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Fuel cell development for space applications: fuel cell system in a closed environment

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Abstract

Polymer electrolyte fuel cell (PEFC) systems targeting applications to transfer vehicles for short-term missions and larger spacecraft in the future are being developed at NASDA.

First, we designed and manufactured a system with a 100 W class fuel cell. Through our tests, we found that a humidifier is not necessary when using pure hydrogen and oxygen supplied from opposite directions.

In a closed simulated environment, the tests could also demonstrate the stable operation of the fuel cell system where the oxygen was recycled and the hydrogen stream was dead-ended.

Keywords: Polymer electrolyte fuel cell; Humidification; Dehydrator; Gas recycle; Aerospace; Closed environment

1. Introduction

Polymer electrolyte fuel cells (PEFCs) have recently received a great deal of attention due to their applicability to electric vehicles and stationary generators. This attention has enhanced their research and development.

In the field of space engineering, fuel cells have been applied to spacecraft like GEMINI and other manned operations since the 1960s. Since a fuel cell produces water as a byproduct of generating electricity, it is very attractive for manned operations in a closed environment. Furthermore, the fuel cell generates energy using hydrogen and oxygen, both of which have high energy density. Due to this high potential, fuel cells are still advantageous for application to large spacecraft with short-term missions [1–4].

Today, PEFCs are beginning to receive a considerable amount of attention again for aerospace applications. One example is the Stratospheric Platform Project [5–8]. A lightweight, long-duration generating system is now required to maintain the altitude and position of an aircraft or airships in the stratosphere. For space applications, a variety of missions are proposed that will need the fuel cell system [9–12]. One example is a transfer vehicle in low-Earth orbit.

The National Space Development Agency of Japan (NASDA) is now developing the H-II transfer vehicle (HTV) to supply materials to the International Space Station (ISS). The HTV is designed using lithium primary batteries combined with solar cells and a lithium secondary battery. The total mass of this craft is calculated to be 15 t, with a mission capability of 6 t, while the total mass of the electrical power units is about 1 t. In the future, a lighter weight, high-output electrical power system for advanced designs of the transfer vehicles will need to be developed.

The fuel cell for space applications has special system requirements, operational conditions, and designs due to its isolated low gravitational environment, which is much different from that for ground applications.

Air can be supplied to the fuel cell as cathode gas for ground applications operated in the atmosphere, and hydrogen (or reformed gas), as anode gas. As is commonly known, the fuel cell reaction produces water with electricity. The water must be vaporized by compressed air, because water remaining on the path in the separator may block the stream of reactant air. In contrast, the proton conduction in

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the membrane requires water, and the air is humidified before being supplied to the fuel cell. Thus, the fuel cell for ground applications requires humidification of the supplied gas and dehydration of the separator in order to balance the complicated humidity conditions inside the fuel cell.

In order to simplify the system, many efforts have been made to eliminate the humidifier even for ground applications. One example is the automobile fuel cell application [13]. Using a thin membrane as a proton conductor, engineers tried to minimize the water content applied to the proton conduction inside the membrane. In this case, they successfully eliminated the humidifier on the cathode side but still installed the humidifier on the anode side. For ground applications, air will be used for the cathode gas. Air contains 79% nitrogen which is inert to the fuel cell reaction. Therefore, a closed system can never be realized since the performance of the fuel cell declined due to the contamination of nitrogen. Thus, some sort of exhaust is always needed, making it difficult to dead-end the outlet port. In [13], cathode and anode gas are exhausted to the outside through the control valve. Furthermore, the exhaust also dries up the membrane due to the flow of the inert nitrogen gas.

If we use fuel cells in space, it is very important to prepare the simplest system. Furthermore, since the spacecraft is very isolated in Earth orbit, all reactant materials must be carried inside the spacecraft. In order to minimize the weight, pure anode and cathode materials must be used and should be consumed completely. Thus, we assumed the following system and proceeded with our research.

(1) Pure hydrogen and oxygen are used as reactants.
(2) Reactant gases are not humidified before being supplied to the fuel cell.
(3) As much of the reactants must be consumed as possible, and the amount of exhausted material should be minimal.
(4) The water produced must be collected.

