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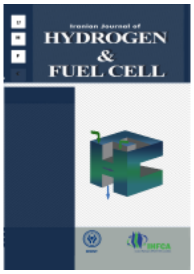

Experimental investigation of a solid oxide fuel cell stack using direct reforming natural gas

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Abstract

In this study, a solid oxide fuel cell (SOFC) stack has been successfully fabricated and tested by using direct natural gas. The main objective of this research was to achieve optimal long-term performance of the SOFC stack without carbon deposition by using low-cost natural gas as a fuel. The stack configuration was improved by a new interconnect design and made of cost-effective raw materials. In this respect, the stack showed maximum power of 31 W while 33 A current was applied at a flow rate of 1000 ccm for H_2 (as fuel) and oxygen. Then, humidified natural gas was employed as an internal reforming technique, which showed degradation of 1.4% after 24 h. Maximum obtained power was 32 W under 33 A current at a flow rate of 1000 ccm. After 48 h of operation, 34 W of power was achieved at the current of 38 A. Therefore, the power was increased from 32 to 34 W after 48 h of operation in upper current. Finally, a suitable SOFC stack made of cost effective materials and using direct natural gas under appropriate conditions was fabricated and developed in this research.

1. Introduction

A solid oxide fuel cell (SOFC) is one of the most prominent devices of energy conversion due to its fuel its fuel flexibility and high energy conversion efficiency [1, 2]. One of the main advantages of high temperature SOFCs are their flexibility with diverse fuels with provides the possibility of internal

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reforming of natural gas or any other fuel [3-5]. Although, one of the challenges in these systems is carbon deposition on the anode side. Therefore, direct oxidation of hydrocarbon fuels in SOFC's anodes have been widely investigated [6-9]. Anodes are made of Ni-based cermet that has high electrical conduction and is potent for hydrogen oxidation [10, 11]. Ni in the anode has a strong catalytic impact to break hydrocarbon bonds. This results in precipitation of coke on the anode's surface which quickly decreases the electrochemical performance of the cell [12-14]. Ni-YSZ can be used when a high amount of steam is used along with fuel to ensure minimization of carbon deposition and appropriate fuel conversion [7, 9].

In this research, the optimized steam to natural gas ratio was applied to improve performance of the stack in 48 h and diminish carbon deposition. Moreover, different flow rates were applied to understand its effect on stack performance and diminish the carbon deposition as much as possible.

2. Experimental

2.1.Materials

The stack used in this study was made of two cells. The green half-cell was prepared by tape casting the electrolyte and anode layers. For this purpose, 8YSZ and NiO-8YSZ (FCM, USA) powders were separately ball-milled with solvents, binder, and plastisizer for 48 h. The electrolyte slurry was cast on Mylar sheets. After drying this layer overnight, the anode slurry was then cast on dried electrolyte. Afterward, the obtained half-cell was pre-sintered at 1000°C for 2h and again sintered at 1450°C for 3h in air atmosphere. The sintered half-cell size was 10*10 cm^2. Then, the LSM (FCM, USA) powder was ball-milled with solvents and additives for 12 h. Finally, the cathode slurry was screen printed and sintered at 1200°C for 2 h. Fabricated cells were placed between interconnects then connections and sealing were applied. Silver paste was used to connect the cathode surface to the steel interconnects on the cathode side whilst nickel mesh and paste were applied to the anode side. Interconnects were made of steel 430 using a lathing machine. After the interconnects were machined and prepared, their surface was polished, degreased and washed with ethanol and acetone. Then, the surfaces were coated with $MnCoO_2$ using the wet powder spraying process. For this purpose, a slurry of $MnCoO_2$ was prepared by jar milling of the powder in ethanol media for 24h using PVB and toluene as binding agents with a weight ratio of $MnCoO_2$: binders : ethanol = 0.6 : 0.1 : 1.0. Afterward, the spray coating was applied four times on both sides of the samples in a horizontal and vertical direction [15].

Coated interconnects were heat treated at 450°C for 12 h in an atmosphere controlled furnace. Additionally, they were treated in a diluted H_2 (5%) atmosphere for 3.5 h until the temperature reached 850°C and further dwelled for 8 h, then they were cooled to room temperature in the furnace. For preparing of the stack, silver and nickel paste were applied on the anode side and cathode side of each cell, respectively. Afterward, the assembly was sandwiched between the interconnects and alumina felt was used to seal the stack.

