Journal of Power Sources 205 (2012) 345–349

Contents lists available at SciVerse ScienceDirect

Journal of Power Sources

journal homepage: www.elsevier.com/locate/jpowsour

Short communication

In situ monitoring of high-temperature proton exchange membrane fuel cell stack using flexible micro temperature and voltage sensors

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ARTICLE INFO

Article history:
Received 14 October 2011
Received in revised form
19 December 2011
Accepted 1 January 2012
Available online 21 January 2012

Keywords:
Micro-electro-mechanical systems
Flexible micro sensors
High temperature fuel cell stack
In situ monitoring

ABSTRACT

Internal local temperature and voltage not only affect reactions that occur in a membrane electrode assembly (MEA); they can also cause dehydration of the MEA in the electrochemical process that occurs in a proton exchange membrane fuel cell (PEMFC) stack, affecting its performance. In this work, micro-electro-mechanical systems (MEMS) are utilized to develop novel integrated micro sensors for use in high-temperature proton exchange membrane fuel cell stack, and integrated micro temperature and voltage sensors on stainless steel foil as a flexible substrate. These micro sensors can measure temperature and voltage data simultaneously and have the following advantages over other sensors: (1) they are small, (2) they are highly sensitive, (3) they can be batch-produced, (4) they can be placed anywhere to make measurements *in situ*.

In this work, six micro sensors are embedded in the flow field of a high-temperature fuel cell stack and used to monitor local temperature and voltage *in situ* given various operating parameters. The results demonstrate that embedding micro sensors do not affect on cell performance and that the temperature and voltage distribution are non-uniform in a high-temperature fuel cell stack.

1. Introduction

The high-temperature proton exchange membrane fuel cell (HT-PEMFC) has been attracting increasing attention over recent years because it poses less of a water management problem than the low-temperature PEMFC, and the HT-PEMFC stack has greater potential to be used in large power supply systems and transportation. However, some problems, such as membrane degradation, catalytic corrosion, and interior monitoring, must still be solved.

The interior temperature and voltage are crucial to the high performance of the HT-PEMFC stack, because temperature very strongly affects the activity of the catalyst, membrane dehydration, mass transfer, and heat management [1], and voltage also importantly influences the performance of the HT-PEMFC stack. Hence, monitoring and understanding local temperature and voltage in such cells are very important.

Ali [2] fabricated thin-film thermocouples (TFTCs) on a polyimide (Kapton) substrate to measure the internal temperature of a polybenzimidazole (PBI)-based high-temperature PEMFC. Gagliardo [3] fabricated 10×10 array sensors on a polytetrafluoroethylene (PTFE) substrate to measure *in situ* the temperature distribution using a signal data-acquisition system, and they analyzed the voltage distribution of a fuel cell by capturing a neutron image. Inman [4] embedded optical temperature sensors in a PEMFC to measure temperature *in situ* using phosphor thermometry. Chang [5] connected conducting wires to the anode and cathode of bipolar plates to measure the local voltage distribution in a fuel cell under various conditions. Büchi [6,7] interlaid thin gold wires as potential probes to decide the resistance distribution across the thickness of membranes. Lebæk [8] inserted T-type thermocouples were into the anode and cathode plates of an HT-PEMFC to measure internal temperature distributions with different operating gas stoichiometries. Andreasen [9] utilized thermocouples and simulation to compare the temperature distributions in two bipolar plates and to determine the output power of a fuel cell stack.

