Effect of Additive to Ni-YSZ Cerment on Reorming of CH_4 and Electrochemical Activity for SOFC

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Power generation by solid oxide fuel cells (SOFCs) is one of the most attracting energy conversion systems because of high efficiency, low pollution and multi-fuel compatibility. The high operating temperature gives rise to excellent fuel flexibility, which allows to reform hydrocarbon fuels on the anode internally in a SOFC module. Such simplified internal reforming operation of SOFC system results in low costs owing to the elimination of pre-reformer. Natural gas is regarded as a relatively cheap and popularly available fuel, which is suitable for SOFC. However, internal reforming of hydrocarbon often accompanies carbon deposition. The active sites of the anode are covered with deposited carbon, resulting in the deactivation, loss of cell performance and lower SOFC reliability

Therefore, in this experiment, alkaline earth metal oxides and precious metals were added to the Ni-YSZ cermet, and the relation between steam reforming of CH_4 and the electrochemical activity was investigated. Before the experiments, equilibrium partial pressure of oxygen and the boundary of carbon deposition region were calculated in the C-H-O phase diagram at temperatures ranging from 400 to 1000°C. These calculations suggested that the development of the anode catalyst without carbon deposition was one of the most promising ways to achieve high efficiency in SOFC because the amount of added water could be reduced.

First, the effect of MgO, CaO, SrO and CeO_2 addition to Ni-YSZ cermets on their catalytic activity and carbon deposition was investigated. Although, the CaO addition slightly deteriorated the electrochemical activity as anode, the CaO addition was effective in suppressing carbon deposition and promoted steam reforming of CH_4. Second, the effect of Ru, Rh, Pt, and Pd additions was investigated. The open circuit voltage for the cells with precious metal-modified Ni-YSZ cermets was 0.90 V, which almost agreed with the expected value. As shown in Fig. 1, Clearly Ru and Pt additions promoted the electrochemical activity as anode, indicating that promotion of reforming and suppression of coke deposition is important for SOFC generation. This also suggested that these precious metals effectively functioned as the anode catalysts.

Impedance spectra for anode/electrolyte interface of fuel cells using Ni-YSZ cermets modified with Ru and Pt were shown in Fig. 2. The impedances are caused by gas diffusion at around 20 Hz. These impedances were greatly reduced by modification with Ru and Pt, since gas diffusion in the Ni-YSZ was rapid owing to no coke formation. These dada means that stability of the abode catalyst of SOFC was considerably high.

In conclusion, it is of primary importance for the investigation of internal reforming of SOFC to evaluate the thermodynamic equilibrium concerning on carbon deposition and oxygen potential in multi-component gas in reformate. The additive to the cermet significantly affected the kinetic parameters of steam reforming of CH_4 on Ni-YSZ cermet. CaO-modified Ni-YSZ cermet was effective in suppressing carbon deposition without deteriorating the reforming activity though the addition of CaO slightly deteriorated the cell performance. Ru and Pt additions promoted the electrochemical activity as anode, indicating that these precious metals effectively functioned as the anode catalysts. The impedances caused by gas diffusion were greatly reduced by addition of Ru and Pd, and this contributed to the high performance and stability of the abode catalyst of SOFC.

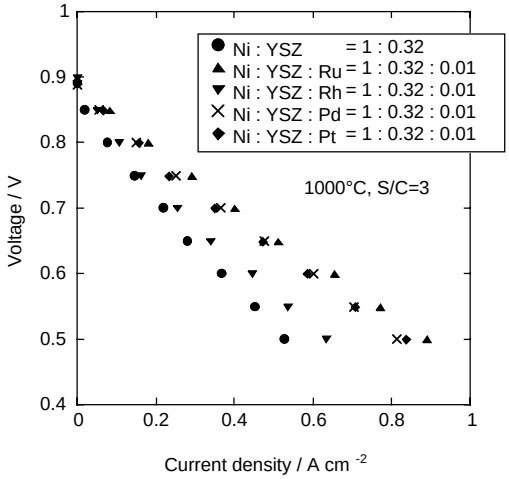

Fig. 1. Current-voltage characteristics in SOFC operation using precious metal-modified Ni-YSZ cermets at 1000°C. Anode gas: 40% N_2, 15% CH_4, 45% H_2O; Flow rate, 150 STP ml/min; Cathode gas: O_2; Flow rate,150 STP ml/min

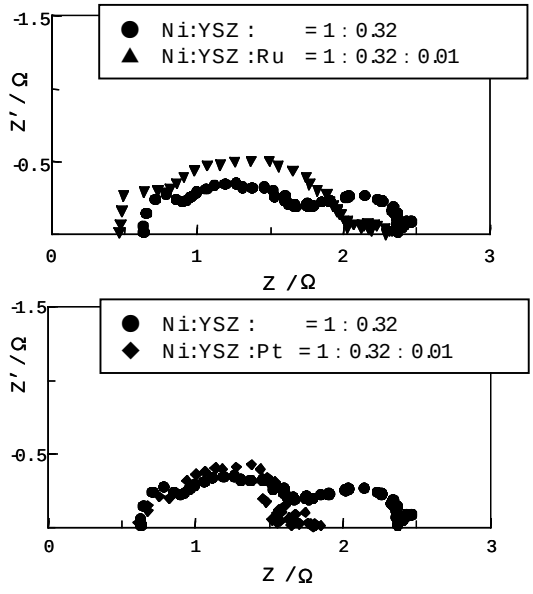

Fig. 2. Impedance spectra for anode/electrolyte interface of SOFC using Ni-YSZ cermets modified with Ru and Pt. 1000°C; Steam/Carbon = 3; Bias= Open circuit voltage.