Design and Fabrication of Miniaturized Fuel Cells Using Microfabrication Technologies

반도체 공정을 이용한 소형 연료전지의 제작

B. -J. Choi, I. -H. Oh*, H. Y. Ha*, S. -A. Hong*, and Y. -K. Sun
Department of Chemical Engineering, Hanyang University
* Fuel Cell Research Center, Korea Institute of Science and Technology

1. Introduction

Fuel cells are becoming attractive solutions to the ever-increasing need for portable electronic power sources, especially in response to the rapid growth of mobile computing devices and portable electronic telecommunications, including cellular phones, personal digital assistants (PDA), notebook computers, game devices and portable music systems. It is expected that these machines will become more numerous and more diverse in the coming years. Fuel cells are well known of high energy density, a variety of fuel sources, and ease of scaling for application-specific power requirements. Fuel cells with polymer electrolyte membranes are particularly attractive because of low-temperature operation and relatively simple construction. Small volume and low mass are naturally critical requirements for portability. Therefore, the miniaturization of PEMFCs can be a successful solution for portable electronic power sources.

Recently, microfabrication has been emerging as an ideal technology for the miniaturization of chemical systems because of its well proven cost-effectiveness, reproducibility, and control [1]. Microchemical systems have many advantages over macrosystems, including increased rates of heat and mass transfer, increased safety as a result of smaller volume and enhanced temperature control, and reduced volume waste streams. Up to date, microchemical systems have been used to analyze DNA [2], release controlled amounts of drugs [3], and fabricate microelectrodes within a flow channel [4]. Some of the strengths of microfabrication techniques include fine feature resolution, high repeatability, batch operations, integrated process sequences, and a variety of material transfer options.

In this study we used microfabrication technique to make miniaturized fuel cells. One fundamental objective of this study is to adapt appropriate microfabrication methods for the production of miniaturized fuel cells within this range of application.

2. Experimental

Silicon substrate for miniaturized fuel cells were fabricated using the microfabrication process as depicted in Fig. 1.

The catalyst ink for electrodes was formulated similar to DMFC catalyst ink. To avoid CO poisoning in the anode side, Ru was added to Pt. $4mg/cm^2$ Pt/Ru black was used and Nafion ionomer was added as the bridge of H+ ion. For the cathode side, Pt black was applied.

The cathode was prepared by spray-coating the prepared catalyst ink onto a carbon ink, previously coated as a slurry onto the active area of an electrode chip by screen-printing. The Pt was dispersed in the liquid by sonication. This ink was applied by spray-coating through a Teflon mask, drying with a convection oven, and repeating the spray-dry cycle to achieve the final mass loading.

The anode was prepared by the same procedure to that used for the cathode side, but instead of using Pt black, Pt/Ru black was spray-coated as a slurry.

The pretreated Nafion 115 membrane was used as the proton exchange membrane.

MEAs(Membrane Electrode Assembly) were fabricated in two methods. One MEA was prepared by spray-coating catalyst slurry directly onto the pretreated Nafion 115 membrane through a Teflon mask. Next, the pretreated membrane and silicon wafer substrates were assembled together by hot-pressing at approximately 45 psi (3 atm) and 150℃ for 5 min. The other one was prepared by spray-coating catalyst slurry directly onto the active area of electrode chips. And then silicon sealant was pasted the edges of an silicon wafer substrate to assemble both chips and membrane.

For direct comparison to the membrane electrode assembly in a typical PEMFC, we also prepared a miniaturized fuel cell with anode and cathode catalyst layers that were directly spray-coated onto a Nafion 115 membrane. Before use in a miniaturized fuel cell, the double-sided, catalyst coated membrane was hydrated in DI water overnight.

Performance of the cells was measured by electronic load (DAEGIL inc.).

3. Results and discussion

Fig. 2 gives microscope image of the active areas of the electrode design tested during the present investigation. Feed hole rib size were the primary variables in this electrode design. The sizes used here insured excellent liquid/gas handling in the individual electrodes. Changing each of the variables could be expected to have serious effects on methanol crossover, catalyst utilization, and cathode flooding. The feedhole and rib sizes gave a total feedhole area of 0.0928 $cm^2$and a total rib area of 0.397 cm^2.

