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# Membraneless Microfluidic Fuel Cell with Silver Electrode Fabricated by Low Temperature Co-fired Ceramic Combined with Photolithography†

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A microfluidic fuel cell fabricated by low temperature co-fired ceramic process was introduced. It was an alkaline fuel cell and produced the electric power *via* electrochemical reaction using a diverse fuel and an oxidant solution. Highly conductive silver was used for an electrode as a less expensive and non-precious metal catalyst. The fuel is 1 M KOH and the oxidant is 1 M $H_2SO_4$ without any electrolytes. In particular, by adding oxygen into the both of solutions by bubbling system, a higher open circuit potential was observed compared to that of normal condition. To enhance the efficiency of potential voltage of single-channel fuel cell, a noble design for dual-channel was employed in a microfluidic fuel cell. After optimizing the flow rates of fuel and oxidant, the open circuit potentials of single-channel and dual-channel microfluidic fuel cell were about 0.25 V and 0.65 V, respectively.

**Keywords: Ceramics, Microfluidic channel, Alkaline fuel cells, Silver electrode.**

## INTRODUCTION

As the market of the portable applications grows up, the technology for high-energy density in micro-structure has been a key issue in the fuel cell technology[1]. A number of investigations have been focused on the fabrication process of micro fuel cells with ion exchange membranes. Therefore, there is a great advance in the technology of proton exchange membrane (PEM)-based fuel cells. However, those have some limitations for mass production due to high operating temperature for drying out of the membrane and fuel crossover[2]. Recently, microfluidic fuel cells have received a great concern due to its potential applications to many specialized potable device, such as mobile phone, tablet device and healthcare sensor system[3]. Microfluidic fuel cell technology was introduced by many groups because it could be operated well without any membrane. A phenomenon of laminar flow in the microfluidic channel with the specific dimensions makes it possible to form the co-laminar streams between the fuel and the oxidant. There is diffusion at the interface between the fuel and the oxidant but it can be minimized by adjusting the flow rate of the streams[4,5]. However, it also has a drawback of low energy conversion efficiency due to short single pass distance[6]. In the microfluidic fuel cell technology, alkaline fuel cell is preferred because it is operated by anion exchange instead of proton exchange during the electrochemical cell reaction and more cost-effective allowing the use of non-precious metals as catalysts[7-10].

In this paper, to fabricate a ceramic–based microfluidic fuel cell, a noble process for low temperature co-fired ceramic (LTCC) technology combined with photolithography was suggested. Silver electrode was embedded in the ceramic body after patterning through screen printing process. Especially dual-channel fuel cell design was employed to enhance the efficiency of potential voltage of single-channel fuel cell. The performance of membraneless microfluidic fuel cells was evaluated by measuring the open circuit potential (OCP) and it was optimized by changing the various experimental factors, such as the configurations and the flow rate of media.

## EXPERIMENTAL

To fabricate a ceramic-based microfluidic membraneless fuel cell, the dimension of micro-channel and the arrangement of silver electrode were designed considering the flow of fuel and oxidant. A microfluidic fuel cell has the Y-shaped micro-channel pattern as shown in Fig. 1. It uses the co-laminar flow of the fuel and the oxidant between two counter parts of electrodes on the side of channel. To enhance the potential voltage of the single-channel type, the dual-channel type

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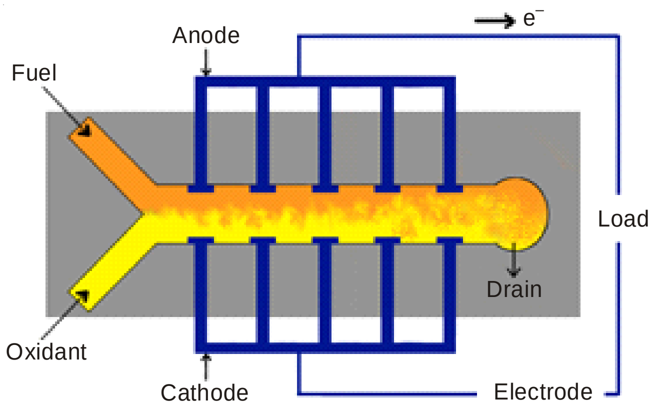

Fig. 1. Schematic diagram of a membraneless microfluidic fuel cell with silver electrode

device was developed to introduce a large amount of fuel and oxidant.