If the fuel cell can be operated using only produced water, the best solution to concept 2 is realized. We decided to demonstrate the fuel cell system without a humidifier. The cathode gas (in this case, oxygen) must be exhausted from the fuel cell to prevent blocking the gas-flow through the separators. In order to realize concepts 3 and 4, we decided to circulate the flow of oxygen and return oxygen to the fuel cell again after dehydration. In this way, thermal control of the dehydrator enables us to control the humidity of the oxygen returned to the fuel cell. Furthermore, we tried to dead-end the hydrogen outlet port. These trials might realize the simplest PEFC system.

The first practical application of the fuel cell in space was for the GEMINI project in the 1960s. The first fuel cell to travel in space was the PEFC. The membrane for the fuel cell was polystyrene, not perfluoro-membrane, and high-power supply, commonly achievable today, was not realized. When the APOLLO and Space Shuttle missions started, PEFC was replaced by an alkaline fuel cell system. After several decades, the research and development of PEFC is again gathering attention due to its applicability to ground facilities. However, little data about the operation with high-power generation in a closed environment has been reported [5–8]. These situations encouraged us to develop a fuel cell system with the above concept using the latest technology such as reinforced perfluoro-membrane and a graphite separator.

Based on the above concepts, we prepared a fuel cell that can generate 100 W. Using this fuel cell, we tried to find a way to operate the fuel cell without humidification. Our research showed that the counter flow of pure hydrogen and oxygen enabled realization of a fuel cell system without humidification even though the outlet port is opened to the atmosphere as in ground applications. Furthermore, this performance was maintained without flooding even when the outlet port of hydrogen was dead-ended. By using pure oxygen as the cathode gas, we could eliminate the humidifier without degradation of the fuel cell performance.

We next tried to find how to circulate the oxygen in order to understand the operational conditions of the fuel cell system for a closed environment. We prepared a dehydrator and gas circulation pump and combined them together to demonstrate a fuel cell system for a closed environment. Though the PEFC system has a long history, very little data has ever been reported concerning the operational conditions as a closed system.

Thus, we successfully realized a fuel cell system where the supplied gas can be totally consumed by the fuel cell reaction. This paper will discuss the details of our research.

2. Experimental

2.1. Preparation of the fuel cell stack

For the oxidant and the fuel gas-flow, we prepared a separator with a serpentine gas-flow channel on both sides of a carbon plate where the surface was coated with graphite to prevent gas leakage. The serpentine channel was 1.60 mm wide and 1.25 mm deep. The membrane electrode assembly (MEA) was sandwiched between the separators. The effective surface area of the Pt catalyst layer on the MEA was 81 cm^2 (= 9 cm × 9 cm). The MEA was PRIMEA5510® produced by GORETECHS ltd., and the 30-μm-thick membrane used for the MEA was GOERE-SELECT®.

We used four pairs of cells to prepare the 100 W stack shown in Fig. 1. In the stack, two cells shared a separator with water coolant to keep the fuel cell temperature between 60 and 65 °C.

2.2. Activation and operation of the fuel cell without humidification

Before we started up the fuel cell, we increased the temperature of the fuel cell stack to 40 °C. We then started to op-