The chip design devised in the present study was simple and compact. However, it could be changed to get better performance in the ongoing study. Feedhole size could be reduced for the ease of fuel distribution, and the number of feedhole could be increased.

In Fig. 4. the shape of voltage versus current density curve is typical for a DMFC. The initial drop of the polarization curve at very low current density was due to an electrochemical activating process, which was caused by the sluggish kinetics of oxygen reduction at the cathode surface. The subsequent linear decrease of the polarization curve was due to ohmic over-potential, which was attributed to the ion flow through the electrolyte membrane, the electron flow through the electrode materials, and current collector.

When it is compared with other macro DMFC unit cell [5-9], the peak power density is about half of it. The reasons for the low peak power density may be described as follows:

(1) When the pretreated membrane and silicon wafer substrates are bonded, we do not use high pressure bonding.

(2) In the result of (1) miniaturized fuel cells have higher contact resistance between Nafion115 membrane and silicon wafer substrate than macro DMFCs.

(3) Miniaturized fuel cells tested with macroscopic support hardware (e.g. copper wires, clip leads, etc.) may actually suffer to a greater extent from interface losses than large-scale cells. The best performance of miniature components can be demonstrated only after improved interfacing solutions are incorporated.

It is possible that direct comparision a miniaturized fuel cell using a new electrode catalyst ink coating method in Fig. 4. with a miniaturized fuel cell using a typical DMFC membrane electrode assembly (MEA) in Fig. 5.

The measured performances of both cells were almost same, but a miniaturized fuel cell using a new method has some advantages:

(1) The ease of substitution for spray-coating method of the electrode catalyst layers (e.g. magnetron sputter deposition, electrodeposition, etc.)

(2) It is possible that the application of thin-flim electrolyte deposited by sputter deposition.

In Fig. 6. cell performance was measured according to various catalyst loading. Both electrodes has the same catalyst loading.

The state of the art miniaturized fuel cells could be realized by incorporation with thin-film electrodes, electrolyte layers and silicon wafer structure manufactured by microfabrication process.

4. Conclusions

Miniaturized fuel cells, which can be applied to portable electronic power sources, have been designed, fabricated, and tested in this study. Adaptations of microfabrication processes such as photolithography and silicon etching have enabled us to manufacture the miniaturized fuel cells.

Some critical fabrication issues have been identified for realizing the design concepts. Especially integration concerns are important, because each component presents a set of mechanical, thermal, and electrochemical requirements that may conflict with other aspects of processing. The optimization of integrated sequences are ongoing works for improved performance. For the variety of application, stacking technologies are currently under development.

5. References

1. Ehrfeld, W., F. Golbig, V. Hessel, H. Lowe, and T. Richter, *Indust. Eng. Chem. Res.*, **38** (1999) 1075.
2. Burns, M. A., B. N. Johnson, et al, *Science*, **282** (1998) 484.
3. Santini, J. T., M. J. Cima, and R. Langer, *Nature*, **397**(1999) 335.
4. Kenis, P. J. A., R. F. Ismagilov,and G. M. Whitesides, *Science*, **285** (1999) 83.
5. M. Waidhas, W. Drenckhahn, W. Preidel, and H. Landes, *J. Power Sources*, **61** (1996) 91-97.
6. Martin Hogarth, et al, *J. Power Sources*, **69** (1997) 113-124.
7. Martin Hogarth, et al, *J.Power Sources*, **69** (1997) 125-136.
8. Xiaoming Ren, et al, *J. Power Sources*, **86** (2000) 111-116.
9. H. Dohle, J. Divisek, and R. Jung, *J. Power Sources*, **86** (2000) 469-477.