The ceramic-based fuel cells were fabricated by low temperature co-fired ceramic (LTCC) process combined with photolithography to form a micro-channel and a silver electrode. The main ceramic material for LTCC process was the Ca-modified amorphous cordierite [$(Mg,Ca)_2Al_4Si_5O_{18}$]. Two kinds of LTCC slurry were prepared to make the ceramic-based green sheets for fuel cells. One was the conventional LTCC slurry to form a rigid substrate and the other was the photosensitive slurry which was used to make a micro-channel pattern for constant fuel and oxidant streamline. To make the LTCC slurry, the main powder was dispersed with a solvent (toluene-to-ethanol ratio of 6:4) including a polyvinyl butyral binder and a dispersant.

Then, the slurry was blended homogeneously by a ball-milling for 24 h. To make the photosensitive slurry for microfluidic channel, photo-initiators, monomers, plasticizers and sensitizers were added to the conventional one. Silver electrode was patterned on a conventional green sheet by screen printing process using highly conductive silver paste (S-3900B, IMD Korea) with a 400-mesh screen. The green sheet for a substrate and the photosensitive green sheet were laminated over 1 mm to sustain a rigid form. After the lamination, the micro-channel was formed by photolithography process using 365 nm ultra-violet (UV) light and photo mask. Finally, it was sintered at 800-850 °C for 1 h. The overall process of LTCC-based microfluidic fuel cell is shown in Fig. 2.

After the sintering procedure, polydimethylsiloxane (PDMS, Sylgard 184, Dow Corning) was bonded on the device cell by a plasma treatment (Femto Science) as a transparent top cover. Then, tygon tube (S-54-HL, Harvard Apparatus, Holliston, MA) were connected to inject the solvent using a micro-pump (Pump 11 Pico Plus: Harvard Apparatus, Holliston, MA). All the aqueous solutions were conducted at room temperature using 1 M KOH for fuel and 1 M $H_2SO_4$ for oxidant. The fuel and oxidant were pumped by a micro-pump (Pump 11 Pico Plus: Harvard Apparatus, Holliston, MA). The evaluation of electrochemical reaction was progressed using a potentiostat (KIKUSUI, Electronic Load PLZ70UA).

## RESULTS AND DISCUSSION

Fig. 3 shows the overall images of single and dual-type microfluidic channels after UV photolithography using photosensitive green sheets. Through applying the conventional photolithography to LTCC process, two types of micro-channel patterns, single-channel cell and dual-channel cell, were obtained. It was confirmed that the straight channels with the width of 3.5 mm and the length of 10 cm were well formed through the UV exposure and hardening process.