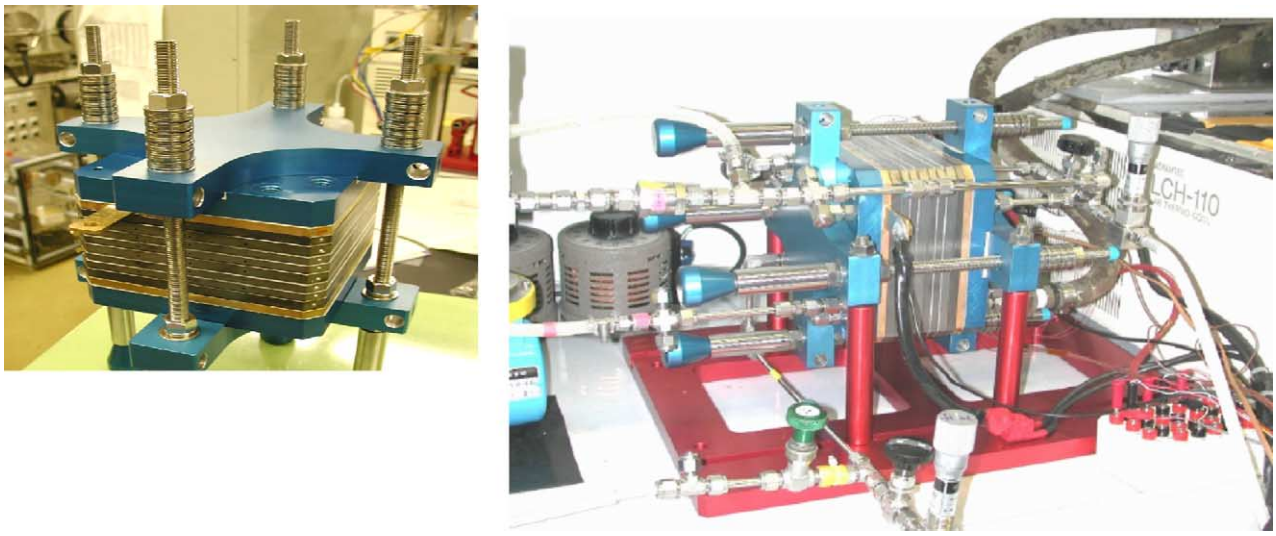

Fig. 1. Photograph of 100 W class fuel cell stack.

erate the fuel cell with a 1.00 A (1.23×10^{-2} A/cm^2) current flow for 1 min, and with 3.00 A (3.70×10^{-2} A/cm^2) for one more another minute. After this treatment, we operated the fuel cell at 20.0 A (2.50×10^{-1} A/cm^2) for 10 min, and then increased the current in steps of 10.0 A (1.23×10^{-1} A/cm^2). At each step, the operation continued for 10 min to activate the fuel cell. The temperature of the fuel cell increased with current. We controlled the circulation of the coolant (water) so that it took more than 30 min to raise the temperature to 60 °C. When we activated the fuel cell, the consumption of hydrogen and oxygen was limited to 30%. The hydrogen used for our tests was 99.99999% (G1 grade), and the oxygen, 99.999% (G2 grade).

2.3. Concept of the fuel cell system

Fig. 2 illustrates the concept of proton and water movement inside the fuel cell. Protons move through the MEA or proton-conducting membrane via water by the Grothuss mechanism. Due to this migration mechanism, water is essential for proton movement [14–16]. Furthermore, migrating protons carry water due to the electro-osmosis [17]. Due to these proton migration mechanisms, the anode usually lacks water.

In contrast, the fuel cell produces water on the cathode side through electrochemical reaction. The produced water is diffused by the concentration gradient inside the membrane.

Fig. 3 depicts a conventional fuel cell system for ground applications. Hydrogen is usually used as the anode gas and is supplied directly from a high-pressure vessel or after being reformed from hydrocarbon. Air is conventionally used as the cathode gas because of its practical applicability.

If the reformed gas is used as the anode reactant and air is used as the cathode gas, it is necessary to exhaust the gases continuously to maintain the fuel cell performance because component gases such as CO, CO_2, and N_2 degrade the fuel cell performance. In addition, these exhausted gases remove humidity from the fuel cell and tend to dry up the membrane, so we need to humidify the reactant gases before supplying them to the fuel cell stack.

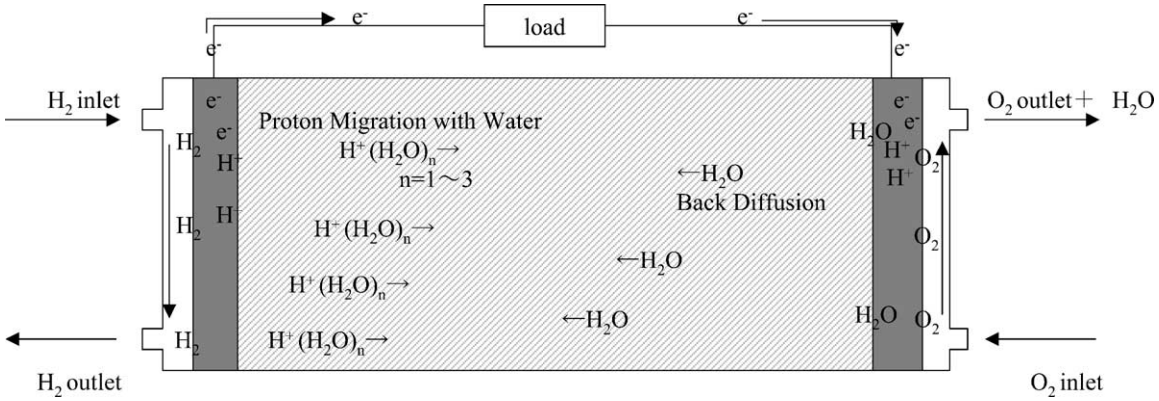

Fig. 2. Concept of proton and water movement inside the fuel cells.

