

IDENTIFICATION OF REACTION MECHANISM OF O₂ REDUCTION AT LSCF/GDC INTERFACE

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Recently much attention was paid to the development of solid oxide fuel cells (SOFC) operated at intermediate/low temperature such as 500-800°C. In order to reduce the increased electrolyte resistance with decreasing operating temperature, reduction in the electrolyte thickness and development of high ionic conducting electrolyte materials such as gadolinia-doped ceria (GDC) were adopted [1-4]. Beside that, the improvement of cathode performance and the development of cathodes with low overpotentials at low temperature become increasing urgent for the development of high performance intermediate temperature SOFC (IT-SOFC) [5].

Doped lanthanum cobaltite such as (La,Sr)(Co,Fe)O₃ (LSCF) have been considered as cathodes for intermediate/ low temperature SOFCs, especially with ceria-based electrolyte because of their high electronic and ionic conductivities as well as their high catalytic activities for oxygen reduction [6-8]. However, the exact oxygen reduction mechanism has not been clarified yet. The clarification of reaction mechanism could enable us to suggest the optimum interface for LSCF/ceria-based electrolyte. In this paper, the impedance of oxygen reduction at La_{0.8}Sr_{0.2}Co_{0.2}Fe_{0.8}O₃ (LSCF)/GDC interface under different oxygen partial pressures (from 1 atm to 0.0011 atm) and different temperatures was investigated. Results show that there are at least three reaction steps involved in oxygen reduction at LSCF/GDC interface.

Fig. 1 shows the impedance of oxygen reduction at LSCF/GDC interface measured at 800°C as a function of oxygen partial pressure. Two depressed arcs are observed under high oxygen partial pressure such as 0.21 atm and 1 atm. Under oxygen partial pressure below 0.21 atm, an additional arc appeared at very low frequency, the resistance of which increased with deceasing oxygen partial pressure. The polarization resistance corresponding to the high-frequency arc showed the weak dependence on oxygen partial pressure, which suggests the high-frequency arc is due to oxygen diffusion in the LSCF bulk cathode. The polarization resistance corresponding to the intermediate-frequency arc showed strong dependence on oxygen partial pressure, which suggests the low-frequency arc is due to dissociative adsorption of oxygen and/or surface diffusion of oxygen-containing species. The polarization resistance corresponding to the additional low-frequency arc is approximately proportional to the reverse of oxygen partial pressure, which suggests the additional low-frequency arc is due to mass transfer of bulk oxygen in porous LSCF cathode.

Fig. 2 shows the impedance of oxygen reduction at LSCF/GDC interface measured at 800°C for two mixed

gas: N₂-O₂ and He-O₂. The oxygen partial pressure was 0.01 atm and 0.0011 atm. The additional low-frequency arc can be seen to change noticeably in N₂-O₂ and He-O₂ under oxygen partial pressure of 0.0011 atm, which supports that the additional low-frequency arc is due to mass transfer of oxygen in bulk LSCF cathode. The polarization resistance corresponding to the additional low-frequency arc is reduced significantly in He-O₂ because of higher gas phase diffusion coefficient in He-O₂ than in N₂-O₂.

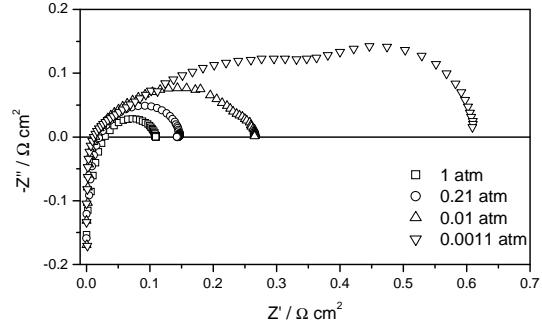


Fig. 1 Dependence of impedance spectra measured at 800°C of LSCF electrode on oxygen partial pressure.

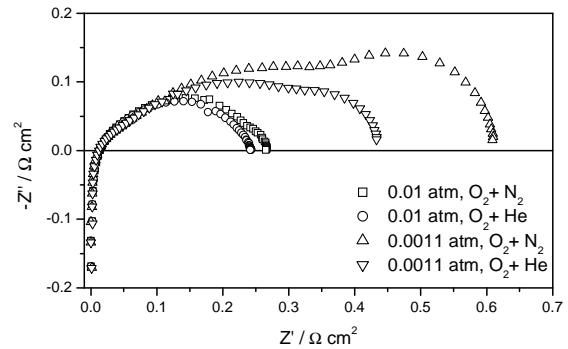


Fig. 2 Impedance spectra measured at 800°C of LSCF electrode with different balance gases.

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