The authors' research team already has experience of developing film-shaped flexible micro temperature and voltage sensors to monitor local physical parameters in a low-temperature proton exchange membrane fuel cell [10]. However, this work develops an innovative approach for fabricating integrated flexible micro sensors (temperature and voltage) on a flexible stainless steel foil substrate using micro-electro-mechanical systems (MEMS). Stainless steel foil has many favorable properties, including corrosion resistance, compression resistance, high temperature resistance, and flexibility. The home-made micro sensors were embedded into a high-temperature proton exchange membrane fuel cell (HT-PEMFC) stack. The experimental results of fuel cell stack tests were

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doi:10.1016/j.jpowsour.2012.01.049

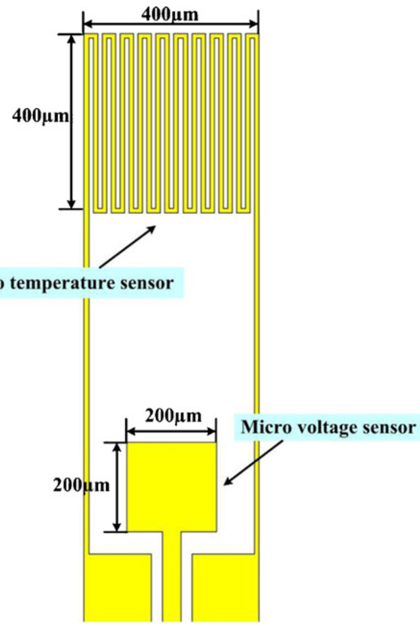

Fig. 1. Schematic drawing of micro temperature and voltage sensors.

used not only to compare the performance curves obtained without and with micro sensors, but also to determine variations of local temperature and voltage under various operating conditions.

2. Design and sensing principle

Flexible micro temperature and voltage sensors that were integrated on a flexible substrate were developed. Fig. 1 schematically depicts a strip-shaped micro sensor.

The micro temperature sensor is a resistance temperature detector (RTD). The electrodes of the micro temperature sensor are serpentine structures, with a sensing area of 400 μm × 400 μm. Sensing is based on the principle that as the environmental temperature increases, the resistance of the micro temperature sensor also increases, because the RTD has a positive temperature coefficient (PTC). When the temperature of the RTD is in the linear region, the relationship between the measured resistance and the change in environmental temperature can be expressed as:

$$R_t = \mathrm{R}_r(1 + \alpha_\mathrm{T}\Delta T) \quad (1)$$

where R_t is the resistance at t °C; R_r is the resistance at r °C, and α_T is the sensitivity of the micro temperature sensor (°C^{-1}).

The miniaturized voltage probe is utilized as the micro voltage sensor herein. The sensing area of the micro voltage sensor, which is in contact with the membrane electrode assembly (MEA), is 200 μm × 200 μm. The rest of the conducting wire is insulated to enable measurements to be made at particular locations inside a fuel cell.

3. Fabrication

Flexible micro temperature and voltage sensors were fabricated and integrated on a stainless steel foil (SS-304, 40 μm thick) as a flexible substrate using micro-electro-mechanical systems (MEMS). Stainless steel foil has several favorable characteristics advantages, including high corrosion resistance, high compression resistance, and high temperature resistance. The fabrication proceeds by the following steps (Fig. 2): (a) sulfuric acid (H_2SO_4) and hydrogen peroxide (H_2O_2) in a ratio of 3:1 are used to remove the passivation layer at 80 °C; (b) aluminum nitride (AlN) is sputtered to form a button insulating layer, with several favorable properties, including good electrical insulation, high thermal conductivity and high compression resistance; (c) an E-beam evaporator is used to deposit a chromium (Cr) adhesive layer and a gold (Au) sensing layer; (d and e) lithography and wet etching are carried out to define the sensing patterns of micro temperature and voltage sensors; (f and g) lithography and phosphoric acid are used to etch away unnecessary AlN; (h and i) double-side lithography is performed to form a protective layer before the stainless steel foil is etched using aqua regia; (j) a photoresist (PR) film is coated as an insulating layer and to protect the micro sensors during fuel cell testing, and lithography is then used to define the sensing voltage area and the pads of micro sensors.

