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Effect of sulfur dioxide concentration on cell performance and sulfur dioxide crossover

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The sulfur dioxide gas is the pollutant gas released in various industrial processes. For example, in the copper refinery process, SO_2 gas is produced to reduce copper sulfide into copper. Most of SO_2 gas produced in the refinery process was converted into sulfuric acid by reaction 1 and 2.

$$2\,SO_2 + O_2 \rightarrow 2SO_3 \qquad (1)$$
$$SO_3 + H_2O \rightarrow H_2SO_4 \qquad (2)$$

To make sulfuric acid and hydrogen simultaneously by using industrial SO_2 off-gas, the SO_2 electrolysis process was proposed. Westinghouse Corporation developed the Hydrid-Sulfur (HyS) process to make hydrogen using sulfur dioxide and electrolyzer [1,2]. The electrochemical reaction has two reactions which are anodic and cathodic reaction.

$$SO_2 + 2H_2O \rightarrow H_2SO_4 + 2H{+} + 2e^- \qquad (3)$$
$$2H{+}\ 2e^- \rightarrow H_2 \qquad (4)$$

The overall HyS process converts water into hydrogen and oxygen by consuming energy. The first proposed process of Westinghouse used the sulfur dioxide dissolved in sulfuric acid. But, this process has significant diffusion resistance for sulfur dioxide. Therefore, Staser proposed a process that gaseous sulfur dioxide used as a feed [3]. After that, USC (University of South Carolina) studied on water and sulfur dioxide transport in the cell [4,5] .

Based on the HyS process, the simultaneous sulfuric acid and hydrogen production process was proposed in the metal refinery process. However, most of SO_2 off-gas in the refinery process contains the air mixed SO_2 because of oxidation process. If without separating air in off-gas, this mixed gas results in overpotential at the SO_2 electrolyzer and it will make more energy consumption. Some processes may be needed to separate the air in the off-gas and economical consideration may be needed.

The electrochemical oxidation studies about HyS process were performed, but the mixed gas effect in the electrolysis reaction was not reported. The objective of this study is to find out the effect of mixed SO_2 gas on the electrochemical cell performance.

Fig. 1 shows the cell voltage difference against SO_2 gas concentration at 0.1 A/cm^2. While the cell voltage increase based on Nernst equation was 25mV, the cell voltage difference was 85 mV between 20% and 100% SO_2 gas concentration. The reason of difference between experimental and theoretical data might be the increase of mass transport resistance due to mixed N_2 gas.

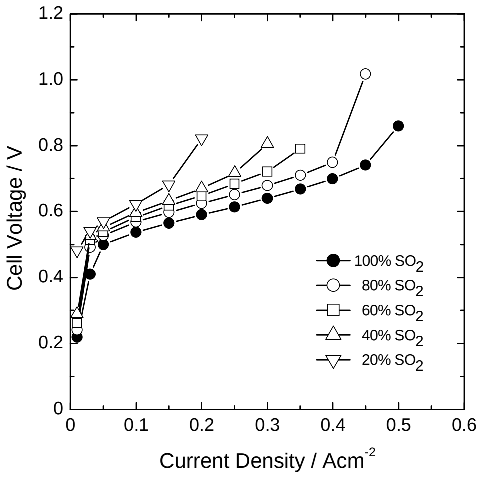

Fig. 1. Galvanodynamic polarization curves measured from the electrolyzer cell with various SO_2 gas concentrations at 90 °C.

Fig. 2 shows the crossover current density with SO_2 concentration change. The crossover current density was sharply decreased with time by about 50 seconds and then it kept stable within 600 seconds in the range of all SO_2 concentration. The crossover SO_2 was reduced with the change of SO_2 concentration. In this experiment, the proton which was produced on the water side was transported to SO_2 gas side and made hydrogen. The proton transport makes the phenomena of the electro-osmotic drag of water. However, the current density of this experiment was so small that the water transport and the SO_2 crossover reduction by electro-osmotic drag were negligible.

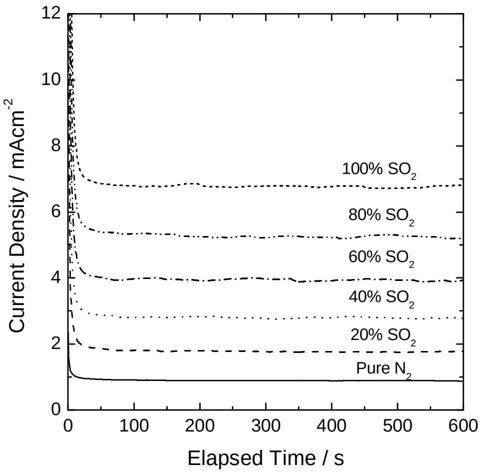

Fig. 2. Plot of current density against elapsed time measured from the electrolyzer cell with various SO_2 gas concentrations.

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