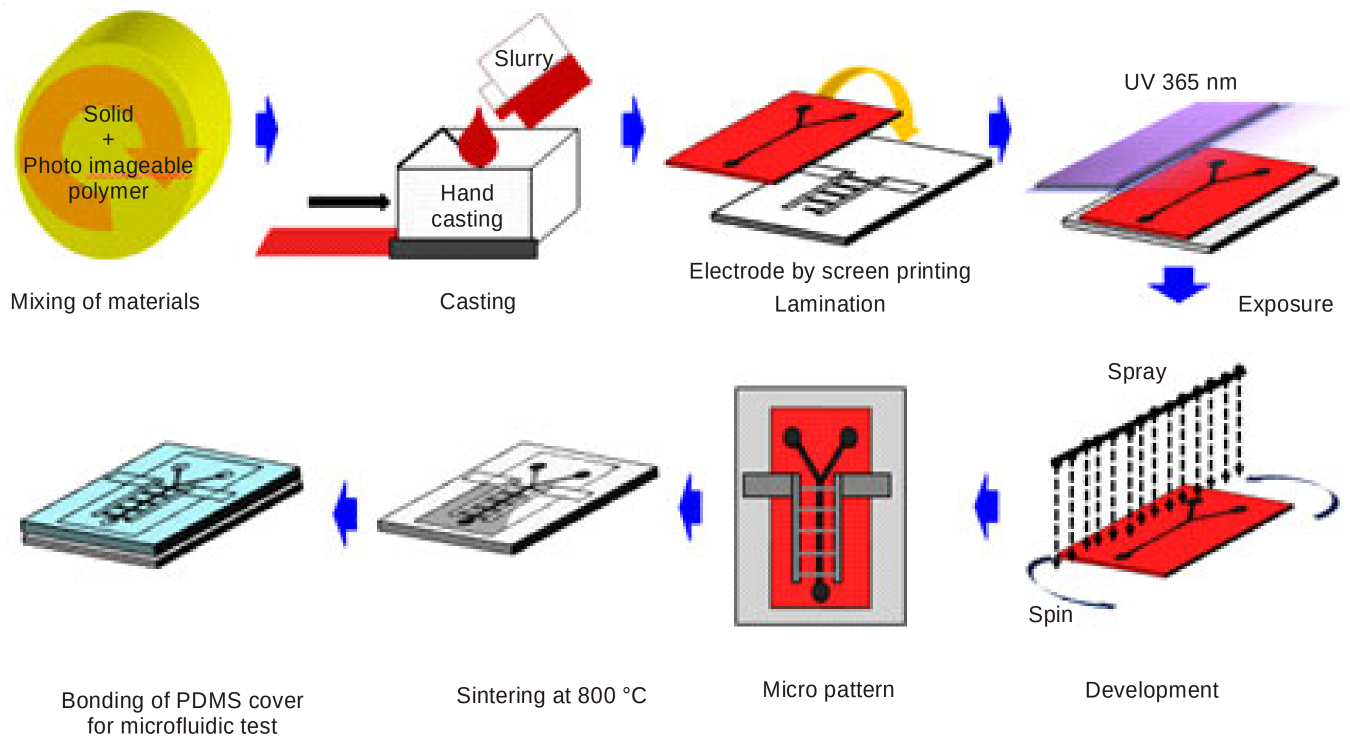

Fig. 2. Fabrication process of the ceramic-based microfluidic fuel cell

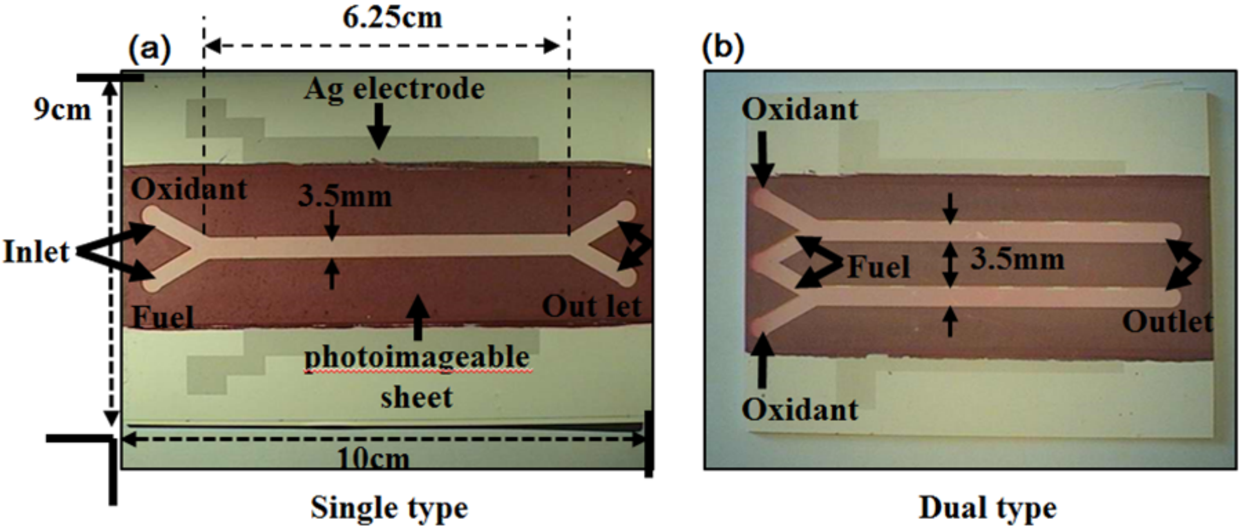

Fig. 3. Optical image of membranless micfofluidic fuel cell before the sintering process. (a) single-channel pattern, (b) dual-channel pattern

There were two injection holes and drain holes for fuel and oxidant in the single-channel cell. In the dual-channel cell, there were three injection holes; two holes at outsides for the oxidant stream and center hole for the fuel stream.

On the conventional LTCC green sheet, silver electrodes were patterned by screen printing process. The 6-pairs of electrode were aligned on the bottom of channel and those showed the length of 50 mm and the thickness of 50 μm. The green sheets were stacked up over 1mm in order to prevent the device from a warp arising during a sintering process. The optical image of membranless microfluidic fuel cell after sintering process was showed in Fig. 4. It showed the width of 5 cm and the length of 7 cm. The microfluidic channels showed the width of 3 mm width and the length of 5 cm. Microfluidic channel height was 150 μm.

