Hydrogen Permeation of Fe-16Cr Alloy in the Atmosphere Simulating SOFC at 1073K

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INTRODUCTION

It is expected that Fe-Cr heat-resistant alloy is applied to interconnect of planar-type solid oxide fuel cells (SOFC). In the previous work of our group, the oxidation kinetics of Fe-16Cr alloy (SUS430) has been studied in the conditions simulating the anode and cathode environments in SOFC respectively[1]. Gas tightness is one of the most important properties which interconnect requires. It's well known that diffusion of hydrogen in metal and alloy are fast. In this study, the hydrogen permeation and oxidation kinetics in the early stage of Fe-16Cr alloy (SUS430) has been studied in air/fuel atmosphere under oxygen and hydrogen potential gradients at 1073 K.

EXPERIMENTAL

Experimental setup for oxidation under air/fuel atmosphere is illustrated in fig. 1. The sample is SUS430 (Fe-16Cr) films of 0.02 mm thick. The samples were cut into the size of 15x15 mm. After ultrasonically cleaned, samples were set at the isothermal zone of oxidation furnace. Sample temperature was up with the rate of 5 K/min, and held at 1073 K for 10.8, 86.4 ks, and after that temperature was down with the rate of 5 K/min. The gaseous mixture of hydrogen/water vapor (H₂/H₂O=97/3) was flowed in the one side of sample, and Ar+21%O2 (dewpoint: less than -40 degree centigrade) was flowed in the other side of sample with 100 ml/min flux. Permeation area is 64 mm². Sample and ceramics cylindrical probe were sealed by two Pyrex glass rings with the size of 15 mm diameter, 1~2 mm thickness. Oxygen partial pressure of gas was measured by zirconia oxygen sensor at the inlet and outlet of fuel side. The reaction furnace was prepared to burn hydrogen completely into water at the outlet of Ar+O₂ side. Water vapor partial pressure of the gas was measured continuously by chilled mirror dew point hygrometer in order to evaluate the hydrogen permeability.

The phase composition of oxide scales of both sides (air/fuel) was identified by X-ray diffraction (XRD). Microstructures and composition of scale were analyzed with scanning electron microscope (SEM) and electron probe x-ray micro analyzer (EPMA).

Prior to evaluation of the hydrogen permeation through SUS430, test measurement was done with Palladium film to confirm properties and accuracy of this apparatus. Palladium has high permeability of hydrogen and that value is well known.

RESULTS & DISCUSSION

Figure 2 shows the results of test measurement. Glass seal was completed at about 950 K and dew point indicates hydrogen permeability quantitatively. Figure 3 shows the change of dewpoint in the early stage of oxidation of SUS430. Decreasing of dew point at about 850 K and drastic decreasing of dew point at about 970 K indicate decreasing of hydrogen permeability. Comparing with theoretical value (Fe without scale) calculated with reported solubility[2] and diffusion coefficient[3], hydrogen permeation of Fe-Cr alloy is apparently decreases with formation of oxide scale.

The peaks of Fe_2O_3 and Fe_3O_4 were detected in the X-ray diffraction pattern in the early stage of oxidation at 873 K. In addition to this, iron oxides nodules were observed on the surface of alloy in SE images and composition maps. Dense scale consists of Cr_2O_3 and $MnCr_2O_4$ spinel was observed on the sample exposed for 10.8 ks at 1073 K.

From these results, hydrogen permeation of actual operation was estimated. In the case of actual operation (thickness of interconnect = 0.1mm), fuel loss is less than 0.01 % after several ks elapsed.

CONCLUSION

Formation of dense scale consists of Cr_2O_3 and $MnCr_2O_4$ spinel decreases drastically hydrogen permeability at 970 K, though the thickness of scale is very thin. These results indicate that oxide scale has a low hydrogen permeation to assure the effective operation of SOFC.

REFERENCES

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Figure 3. Dewpoint change of Ar+O₂ side of Fe-Cr alloy.