Fig. 3 presents an optical microscopic photograph of flexible micro temperature and voltage sensors. The home-made flexible micro temperature and voltage sensors have numerous favorable characteristics, including smallness, high sensitivity, precision of

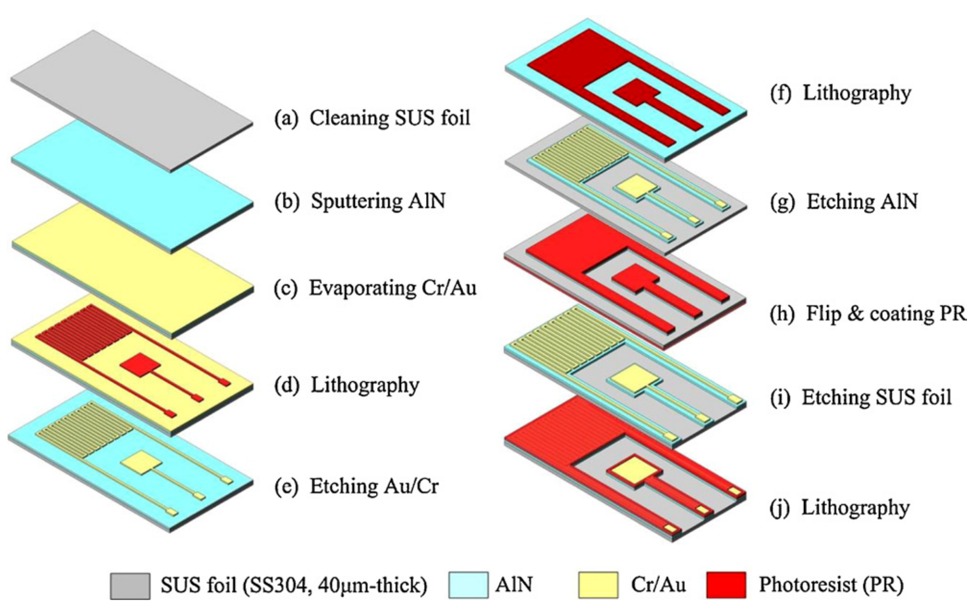

Fig. 2. Fabrication flowchart of flexible micro temperature and voltage sensors.

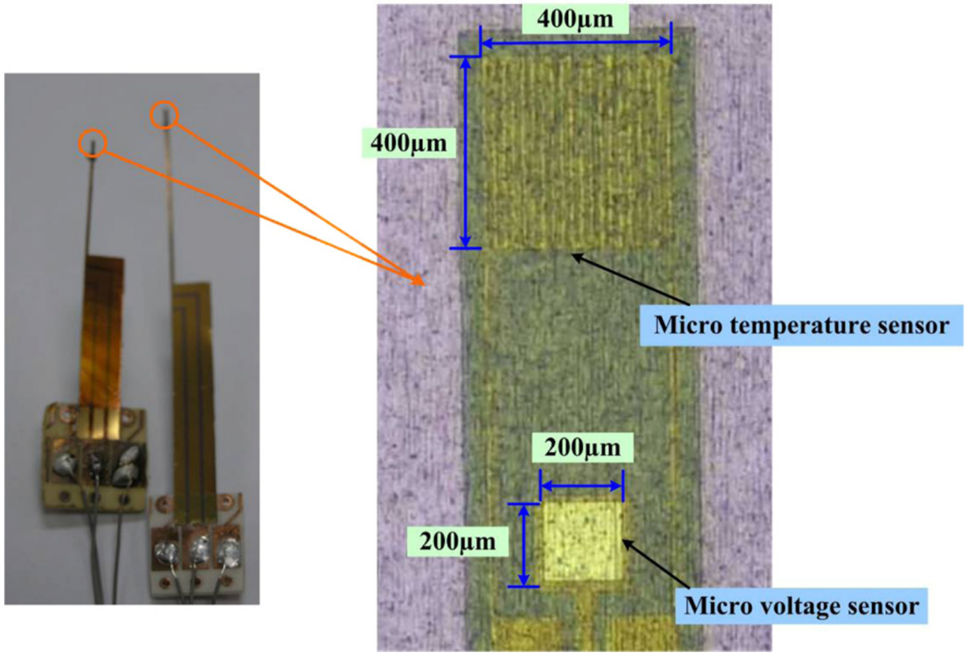

Fig. 3. Optical microscopic photograph of flexible micro temperature and voltage sensors.

measurement position, and the ability to make measurements *in situ*.