2.2.Methods

The furnace was heated gradually to 800°C while the stack was in it. After reaching 800°C, N_2 gas was purged into the stack to reach a stable OCV. Then diluted H_2 (5%) was purged into the stack to reduce nickel oxide and reach optimal OCV. The flow rate was increased gradually to avoid any damage to the cell. After adjusting gas flow rate and purging conditions, voltammetry and chronopotentiometry tests were utilized to study the stack performance. SMART2 (WonATech Co., Ltd.) was used to adjust gas flow and Autolab (302N) was used to measure electrochemical performance. Linear sweep voltammetry (LSV) was applied in order to investigate the stack electrochemical performance and evaluate the stack performance. The voltammetry was done with a scan rate of 1A s^{-1}. The output power was also calculated by multiplying the measured voltage and applied current. After partial reduction of the anodes, the purging gas was switched to H_2 (5N) and continued until the anode was fully activated, this was revealed by reaching an open circuit voltage (OCV) of higher than 1.0 V per cell. In the following testing procedure, natural gas mixed with steam with the ratio of about 2:1 (Steam to Carbon) and was purged into the stack. The optimized steam to carbon (S/C) ratio was applied based on previous research [16].

3.Results and Discussion

Fig.s 1 and 2 show the performance of the SOFC stack in hydrogen and humidified natural gas, respectively, taken in the first 24 h of operation. After a while, OCV reaches 1.97 V with an applied current of 11 A. OCV then further increased to 1.996 V at the current of 14 A. This is equal to approximately 1 V for each cell, which is very close to the theoretical value. From the I-V measurement we can see that a maximum power of 30.72 W was obtained in this condition with an applied current of 30 A at 800°C as shown in Fig. 1.a. In addition, I-V measurements at higher currents showed that power increased to 37 W when the hydrogen flow increased to a rate of 2000 ccm, as shown in Fig. 1.b. Increasing the fuel flow rate led to a decrease in the polarization due to higher mass transfer which increased the voltage and power of the cell. It was also seen that the current density at maximum power shifted to higher values due to the presence of much more dissociated hydrogen.

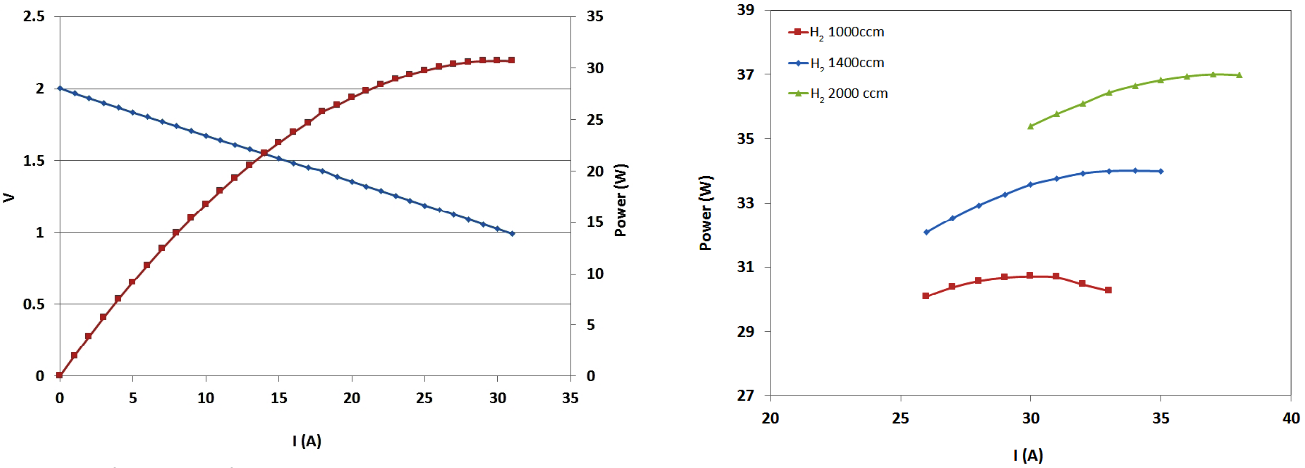

Fig. 1. Performance of the SOFC stack, a) I-V curve at 800°C in H_2 with flow rate of 1000 ccm and O_2 flow rate of 1000 ccm. b) Power performance under different H_2 flow rate at 800°C (area of the cells: 2×100 cm^2).