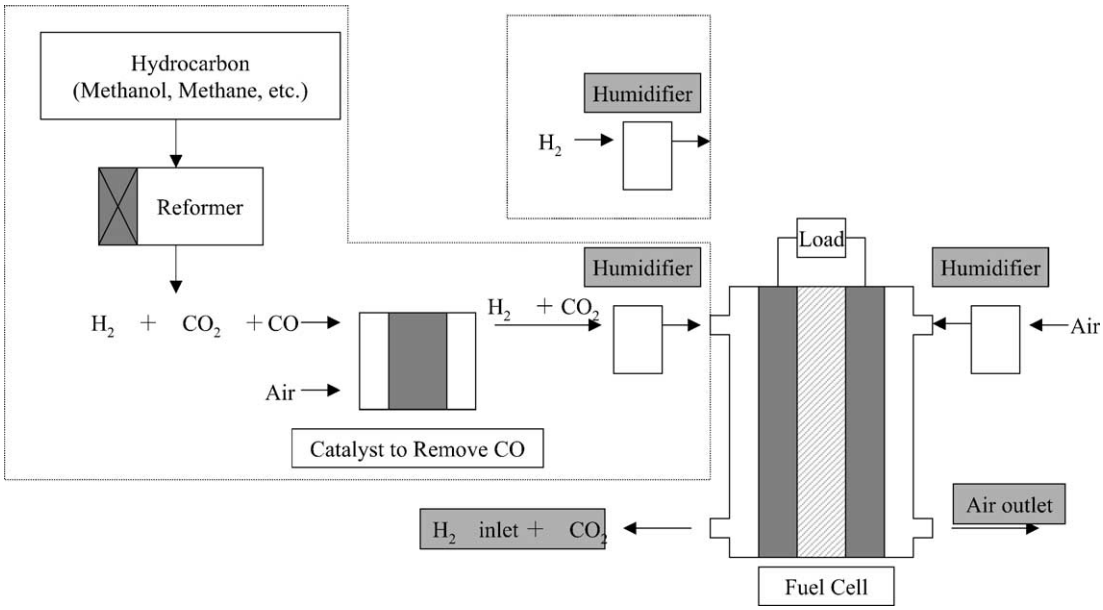

Fig. 3. Conventional fuel cell system for ground applications.

Fig. 4 depicts the fuel cell system we prepared for our applications. In contrast to the ground applications, we need to carry both anode and cathode gases inside the space vehicle, so we can only use pure hydrogen and oxygen. In this case, we used high-grade hydrogen (99.99999%) and oxygen (99.999%) when we simulated the closed environment conditions to minimize the degradation of cell performance.

In addition, we tried to eliminate the humidifier to simplify the system. One of the main objectives was to demonstrate the stable operation of the fuel cell using its own water product without extra humidification. We tried different directions of gas-flow to realize the operation.

Based on these concepts, we prepared a fuel cell system that combined a fuel cell stack, gas circulator, and dehydrator as shown in Fig. 5.

We will describe in detail how the dehydrator removes the humidity for the gas circulation. In the low gravity of orbit, we cannot separate water from humid gas using water's weight, even if we chill the humid gas and increase the density of water. In this case, a wick was installed inside the dehydrator. Gas passing through the dehydrator is cooled to 5 °C and the humidity in the gas is absorbed into the wick as water to produce the low humidity gas for circulation.

3. Results and discussions

3.1. Performance without humidification

Fig. 6 illustrates the power–current relationship of a four-cell stack. In this case, the gas was not recycled. The excess gas for the fuel cell reaction was exhausted to the air, and the reactant gas was not humidified before being supplied to fuel cell stack. As shown in this figure, 140 W at 50 A and 250 W at 100 A were generated.

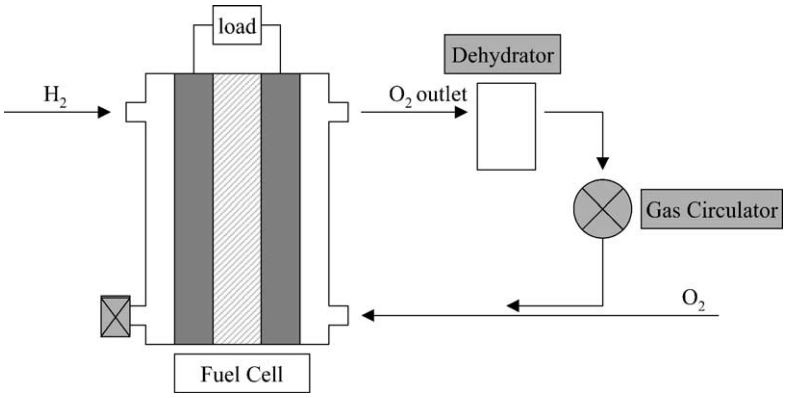

Fig. 4. Conceptual design of our fuel cell system for space applications.

