

## Fe-containing Lanthanum Nickel Oxide as the Cathode for the Improvement of Solid Oxide Fuel Cell Performance

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Partial substitution of iron for nickel in  $\text{LaNiO}_3$  stabilizes the perovskite phase with an increase in the electronic conductivity. We found that the  $\text{LaNi(Fe)O}_3$  (LNF) cathode combined with a 10 mol%  $\text{Sc}_2\text{O}_3$ -1 mol%  $\text{Al}_2\text{O}_3$ -89 mol%  $\text{ZrO}_2$  (SSAZ) electrolyte showed higher solid oxide fuel cell (SOFC) performance than the conventional  $\text{La}_{0.6}\text{Sr}_{0.4}\text{MnO}_3$  cathode. Crystalline phase stability and reactivity with the SSAZ electrolyte were found to affect the ability of the  $\text{LaNi}_{0.6}\text{Fe}_{0.4}\text{O}_3$  cathode at 800-1100 degrees Celsius. Cathodic overvoltage decreased with prolonged operation time at 800 degrees Celsius. Phase identification by X-ray diffraction and observation of the interfacial structure between the anode and electrolyte by scanning electron microscopy indicate an improvement of the charge transfer property occurring at the interface.

The Ni-SSAZ cermet anode was prepared by painting a mixture of NiO and SSAZ powders with a binder in an appropriate molar ratio to one face of the SSAZ disk and subsequent heating at 1300 degrees Celsius for 2 h in air. LNF powder with the binder was painted to the other face of the SSAZ disk and heated at 1000 degrees Celsius for 2 h in air. The platinum reference electrode was attached to a small portion of the cathode face. The SSAZ disk was 15 mm in diameter and 0.3 mm thick.  $\text{H}_2$  was supplied as the fuel and air was supplied as the oxidant. The cathodic overvoltage was measured with the current interruption method.

Fig. 1 shows characteristics of the fuel cell performance for the LSM and LNF cathodes. The  $I$ - $V$  curves in this figure imply a decrease of the activation overpotential of the LNF cathode. This decrease probably resulted in approximately 25% increase in the maximum power density for the LNF cathode compared with the LSM cathode. Fig. 2 shows a comparison of the cathodic overvoltage between the LSM and LNF cathode under the condition that the cell was continuously discharged at  $78 \text{ mA cm}^{-2}$  except for the overvoltage measurement. The overvoltage for the LSM cathode was higher than that of the LNF cathode at least up to 60 h. The overvoltage of LNF was gradually decreased with increasing cell operation time, whereas that of LSM was increased. We could not identify any morphological changes at the interface between the LNF cathode and SSAZ electrolyte with a scanning electron microscope. Phase identification by X-ray diffraction for the powder mixture of LNF and SSAZ after heat treatment above approximately 1000 degrees Celsius revealed that the solid-state reaction between these oxides produces the  $\text{La}_2\text{Zr}_2\text{O}_7$  phase, which increases the interfacial resistance.

These results indicate that activation of the cathodic reduction of oxygen occurs near the interface between the LNF cathode and SSAZ electrolyte. Substitution of Fe for Ni in  $\text{LaNiO}_3$  could affect the defect structure of the perovskite phase, which results in an increase in the incorporation of oxygen atoms into the LNF from the gas phase and the subsequent reduction of oxygen atoms into oxide ions. Iron ions can exist in the divalent or trivalent state in oxides, however nickel ions exist only in the divalent state. This difference can explain the decrease of the activation overpotential for the LNF cathode. Fig. 3 shows Arrhenius plots of the inverse polarization resistance for the LNF cathode. Prolonged cell operation time resulted in a slight increase in the apparent activation energy for the cathode reaction. This indicates that the rate-determining step is affected by the total amount of transported oxide ions into the electrolyte. Fine structural change of LSM near the LSM-SSAZ interface could account for this phenomenon.

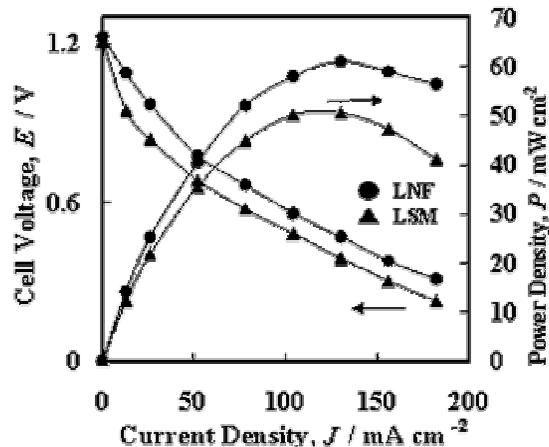


Fig.1  $I$ - $V$  and  $I$ - $P$  curves at 800 degrees Celsius.

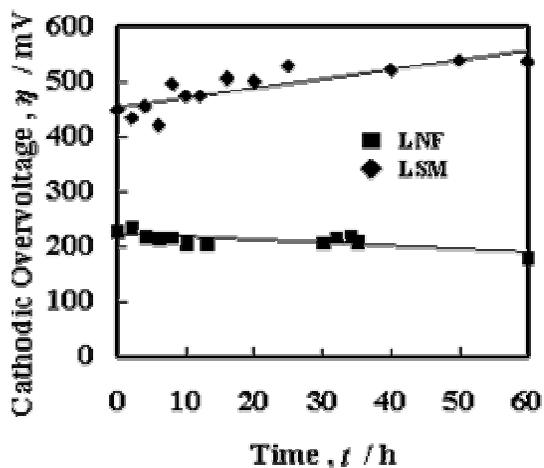


Fig.2 Cathodic overvoltage as a function of time at 800 degrees Celsius.

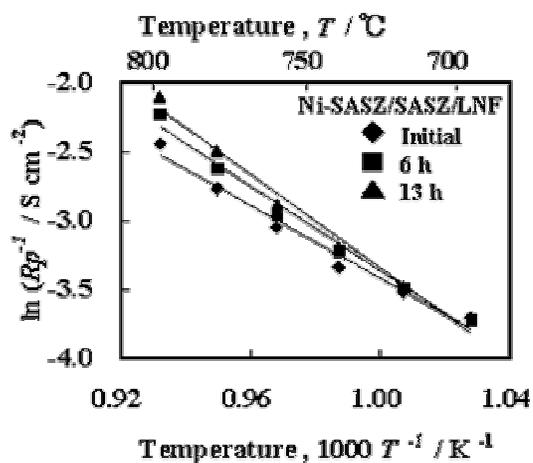


Fig.3 Arrhenius plots of the inverse polarization resistance