Electrocatalytic reduction of gas-phased CO_2 on nano-sized Sn electrode surface

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Carbon dioxide (CO_2) is considered as one of the main contributors to global warming, which is becoming an increasing concern in recent years [1,2]. Many methods to reduce CO_2 have been explored for a long time now. In particular, electrochemical conversion of CO_2 (ECC) into other useful chemicals has attracted interests as a possible technique. Furthermore, ECC has a prognostic significance in utilizing CO_2 as a carbon source and creating valuable chemicals.

During ECC, it is necessary to use the appropriate catalyst because different chemicals, such as methane, ethylene and formic acid, are produced depending on the type of catalyst. Electrocatalysts for ECC are classified into several groups based on the nature of the primary product obtained from the electrochemical reduction [2]. In particular, CO_2 has been reported to be converted to formic acid on metal electrodes such as Sn, Pb [3-5]. ECC has been studied generally in the aqueous phase. However, problems regarding the solubility of CO_2 and products separation from electrolyte as well as deactivation of electrode catalysts are encountered. It is therefore difficult to operate stably for long period of time. A new approach to overcome these problems is the direct ECC in gas-phase using zero-gap cell with GDE and Nafion [4,5]. Applying the zero-gap cell to ECC raised the possibility of long term operation in solving the mass transfer limitation of CO_2 on the electrode surface.

In this study, Sn electrode was used as catalyst for highly selective production of formic acid. Nano-sized Sn was sprinkled onto the gas diffusion electrode (GDE) to increase the reaction interface between CO_2 and Sn catalyst. Catalyst loading was optimized by varying the Sn catalyst amount on the electrode. Fig. 1 shows a promising result of continuous ECC process with stable production of formic acid.

Experiments to optimize operating condition have been repeated in different temperature and applied voltages. Furthermore, to identify the carbon source of formic acid, ECC was performed during purging Ar and CO_2 isotope (C_{13}). The possibility of carbon conversion to formic acid from the catalyst or support material was investigated. Finally, stability in long term operation was also performed and produced formic acid was analyzed by UV-spectroscopy and HPLC.

References

1. J. Lee, Y. Kwon, R. L. Machunda, H. J. Lee, *Chem.-Asian J.*, **4**, 1516 (2009).
2. J. Wu, F. G. Risalvato, F. Ke, P. J. Pellechia, Xi. D. Zhou, *J. Electrochem. Soc.*, **159**, F353 (2012).
3. Y. Kwon, J. Lee, *Electrocatal.*, **1**, 108 (2010).
4. R. L. Machunda, H. Ju, J. Lee, *Curr. Appl. Phys.*, **11**, 986 (2011).
5. R. L. Machunda, S. Lee, H. Ju, H. Jeon, J. Lee, *Energy Environ. Sci., submitted* (2012).

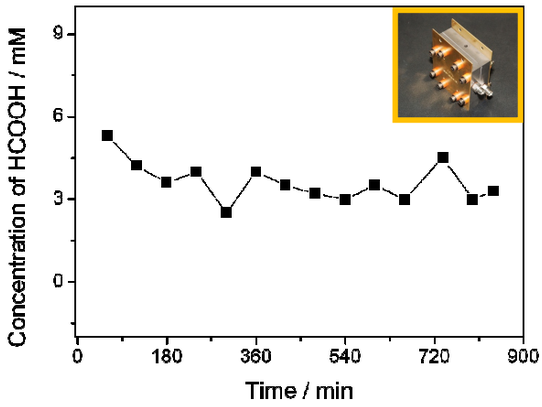

Fig. 1. Long term stability of electrochemical conversion of CO_2 process for zero-gap cell with Pt/C anode in H_2 (20 mL min^{-1}) and nano-sized Sn cathode in CO_2 (100 mL min^{-1}). Applied cathodic potential = -1.6 V (*vs.* NHE) at 25 ℃. The concentration of formic acid was measured every hour. Inset image is the photograph of home-made zero-gap ECC cell.