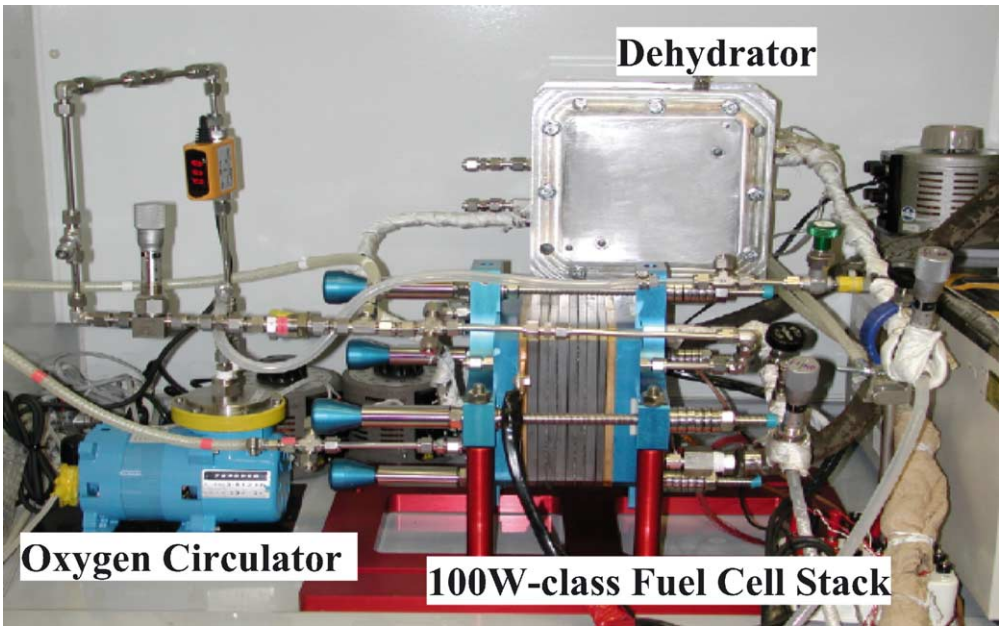

Fig. 5. Fuel cell system combining four-cell stack, gas circulator, and dehydrator.

Fig. 7 presents the relationship between the cell voltage and current. The voltage differences among the four cells were very small, and the performance among them was uniform.

The important point for this operation was the direction of gas-flow. Fig. 8 shows how performance differs when the gas stream direction was changed. The concepts of "parallel" and "counter" are described in Fig. 9. "Parallel flow" means that both hydrogen and oxygen are supplied from the inlet port located on the same side of the fuel cell, while "counter flow" means that hydrogen and oxygen are supplied from inlet ports located on opposite sides of the fuel cell.

In operations without humidification, the fuel cell performance degraded immediately if the gases were supplied in the same direction across the MEA. The amount of water produced inside the fuel cell can be minimal in the upper part and higher in the lower part of the gas stream. This gradient accelerated the heterogeneous distribution of water in the cell.

In order to obtain stable fuel cell performance, water must be distributed homogeneously. We should mention that both anode and cathode gases flow parallel due to the design of the serpentine channel even in counter flow. However, when hydrogen and oxygen are supplied from the opposite sides of the fuel cell, the water produced on the cathode side is carried to the lower stream of oxygen and migrates to the an-

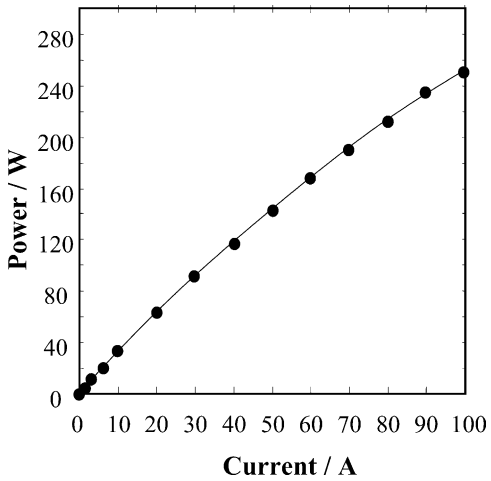

Fig. 6. Power–current relationship of four-cell stack. The fuel cell was operated at 65 °C without humidification. Both hydrogen and oxygen were exhausted.