The microfluidic fuel cells test was performed using 1 M KOH and 1 M $H_2SO_4$ in 18.3 Ω-cm Millipore water for fuel and oxidant, respectively. The performance of microfluidic fuel cell was characterized by observing open circuit potential. To enhance the value of open circuit potential, the solution of fuel and oxidant was saturated with oxygen by bubbling throughout an aqueous solution of 1 M KOH and 1 M $H_2SO_4$ for 30 min (5 L/min). KOH solution played a critical role that it was not only for fuel but also for electrolyte to move electron ($e^-$) from anode to cathode.

Fig. 5 showed the variation of open circuit potential for 300 sec at different flow rates from single-channel type fuel cell [Fig. 5(a)] and dual-channel type [Fig. 5(b)]. At the beginning, open circuit potential of single and dual-channel type of microfluidic fuel cell were roughly obtained in excess of 0.2-0.25 V and 0.5 V respectively. The flow rates of single-channel type device was changed from 100 to 400 mL/min, while different flow rates between fuel and oxidant stream of dual-channel cell were applied to form a balanced laminar flow both of channels from 500 to 800 mL/min of the fuel solution and from 100 to 400 mL/min of the oxidant solution. It was clearly observed that the open circuit potential of dual-channel fuel cell was about two times higher than that of single-channel cell.

Fig. 6 showed the values of open circuit potential from single and dual-channel types of microfluidic fuel cell with fuel and oxidant saturated with oxygen. In the single-channel

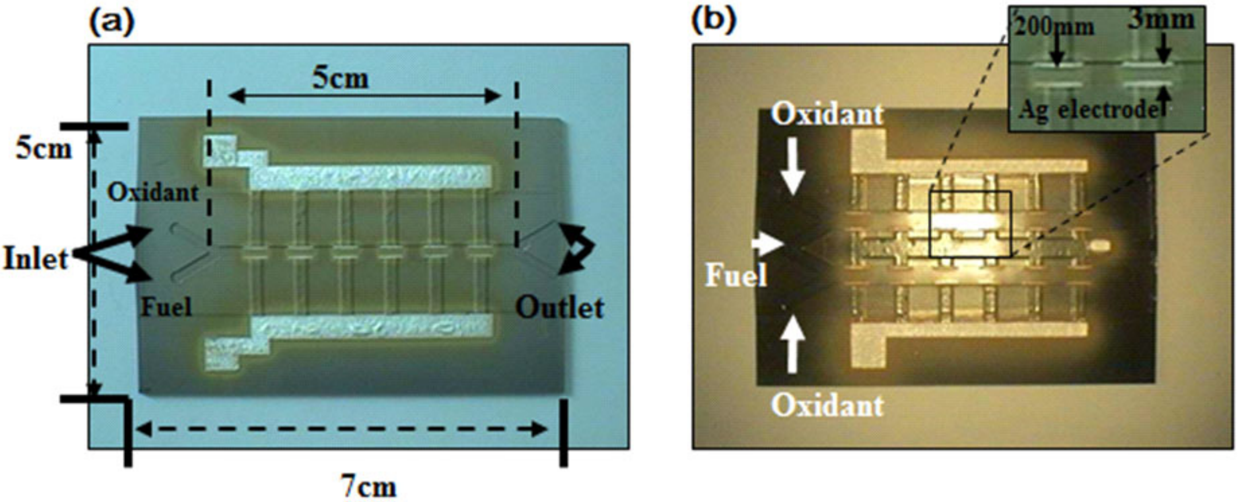

Fig. 4. Optical image of membranless micfofluidic fuel cell after the sintering process. (a) single-channel fuel cell and (b) dual-channel fuel cell