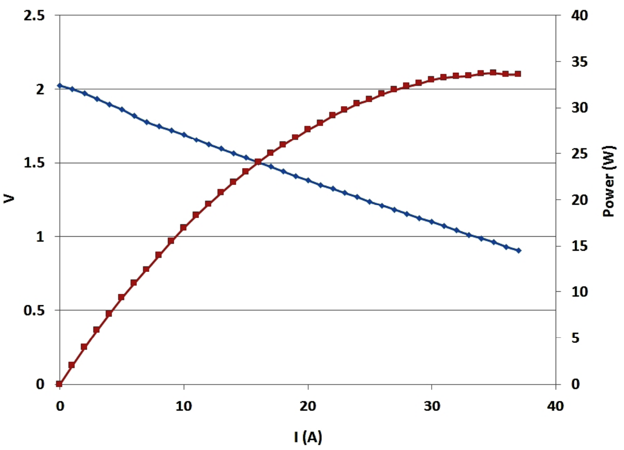

Fig. 2. Voltage and power versus current measured in humidified natural gas with flow rate of 1000 ccm and oxygen flow rate of 1000 ccm at 800°C (area of the cells: 2×100 cm^2).

Furthermore, a current of 22 A was applied to the stack and the fuel and O_2 flow rate were set to 1000 ccm in order to obtain a voltage of 1.298 V. After switching to humidified natural gas with a flow rate of 2000 ccm, the voltage increased to 1.38 V. In this condition, I-V measurement on the stack was conducted and a maximum power of 33.74 W and an OCV of 2.024 V were obtained as shown in Fig. 2.

The duration of the test was divided in two 24 h stages. In the first 24 h term, the applied current was 16 A, and it was operated under a natural gas and oxygen flow rate of 600 ccm at 800°C showing a voltage of 1.42 V. After the first day of operation, the voltage was still stable at 1.42 V. At this condition, I-V measurement was conducted, indicating a maximum power of 32.34 W under a current of 33 A as shown in Fig. 3. The power faded with respect to the cell before this 24 hour period. The main cause for this phenomenon is degradation of the nickel anode introduced by carburizing.

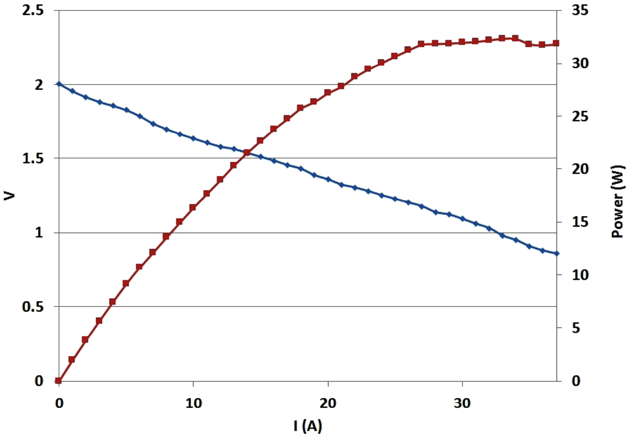

Fig. 3. Voltage and power versus current measured in humidified natural gas with flow rate of 1000 ccm and oxygen flow rate of 1000 ccm at 800°C after 24 h of operation in natural gas (area of the cells: 2×100 cm²).

For the second 24 h term, the natural gas and oxygen flow rates were set to 400 ccm, and a 14 A load was applied in order to obtain a voltage of 1.4 V. The I-V measurement for this condition is illustrated in Fig. 4. After the second stage of operation (48 h of operation), the I-V measurement displayed a voltage of 1.38 V which indicated a 1.4 % reduction in voltage as shown in Fig. 5. It should be noted that under a current of 38 A, the power reaches its highest value of 34.23 W, which is even higher in comparison to the first day of operation. The maximum power of the stack increased after 48 hours while the OCV dropped from 2.024 to 1.899 V. This behavior is caused by carburizing of the anode. This can be confirmed by increasing the current at the similar potential, which is observed in Fig.s. 2, 3 and 4. This shows the higher activity and improved electrical conduction of the cell. It should also be noted that in the early hours the power decreased from 33.74 to 32.34 W after 24 hours, and raised again to 34.23 W after 48 hours. Moreover, obtained voltage at different times for 48 h in humidified natural gas and air is shown in Fig. 5.