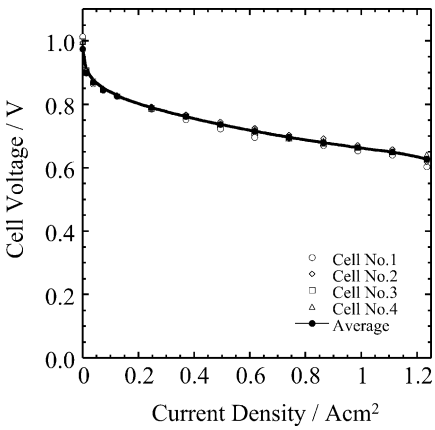

Fig. 7. Relationship between cell voltage and current of four-cell stack. The fuel cell was operated at 65 °C without humidification. Both hydrogen and oxygen were exhausted.

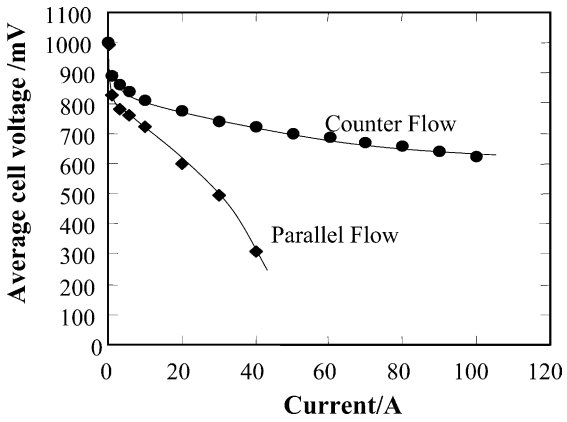

Fig. 8. Difference of performance with gas-flow direction changes. The fuel cell was operated at 65 °C.

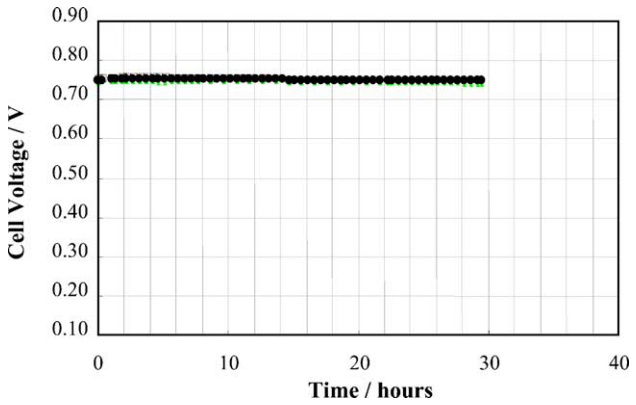

Fig. 10. Continuous operation of 100 W class fuel cell. The fuel cell was operated at 65 °C without humidification. Both hydrogen and oxygen were exhausted. The conversion of hydrogen and oxygen was 40 and 23%, respectively. All four-cell voltages are listed; (●) indicates the average voltage of four cells.

ode side. This migrating water humidifies the counter flow of dry hydrogen across the MEA and carries the humidity to its lower stream. Furthermore, the humidity carried by the hydrogen stream migrates through the MEA in the upper stream of the cathode gas where the membrane is rather dry. As a result, there is a favorable distribution of water inside the fuel cell. In this case, the gas-flow channel was serpentine, but we observed the same effect of gas-flow directions even in a comb-shape type [12].

Fig. 10 presents the results of continuous 30 h operation of a four-cell stack. Cathode and anode gases were fed from opposite directions without humidification. During the entire operation, each cell voltage was operated at 0.75 V for 30 A (0.37 A/cm^2), continuously, and we could successfully demonstrate stable fuel cell performance for hours without humidification.

As shown in Fig. 2, the conduction of protons inside the membrane requires water. In ground applications, the cathode gas is conventionally air, and about 79% of the cathode gas is inert nitrogen. Due to this inert gas, the humidity of the cathode gas remains low if water is not supplied through the humidifier. Furthermore, the inert gas must be purged continuously from the fuel cell, which also decreases the humidity. Therefore, air cannot be used as the oxidant in a closed system. However, when the cathode gas is pure oxygen, the cathode humidity can be increased by the change of oxygen utilization. The high humidity supported the performance of our fuel cell.