4. Results and discussion

4.1. Calibrated testing of flexible home-made micro sensors

After the micro temperature and voltage sensors had been fabricated, the micro sensors were connected to printed circuit boards using a wire bonder. Then, the developed micro sensors were calibrated in a DENG YNG Drying Oven, and their resistances were recorded using an NI PXI 1033, to ensure their reliability. Fig. 4 schematically depicts the setup for temperature calibration. Fig. 5 plots the calibration curves of three micro temperature sensors from 140 °C to 200 °C. The results reveal that the micro temperature sensors have high linearity and a sensitivity of $2.8 \times 10^{-3}\,°C^{-1}$.

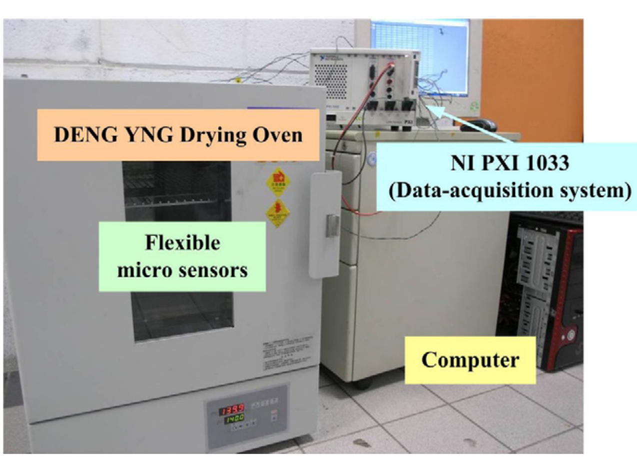

Fig. 4. Temperature-calibrated testing system.

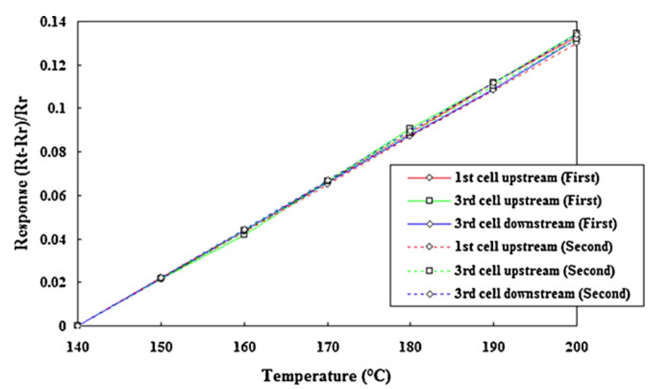

Fig. 5. Calibration curves of three micro temperature sensors from 140 °C to 200 °C.

4.2. Testing PEM fuel cell stack (four cells)

Flexible micro temperature and voltage sensors were embedded between the cathode flow channel plate and the membrane electrode assembly (MEA) of a high-temperature proton exchange membrane fuel cell stack (HT-PEMFC stack; four cells). The micro sensors were embedded upstream of the first, second and fourth cells, and upstream, midstream, and downstream of the third cell, as displayed in Fig. 6. A thermocouple was placed side the flow channel plate to compare its measurements with those of the micro sensors fabricated herein. The PEM fuel cell stack was tested using a 500 W PEM Fuel Testing Station; Fig. 7 and Table 1 present the operating conditions in detail. The performance curves without and

Table 1
Operating conditions of tested PEM fuel cell stack.