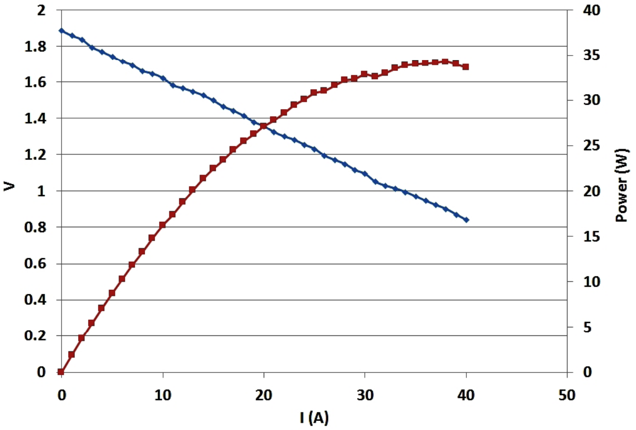

Fig. 4. Voltage and power versus current measured in humidified natural gas with flow rate of 1000 ccm and oxygen flow rate of 1000 ccm at 800°C after 48 h of operation in natural gas (area of the cells: 2×100 cm²).

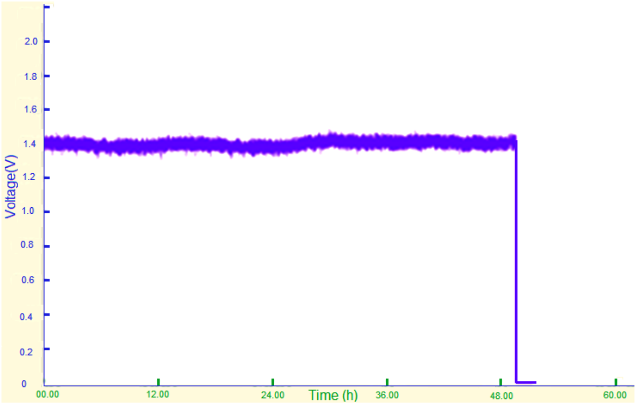

Fig. 5. Voltage versus time measured after 48 h in humidified natural gas and O_2.

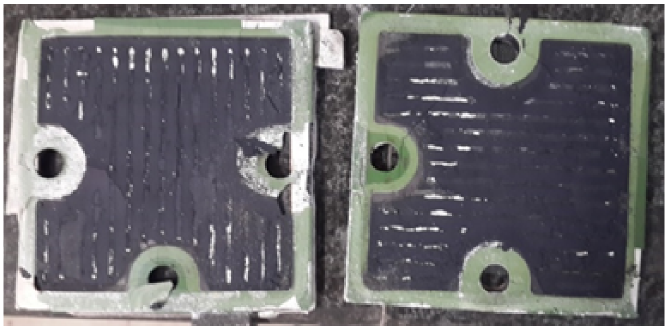

Fig.6. Image showing the two cells used in the stack after operation.

Table 1. Comparison of the result of this work with the previous reports

	Ref [17]	**Ref [18]**	**this work**
Power Density in wet methane (W cm^{-2})	0.27	0.35	0.21
Active area (cm^2)	84	16	80
Cell dimension (cm×cm)	-	-	10×10
Working Temperature (°C)	750	850	800
Reforming	Internal	Internal	Internal

The image of the cells after operation in the stack for 48 h is shown in Fig. 6 which illustrates full reduction of the cells. In addition, a trace of silver paste can be observed in the cathode sides of these cells.

Table 1 presented the comparison between some prestigious studies for stack testing of SOFCs [17, 18]. As it can be seen, the power densities is in the range of 0.21-0.35 W cm^{-2}. Internal reforming was used in all of these studies. It should be noted that the power density of the SOFCs is strongly dependent on the working temperature, the active area, the design of the interconnects and flow rates of the fuel and oxidant.

4.Conclusion

In summary, the present results demonstrate that using humidity reduces coke formation in the anode, ensuring the possibility of long-term operation. In the current stack configuration, the ratio of steam to fuel was 2:1. Combining steam with natural gas also led to direct reforming on the anode surface and the formation of carbon monoxide and hydrogen. As a result, after 48 h of operation no significant sign of reduction in over potential was observed. In addition, it was shown that the power increased from 32 W to 34 W after 48 h of operation. In conclusion, the SOFC stack suitable for direct natural gas was successfully fabricated using cost effective materials.

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