Characteristics of Anodic Polarization of Solid Oxide Fuel Cells under Pressurized Conditions Ryuji Kikuchi, Tatsuya Yano, Tatsuya Takeguchi, and Koichi Eguchi Department of Energy and Hydrocarbon Chemistry, Kyoto University Yoshida, Sakyo-ku, Kyoto 606-8501, Japan

One of the advantages of solid oxide fuel cell (SOFC) systems is their high conversion efficiency in power generation attained by direct conversion of fuels into electricity and thermal energy. It is widely recognized that SOFCs combined with a gas turbine system can achieve high efficiency. In such systems, SOFCs are operated at high pressure and polarization characteristics are dependent on operating conditions. It is, therefore, of importance to understand the anodic polarization of SOFCs operated under pressurized conditions. In this study the anodic polarization characteristics under pressurized conditions were investigated by AC impedance spectroscopy measured using a Ni-YSZ/YSZ half cell. AC impedance measurements were carried out at the temperature of 900 and 1000°C in  $H_2$ - $H_2O$  system with a constant H<sub>2</sub>/H<sub>2</sub>O ratio, or a constant partial pressure of H<sub>2</sub> or H<sub>2</sub>O for different total pressures of 1 to 10 atm.

Figure 1 shows AC impedance spectra measured for the total pressure of 1, 4, and 10 atm at the temperature of 900 and 1000°C and the oxygen partial pressure of 1.1 x  $10^{-18}$  and 4.4 x  $10^{-20}$  atm with an applied bias potential of 0 V and stimulus amplitude of 10 mV. The constant oxygen partial pressure for the different total pressures was attained by keeping the H<sub>2</sub>/H<sub>2</sub>O ratio constant according to the measurement temperatures. The impedance spectra in the Nyquist representation consists of two semicircles: one on the left side of the spectra has a high characteristic frequency of a few hundred Hz, and the other on the right side exhibits a characteristic When the range of the frequency of a few Hz. characteristic frequencies is considered, it is likely that the high frequency arc is attributable to the anodic reactions and that the low frequency semicircle is related to diffusion of H<sub>2</sub> and H<sub>2</sub>O to/from the reaction sites of the electrode.

At temperature =  $1000^{\circ}$ C and O<sub>2</sub> partial pressure = 4.4x 10<sup>-20</sup> atm, the semicircle with the high characteristic frequency is reduced and the arc at the low frequency is expanded as the total pressure was raised from 1 atm, up to 4 and 10 atm. The increase in the total pressure could enhance the collisional frequency of H<sub>2</sub> with the electrode and thus apparent reaction rate of H<sub>2</sub>, leading to the decrease in the resistance characterized by the high frequency arc. As the total pressure is increased, the partial pressure of H<sub>2</sub>O near the electrode surface can be raised due to the enhanced reaction rate as well as the low diffusivity of H<sub>2</sub>O, resulting in the increase in the oxygen partial pressure in the vicinity of the triple phase boundary. Consequently, the pressure gradient of O<sub>2</sub> between the reaction sites and the bulk is enlarged, that is, the concentration polarization becomes significant, as represented by the increase in the arch of the low frequency component in the Nyquist plot.

The comparison of the impedance spectra taken at the different  $O_2$  partial pressures with a constant temperature indicates that the semicircle at the low frequency is larger

at the low  $O_2$  partial pressure, and that the effect of the total pressure on the variation of the low frequency arc is more evident at the low O<sub>2</sub> partial pressure than at the high O2 partial pressure. These results mean that the concentration polarization of the anode is more significant at low O<sub>2</sub> partial pressures, and that H<sub>2</sub>O produced by the anodic reactions is expected to contribute substantially to the formation of the concentration polarization at low O<sub>2</sub> partial pressures, especially in the situations close to open circuit condition. When the temperature is varied with constant O<sub>2</sub> partial pressure, a distinct change in the impedance spectra appears only in the high frequency arc; the high frequency semicircle is expanded as the temperature is decreased, while the low frequency arc is almost unchanged. This is consistent with the fact that the resistance of the anodic reaction is large at low temperatures and diffusivity is almost insensitive to the temperature change examined in this work.

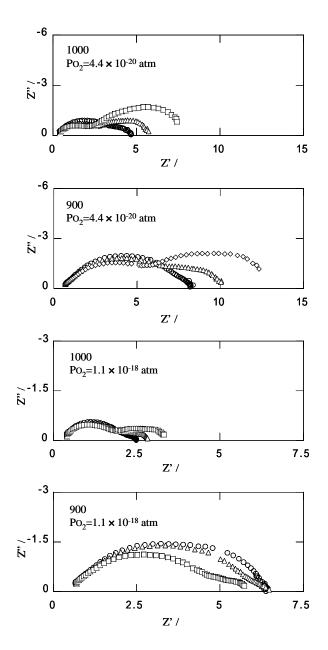


Fig. 1 Impedance spectra for Ni-YSZ/YSZ half cell. Cell: Ni-YSZ/YSZ/Ni-YSZ; atmosphere: H<sub>2</sub>-H<sub>2</sub>O; bias = 0 V; applied amplitude = 10 mV; total pressure: O 1atm,  $\triangle$  4 atm,  $\Box$  10 atm.