fuel cell. Hence, these systems use the stored oxygen, produced from electrolysis process and they are totally hermetic to the environment, without any mass exchange with the environment. This aspect can be decisive for the applications whereas there is the need to separate the environment from the system (such as submarines, space applications and harsh environments) [3]. The main space agencies are investigating the potential of the fuel cell technology in different scenarios, mainly for space exploration. It seems that fuel cell can be suitable [4], in the medium-term, for advanced robotic exploration missions and, in the longer term (>2030), for possible human explorations of the Moon and Mars. Fuel cell technology was already used before [5–11] as power supply system for space vehicles; this technology can be also used in the life support systems and in-situ resource utilizations systems. However, the operation conditions of a closed-loop system are generally very far from the standard conditions of a PEFC [12,13]. Pure oxygen fuel cell operation results in a higher cell voltage compared to air operation [1,2]. The gain, due to a higher partial pressure of oxygen, is usually higher than that solely predicted by the Nernst equation, particularly due to the higher diffusion rates of pure oxygen if compared to the oxygen/nitrogen mixture. In addition, a fuel cell operating with air requires fans or compressors to feed air through the fuel cell, resulting in additional parasitic

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Fig. 1. Single cell and laboratory connection.

losses, meanwhile passive air supply based on natural convection may be only used for very low-power applications usually featured from a poor performance. In order to best identify the criticalities, an experimental test campaign has been conducted to finalize the system configuration. So, a standard single cell using commercial components has been connected to a test station and tested in different conditions of *T*, *P*, RH with O_2 as an oxidant. This should lead to identify the critical aspects of the technology to be overcome in the future development of the fuel cell technologies. The results of these experiments helped the successive design of the fuel cell stack, the fuel cell size developed to provide a nominal power of 300 W with a single cell voltage of 0.65–0.8 V.

2. Experimental

A commercial MEA, purchased from IRD fuel cells, was used for this investigation. It was selected, among several commercial MEAs, after a successful testing in H_2/O_2 conditions. IRD-FC MEA is a catalyst coated membrane (CCM) on the reinforced Nafion membrane XL-100. Pt loadings of 0.6 mg/cm^2 and 0.3 mg/cm^2, for cathode and anode respectively, were used and a carbon felt based on a Gas Diffusion Layer (GDL) Sigracet24BC (SGL Group) was applied to the CCM.

3. Electrochemical characterization

MEAs were tested in a 25 cm^2 single cell. The channel flow design is a combination between serpentine and parallel channel configurations and it is based on the same design of the fuel cell stack to be developed. Single cell plates were realized by machining graphite (XM9612 graphite grade, SGL Carbon) with a thickness of 10 mm and channel dimensions of 1.2 × 1.0 mm. Current collectors were realized in copper and protected against oxidation interposing a flexible graphite sheet between the copper and graphite plates. Single cell tests were performed by a PC-controlled fuel cell test station. A bubble gas humidifying system was used to manage the gases relative humidity (RH) levels varying the humidifier bottles temperature. In Fig. 1, an image of the single cell and the connection to the test station was reported.

Moreover, Fig. 2 shows a scheme of the Balance of Plant implemented for the experimental tests.

The reference working conditions of the cell were based on the following standard conditions of a PEFC: $T_{cell} = 80$ °C, $P_{H2/O2} = 3$ bar, H_2/O_2 stoichiometry = variable, and RH = 100%. To simulate a closed loop system and investigate the most favorable operating conditions, single cell *I–V* polarization curves were carried out at $P_{H2}/P_{O2} = 3.0/3.0$ with a fixed stoichiometry for fuel and oxidant.

Table 1. Performed test conditions.

Test	*T* (°C)	RH (%)	*P* (bar)	H_2/O_2 (stoichiometry)
1	80	100	3	1.1/1.1
2	80	100	3	1.2/1.2
3	80	100	3	1.2/1.5
4	80	100	3	1.5/1.5
5	80	100	3	1.2/2.0
6	60	100	3	1.2/1.5
7	80	75	3	1.2/1.5
8	60	75	3	1.2/1.5
9	80	50	3	1.2/1.5
10	60	50	3	1.2/1.5

Several conditions were investigated varying the cell temperature from 60 to 80 °C, the reactants stoichiometry between 1.1 and 2.0 and the RH between 50% and 100%. In Table 1, all the performed tests are summarized.

The cell resistance (R_{cell}) was measured with an Agilent milliohm meter by a static method at a frequency of 1 kHz.

4. Results and discussion

This study was carried out to verify the possible application of PEFC in a closed-loop system and to individuate the optimal stack operative conditions. In fact, testing conditions were far from those of a standard single cell characterization reference, whereas the application is very different from the typical application of a PEFC system. While useful for qualitative comparisons between cells, this data are generally not very representative of a stack performance, which needs further developments, especially on the engineering side [13–18].

When operating in a closed-loop configuration, without any external humidification system, the RH at the stack inlet can significantly vary depending from the power output, from 100% (stand-by or low power operation with a non-zero minimum recirculation flow) to very low values (i.e. 17% at 1,2 stoich. or 33% at 1,5 stoich.). This can be calculated considering the fuel cell mass and heat capacity, heat losses along the gas pipeline and the operation temperature. The gas phase at the anode and cathode outlet can be assumed to have the fuel cell temperature, because the fuel cell is maintained at a fixed temperature, either by means of electrical heaters (in case of a single cell) or an external cooling circuit (in case of a stack) and the mass of the fuel cell is much greater than that of the gas inside the cell. The gas at the fuel cell outlet has the same temperature of the fuel cell and the gas is fully saturated by water (with a significant liquid phase). The gas coming from the storage vessel has the ambient temperature and its RH can be assumed equal to zero. The temperature and the RH of the gas resulting from the mixing of the recirculated and the fresh gas from the storage vessel can be calculated from mass and energy balance:

$$\dot{m}_1(\mathrm{g}) \cdot c_\mathrm{p}(\mathrm{g}) \cdot (1+\alpha) \cdot (T_1 + T_2) + \dot{m}_2(v) \cdot c_p(w) \cdot T_2 = ((\dot{m}_1(\mathrm{g}) \cdot c_p(\mathrm{g}) \cdot (1+\alpha) + \dot{m}_2(\mathrm{v}) \cdot c_p(\mathrm{w})) \cdot T_3 - \dot{m}_3(\mathrm{l}) \cdot r$$

where, $\dot{m}_i(\mathrm{g})$ is the mass flow of the gas. When $i = 1$, the gas comes from the storage vessel; when $i = 2$, the gas comes from the recirculation loop; when $i = 3$, the gas is the sum of gases coming from storage vessel and recirculation loop at the fuel cells inlet (kg/s); $c_\mathrm{p}(x)$ is the specific heat of the gas ($x = \mathrm{g}$) or the water ($x = \mathrm{w}$) (J/kg/K); α is the difference between the actual gas flow and the stoichiometric value divided by the stoichiometric value and it represents the gas utilization factor. $\alpha = 0$ indicates an actual flow equal to the stoichiometric flow, while 0.5 indicates an actual flow which is 50% higher than the stoichiometric one (adimensional); *T* is the temperature (°C) of the gas coming from the