Items	Conditions
Cell operating temperature (°C)	160
Constant current (A)	5, 20
H_2 flow rate (anode) (slpm)	3
Air flow rate (cathode) (slpm)	1.2
Bipolar plate type	Poco graphite
Reaction area (cm^2)	31.4

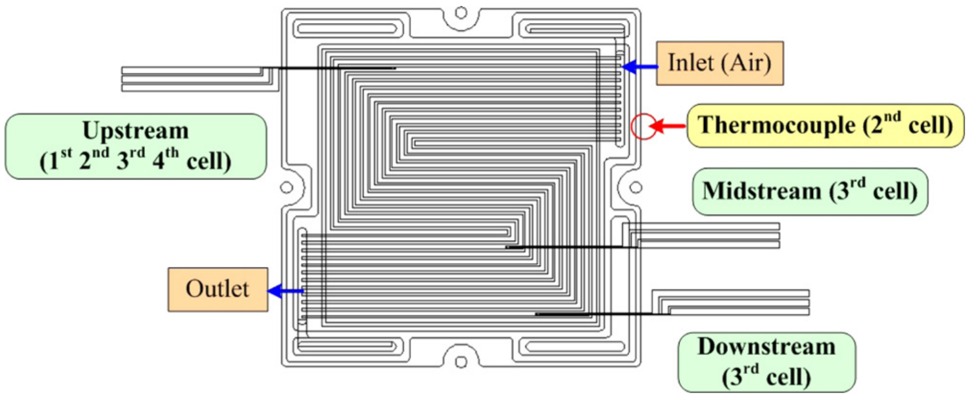

Fig. 6. Locations of embedded micro sensors in cathode flow channel plate.

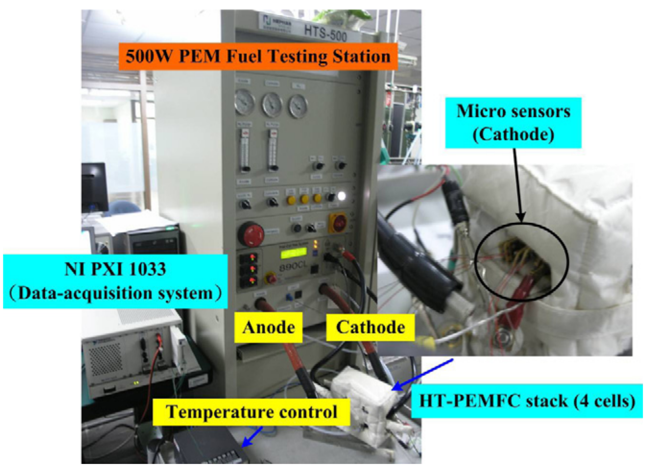

Fig. 7. Set-up of HT-PEMFC stack testing.

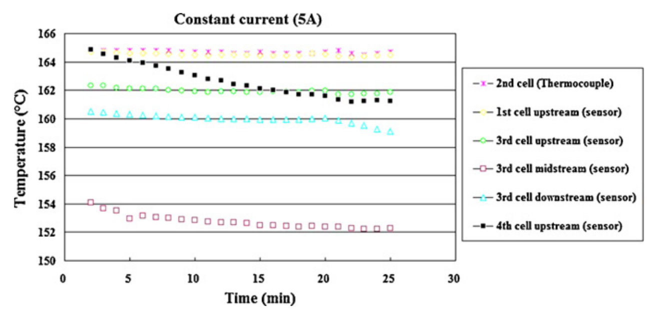

Fig. 9. Output local temperature variations in HT-PEMFC stack at constant current of 5 A.

with micro sensors are compared, and the local temperature and voltage were measured using the micro sensors and analyzed.

4.2.1. Difference between performance curves without and with micro sensors

Fig. 8 plots the performance curves without and with micro sensors inserted into the HT-PEMFC stack at 160 °C. Although the micro sensors that were embedded in the HT-PEMFC prevented the reaction between the reaction fuel and MEA, the results revealed that it less influence for stack performance because the blocked areas of micro sensors are 0.4% of the reaction areas of MEA.

4.2.2. Variations of local temperature and voltage at constant currents of 5 A and 20 A

Figs. 9 and 10 plot the output local temperature variations in HT-PEMFC stack at constant currents of 5 A and 20 A. The figures indicate that the distributions of local temperature in the cell stack were non-uniform. The temperature variations were gentle because of the mitigation of the reaction in the cell stack at low current; lower temperatures were obtained from the micro sensors midstream in the third cell. Fig. 10 demonstrates that the local temperature curves rose gradually because of the violent reaction, and the rising curves obtained using the micro sensors are consistent with that of the thermocouple to which the sensors were compared.