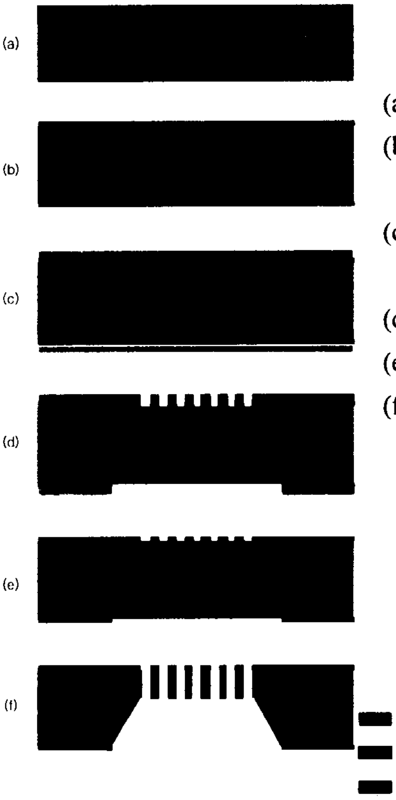

Fig. 1. A schematic fabrication process of silicon substrate chip.

(a) A bare, double-side polished silicon wafer.

(b) Deposit silicon dioxide (SiO2) layer on both-side of the wafer by thermal oxidation..

(c) Coating photoresist on both-side of the wafer by spin-coating.

(d) Pattern and etch window in SiO2 layer.

(e) Remove photoresist layer by acetone.

(f) Etch both-side of the wafer in hot TMAH solution to form feed holes.

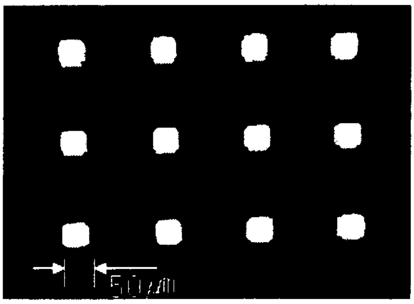

Fig 2. Optical microscope photograph of feed holes in electrode chip

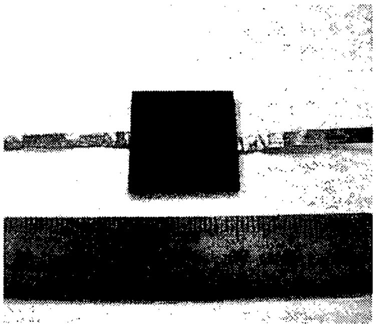

Fig 3. Photograph of a miniaturized fuel cell

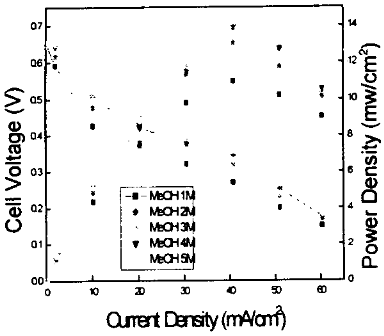

Fig. 4. Performance curves of a miniaturized fuel cell with a new method: cell temp.25℃

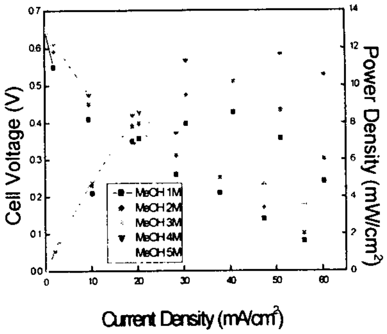

Fig. 5. Performance curves of a miniaturized fuel cell with a typical MEA: cell temp.25℃

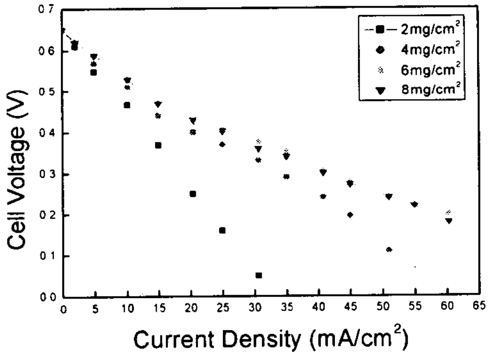

Fig. 6. Effect of catalyst loading on the cell performance