In-situ visualization of generated gas distribution in an operating direct hydrazine hydrate fuel cell by soft X-ray radiography

T. Sakamoto[a], K. Asazawa[a], P. Deevanhxay[b], S. Tsushima[b], S. Hirai[b], and H. Tanaka[a]

[a]*Frontier Technology R & D Division, Daihatsu Motor Co., Ltd., 3000 Yamanoue, Ryuo, Gamo, Shiga 520–2593, Japan*

[b] *Dept. of Mechanical and Control Engineering, Tokyo Institute of Technology, 2-12-1 O-okayama, Meguro-ku, Tokyo 152-8552, Japan*

Direct liquid fuel cells, such as the methanol, ethanol, borohydride, formic acid and hydrazine hydrate systems, are attractive candidates for portable electronics and vehicular applications, because of their high energy density. Direct hydrazine hydrate fuel cells (DHFCs) have been studied since the 1960s. DHFCs can be operated without any previous metal catalyst, which potentially contributes to cost reduction of the system. Recent developments in DHFCs technology have led to full-sized demonstration DHFCs indicating their feasibility and promise in future fuel cell vehicles. In DHFCs, electrochemical reactions take place in the anode and the cathode, respectively.

Anode: N_2H_4 (liq) + $4OH^- \rightarrow N_2$ (gas) + $4H_2O$ + $4e^-$ [1]
Cathode: $O_2 + 2H_2O + 4e^- \rightarrow 4OH^-$ [2]

Hydrazine electrooxidation leads to harmless N_2 and H_2O products, and the theoretical efficiency and theoretical potential (1.62 V) of DHFCs are higher than H_2/O_2 PEFCs.

For further improvement on cell performance, reduction of fuel crossover and efficient removal of generated gas are of great importance. Nitrogen gas electrochemically-generated in the anode can be remained at the reaction sites, which could deteriorate the cell performance. Nitrogen gas retention in DHFCs could cause inhomogenous electrochemical reaction in the anode catalyst layer. Therefore, a fundamental understanding of anode gaseous generation and their effect on the fuel cell performance is essential to improve the DHFCs performance. In this study, we first report insitu visualization of gas distribution by using high-resolution soft X-ray radiography and discuss gas generation behaviors with current density variation.

Fig. 1 shows the schematic of cell. The observation area of the rib and channel are shown in Fig.1b. Fig. 2 shows the current density and cell voltage as a function of time on visualization test using an in-plane cell. The imaging area was a 0.5 × 0.5 mm region in the outlet part of the cell, corresponding to the region under the rib. The I-V test was performed using 1 M KOH + 1 M hydrazine hydrate as an anode fuel and oxygen gas at room temperature at flow rate of 2 ml·min^{-1} and 300 ml·min^{-1}, respectively. As shown in Fig. 2, cell performance readily becomes stable after the load is changed. The cell contains no generated nitrogen under the initial conditions, but 815 s after the initiation of power generation, generated nitrogen that grow inside the GDL under the rib is observed as shown in Fig. 3. Fig. 4 shows the generated nitrogen distribution as a function of time. The nitrogen gas is generated as current density has increased and is observed near the rib. This suggests that the generated nitrogen pass out from rib to channel using localized pathway in the GDL. On the other hand, no generated nitrogen is observed in the center of the imaged region. Even though hydrazine hydrate and oxygen are uniformly supplied to the plane of the catalyst layer, a non-uniform distribution of generated nitrogen is observed. One possible explanation for this non-uniform distribution of generated nitrogen is inhomogeneity of the electrochemical reaction inside the catalyst layer at the sub-millimeter scale.

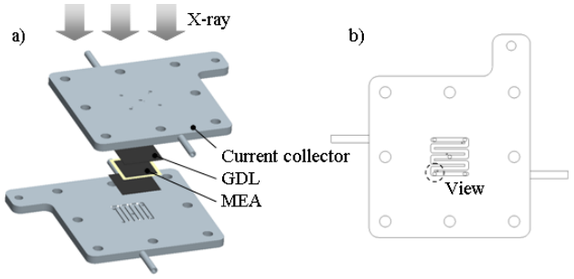

Fig. 1 Schematic of the through-plane visualization cell for soft X-ray microimaging: (a) cell configuration, (b) schematic of flow field plate.

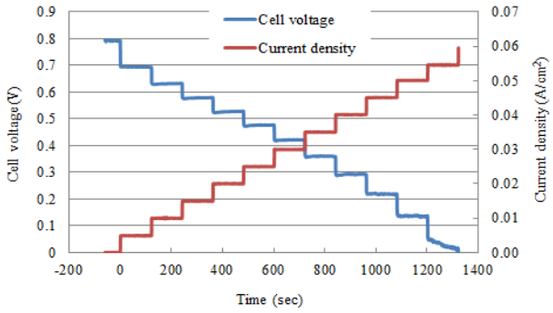

Fig. 2 Current density and cell voltage as a function of time during visualization test.

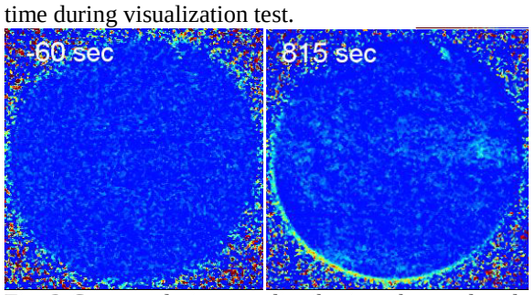

Fig. 3 Generated nitrogen distribution observed under the rib area as a function of time.

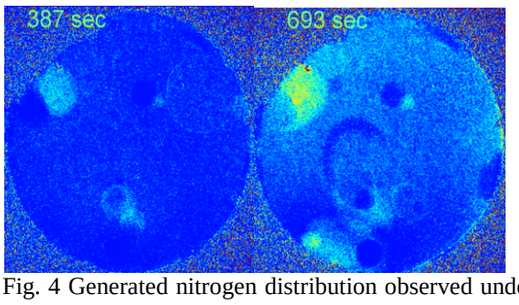

Fig. 4 Generated nitrogen distribution observed under the channel area as a function of time.

Acknowledgements

We would like to thank Dr. T. Sasabe (Nagoya Univ.) for his advice on construction of cell for the visualization test.