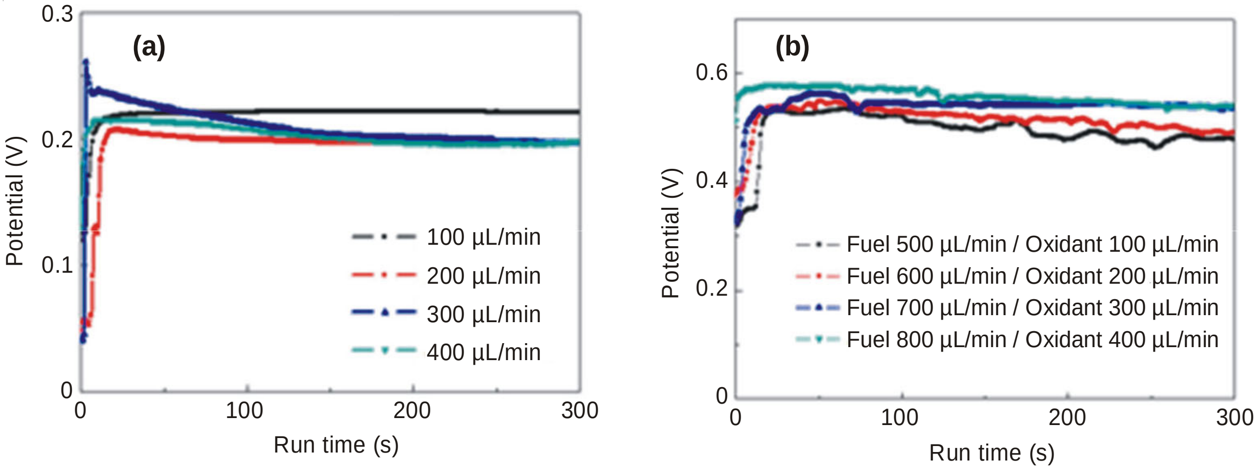

Fig. 5. Open circuit potential (OCP) values *vs.* run time of electrochemical reaction from (a) single-channel and (b) dual-channel fuel cell

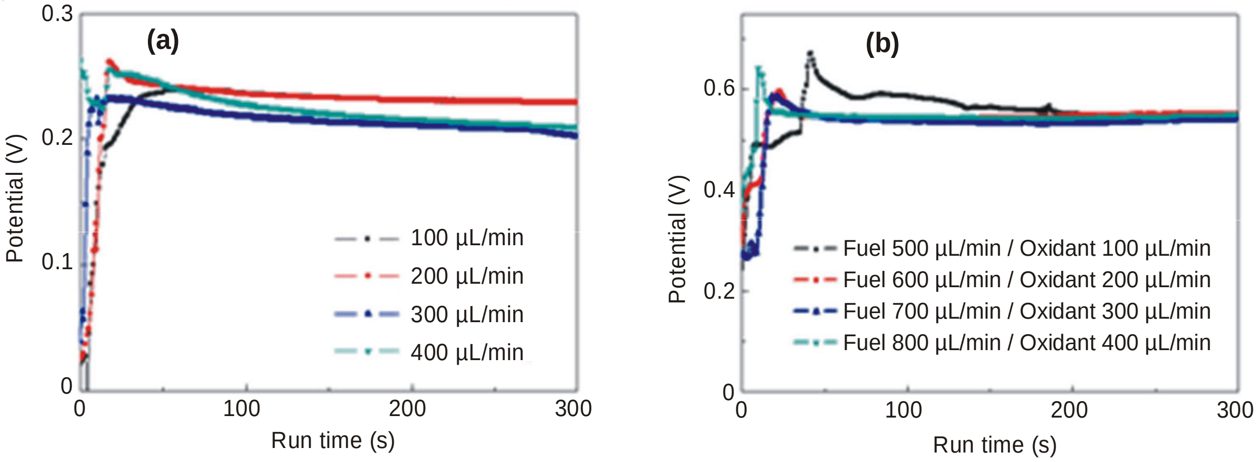

Fig. 6. Open circuit potential (OCP) values *vs.* run time of electrochemical reaction with oxygen saturated solution from (a) single-channel and (b) dual-channel fuel cell

microfluidic fuel cell [Fig. 6(a)], the open circuit potential value was similar to the previous result shown in Fig. 5(a). In the dual-channel type fuel cell, however, the open circuit potential value saturated with oxygen was 0.55 V with average and showed the highest point of 0.65 V at the flow rate of 800 mL/min for fuel and 400 mL/min for oxidant. After reaching the open circuit potential value of 0.65 V, it was decreased continuously as the run time was progressed. It might be caused by the increase of resistance in the solution through the formation of bubbles.

## Conclusions

A membraneless microfluidic fuel cell was fabricated by LTCC process combined with photolithography. It was confirmed that microfluidic channel was well formed by photolithographic technique using photosensitive green sheet and silver electrode was embedded successfully into the LTCC-based fuel cell device. Two types of fuel cells, single-channel and dual-channel, were designed and the performance was compared using 1 M KOH fuel and 1 M $H_2SO_4$ oxidant. The open circuit potential values from single-channel and dual-channel fuel cell were about 0.25 and 0.5 V, respectively. When fuel and oxidant with oxygen saturated were used in electrochemical reaction, open circuit potential was showed the highest value of 0.65 V.

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