Through these tests, we successfully demonstrated that a fuel cell system could be operated without humidification of reactant gases by applying pure hydrogen and oxygen.

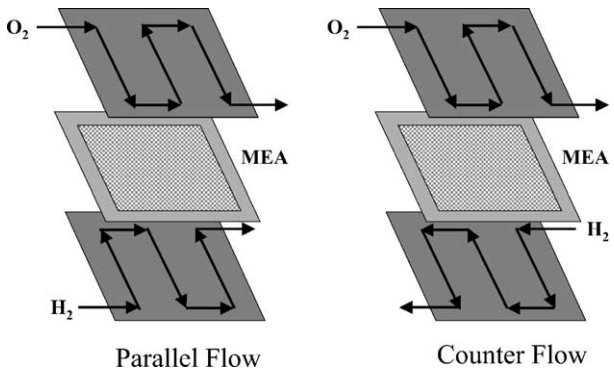

Fig. 9. Concept of gas-flow direction inside the fuel cell.

3.2. Simulation of closed environment

We also performed a simulation of closed gas conditions. All reactant must be carried inside the vehicle for space applications. Ideally, reactant gases must be fully consumed without being exhausted. This environmental restriction caused us to test the fuel cell performance under closed conditions in which we tried to circulate oxygen and close the hydrogen outlet port.

When oxygen is used, the produced water must be constantly removed from the catalyst layer of the MEA. We therefore installed a dehydrator along the gas recycling line together with a gas circulation pump. The dehydrator can remove water by condensation. The removed water was wicked up and applied to humidify the fuel cell by controlling the temperature of the dehydrator. Gas circulation also plays an important role in inhibiting the flooding on the cathode side. Fig. 11 illustrates the relationship between the output voltage of the fuel cell and the oxygen circulation rate (circulated mass / theoretical mass). The appropriate circulation rate resulted in constant fuel cell performance.

Ideally, if the outlet of hydrogen at the anode were to be closed, the simplest system could be realized. For this purpose, we must prevent the condensation and accumulation of water in hydrogen and subsequent flooding on the anode side.

Fig. 12 depicts the amount of water obtained from the fuel cell anode. We closed the hydrogen outlet port stepwise and

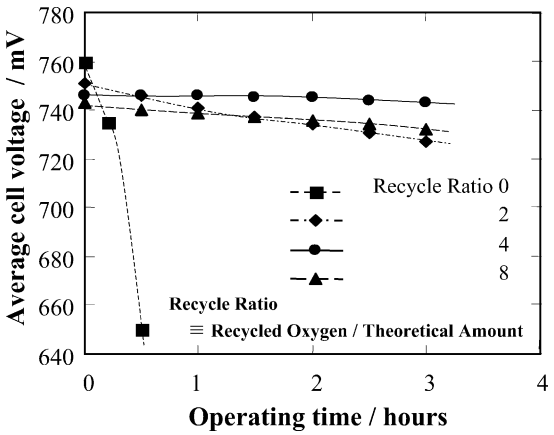

Fig. 11. The relationship between the output voltage of fuel cell and the circulation rate of oxygen.

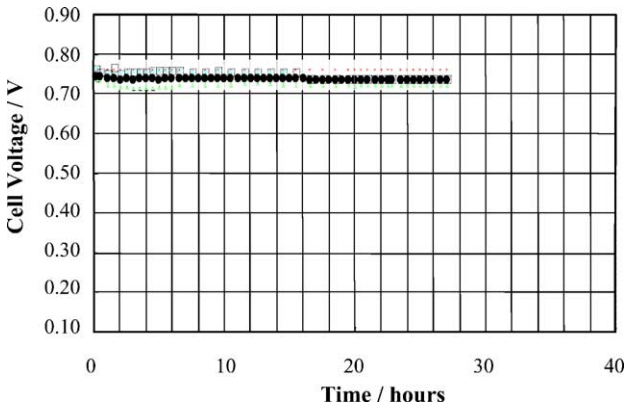

Fig. 13. Continuous operation of 100 W class fuel cell. The fuel cell was operated at 65 °C without humidification. Hydrogen outlet was closed, and oxygen was recycled with a recycle ratio of five. All four-cell voltages are listed; (●) indicates the average voltage of four cells.

allowed the water to condense. This figure indicates that the water obtained from the anode was linearly dependent on the amount of outlet gas. This linear line intersected the *x*-axis when the theoretical amount of hydrogen was consumed by the fuel cell reaction, indicating that water does not exist in liquid form on the anode side.