Table 2 presents the local voltage values in the HT-PEMFC stack at constant currents of 5 A and 20 A. The results reveal that the local voltages varied among all cells and were clearly non-uniform at the high current of 20 A.

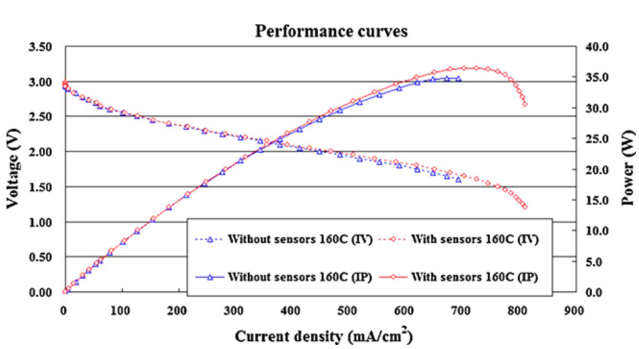

Fig. 8. Performance curves without and with micro sensors.

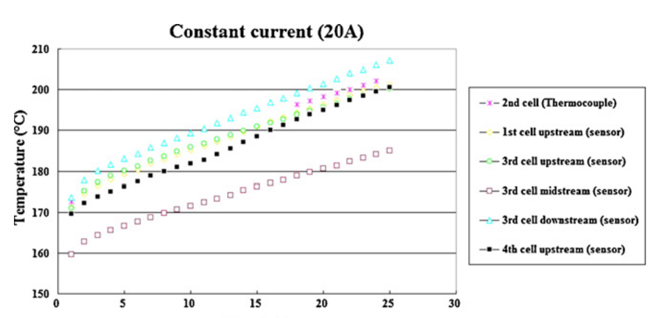

Fig. 10. Output local temperature variations in HT-PEMFC stack at constant current of 20 A.

Table 2
Local voltages in HT-PEMFC stack at constant currents of 5 A and 20 A.

Parameters	Location	Micro voltage sensor	Commercial voltage sensor
Constant current (5 A)	2nd cell upstream	0.606 V	0.605 V
	3rd cell upstream	0.523 V	0.612 V
	3rd cell midstream	0.609 V	
	4th cell upstream	0.598 V	0.597 V
Constant current (20 A)	2nd cell upstream	0.465 V	0.429 V
	3rd cell upstream	0.418 V	0.48 V
	3rd cell midstream	0.464 V	
	4th cell upstream	0.437 V	0.415 V

5. Conclusions

Flexible micro sensors, which integrated micro temperature and voltage sensors, were successfully developed using micro-electro-mechanical systems (MEMS). The home-made flexible micro sensors have the following benefits: (1) they are small and flexible, (2) they are highly sensitive, (3) they can be batch-produced, and (4) they can be placed anywhere to make measurements *in situ*.

In this innovative investigation, non-invasive micro sensors were inserted into the cathode flow channel plate in an HT-PEMFC stack (four cells) to compare their performance curves, and to measure and analyze the local temperature and voltage in the stack. Experimental results confirmed that the embedded micro sensors had little effect on the proposed HT-PEMFC stack performance, and the distributions of local temperature and voltage were non-uniform. The interior measurements made using the micro sensors on the experiment are made herein for the first time. In the future, the number of embedded micro sensors should be increased and relevant parameters varied to analyze completely local conditions on a micro-scale in an HT-PEMFC stack.

Acknowledgments

This work was accomplished with much needed support and the authors would like to thank for the financial support by Bureau of Energy, Ministry of Economy Affair of ROC through grants 100-D0204-4. The authors also like to thank Shih Hung Chan, Shuo-Jen Lee and Ay Su of Yuan Ze University for their valuable advice and assistance in the experiments. In addition, we would like to thank the YZU Fuel Cell Center and NENS Common Lab, for providing access to their research facilities.

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