This result suggested that the closed hydrogen outlet did not cause accumulation of liquid water. We could expect an appropriate amount of water on the anode side where the electro-osmosis and back diffusion were well balanced. Based on these results, we tried to operate the fuel cell with closed gas systems.

Fig. 13 shows the result when a current load of 30 A (i.e., current density of 0.37 A/cm^2) was constantly applied to the fuel cell. In this case, as shown in Fig. 4, oxygen was recycled to remove the produced water and the hydrogen outlet was closed. We observed no degradation of performance of the stack during 28 h of operation.

Even though the hydrogen gas outlet is closed, there is still a hydrogen gas stream due to the consumption by the fuel cell reaction. This stream might be enough to carry the humidity in the lower direction of the oxygen stream, which achieved the proper performance of the fuel cell.

The purity of reactant gas is also very important for gas circulation or closed gas operation. When we used low purity gases, we observed degraded fuel cell performance. Impurities such as nitrogen inside the fuel cell might also decrease performance. Specifically, for continuous operation, we used 99.99999% pure hydrogen and 99.999% pure oxygen. Purging will need to be done at regular time intervals for future operation of a fuel cell in a closed environment.

Using this four-cell stack, we tested the performance by intermittent operation for over 100 h in total. Fig. 14 shows the whole operation of the stack including the results mentioned above. We observed stable performance throughout the measurement.

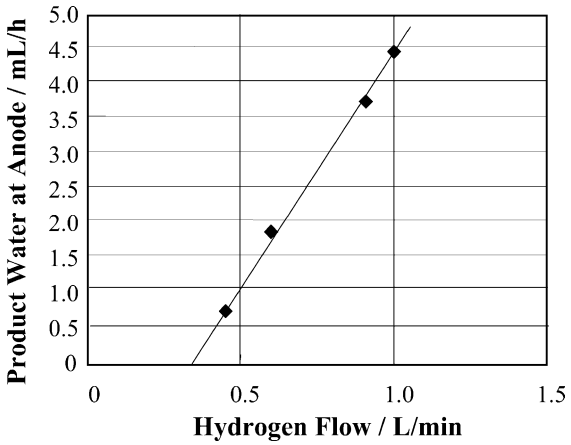

Fig. 12. Linear dependence of water content obtained from fuel cell anode.

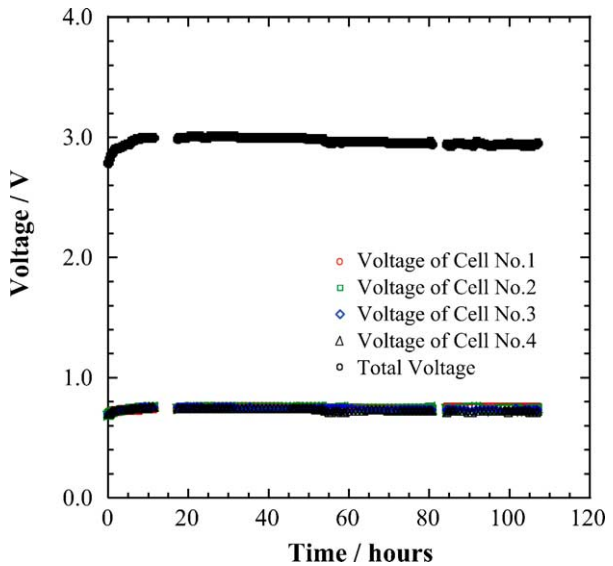

Fig. 14. Intermittent operation of 100 W class fuel cell system without humidification. The fuel cell was operated at 65 °C without humidification.

4. Conclusion

NASDA is now developing a PEFC system for future operation of transfer vehicles and larger spacecraft.

We tested the PEFC performance without humidification in the case of 100 W class stack for applications in a closed environment. Our experiments confirmed that fuel cells did not need a humidifier if they were operated with pure hydrogen and oxygen supplied from opposite directions. We also demonstrated closed gas operation to determine the applicability of the fuel cell system in a closed environment, where hydrogen gas was dead-ended and oxygen gas was circulated with produced water removed.

Results of constant operation for 30 h in a closed gas system and intermittent operation for more than 100 h provided information and guidance for future designs and operation in space applications.

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