Oxygen Permeability in Gold by Using High Temperature Oxidation of Gold Alloy

Kenichi Kawamura, Tomohiro Kajino, Makoto Nanko,* and Toshio Maruyama

Tokyo Institute of Technology, 2–12–1 Ookayama, Meguro-ku, Tokyo 152–8552, JAPAN.

* Nagaoka University of Technology 1603–1 Kamitomioka-machi, Nagaoka, Niigata, 940–2188, JAPAN.

INTRODUCTION

 ${
m LaCrO_3}$ ceramics system is a candidate for a separator of solid oxide fuel cell (SOFC), which separates between anode and cathode atmosphere and connects unit cells to boost voltage of the cell. When we use a heat resisting alloy instead of LaCrO₃ ceramics, SOFC become having a mechanical strength and reducing a cost. Recently, operating temperature of SOFC are tried to decrease bellow 1073 K for developing a durability and a mobility. The decrease in the operating temperature decreases a rate of oxidation of an alloy, and reduces an effect of electrical resistivity of scale formed on the surface of the alloy. Therefore, the SOFC operated at middle and/or low temperature would use the heat resisting alloy as the separator. Since thermal expansion coefficient of Fe-Cr ferritic alloy is similar to that of stabilized ZrO₂ electrolyte, the Fe-Cr ferritic alloy is candidate for the separator.

Under the both electrode condition, the Fe–Cr ferritic alloy is oxidized and forms $\mathrm{Cr}_2\mathrm{O}_3$ scale. $\mathrm{Cr}_2\mathrm{O}_3$ scale volatilize as $\mathrm{Cr}\mathrm{O}_3$ and $\mathrm{Cr}\mathrm{O}_2(\mathrm{OH})_2$, and condense on the electrode spoiling an electrode performance. Our group proposed that gold coating was effective to prevent this phenomenon.

When the alloy separator with $\mathrm{Cr_2O_3}$ scale is covered with Au , Cr dissolves into gold as $\mathrm{Cr_2O_3} \to 2\mathrm{Cr} + 3/2\mathrm{O_2}$ at $\mathrm{Cr_2O_3}$ scale/Au interface. The dissolved Cr diffuses by a chemical potential gradient between the interface of the $\mathrm{Cr_2O_3}/\mathrm{Au}$ and a surface of Au, and volatilizes at the Au surface. Therefore, the volatility of Cr depends on an activity of Cr at the $\mathrm{Cr_2O_3}/\mathrm{Au}$ interface, Since activity of $\mathrm{Cr_2O_3}$ formed as scale, is unity, activity of Cr depends on an oxygen partial pressure at the $\mathrm{Cr_2O_3}/\mathrm{Au}$ interface. The oxygen partial pressure is dependent on balance between permeating oxygen through Au and consumed oxygen by forming oxide. In present study, the oxygen permeability in Au was determined by using internal oxidation of Au alloy.

EXPERIMENTAL

Au-0.42!\$.14 mol% Ni and Au-0.07!\$.45!\$.69 mol% Cr alloys were prepared by melting technique. It confirmed that the sample alloy was comprised of the homogeneous single phase of Au without oxide. The samples were oxidized at 1073-1273 K for 86.4-2419.4 ks in air. After oxidation, the sample was evaluated by X-ray diffraction (XRD), SEM, electron micro probe analyzer (EPMA).

RESULTS AND DISCUSSION

After the oxidation of Au–Ni alloy, spherical NiO formed as internal oxide. $\rm Cr_2O_3$ was formed as spherical or columnar internal oxide in Au–Cr alloy. Figure 1 shows the typical relation between oxidation time and thickness of the internal oxidation zone (IOZ). The thickness of the IOZ increased parabolically.

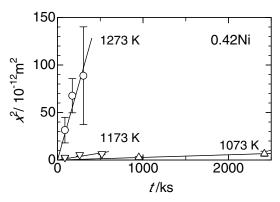


Figure 1

The thickness of internal oxidation layer as a function of time in Au-0.42Ni.

Wagner[1] indicated a relation between the oxidation time, t, and thickness of IOZ, x, as follows.

$$x^2 = 2 \frac{N_{\rm O} D_{\rm O}}{\nu N_{\rm (Ni \ or \ Cr)}} t \tag{1}$$

The parabolic rate constant, $k_{\rm p}$, is represented by

$$k_{\rm p} = \frac{N_{\rm O}D_{\rm O}}{\nu N_{\rm (Ni~or~Cr)}} N_{\rm O}D_{\rm O} \tag{2}$$

Figure 2 shows a oxygen permeability, $N_{\rm O}D_{\rm O}$ using the obtained $k_{\rm p}$. The activation energy of the $N_{\rm O}D_{\rm O}$ in Au is similar to those in the other fcc metals.

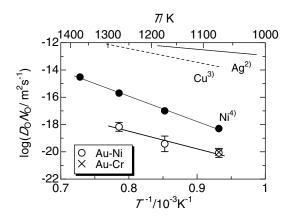


Figure 2

The oxygen permeability in fcc metals as a function of temperature. [2,3,4]

REFERENCES

- 1. C. Wagner, Z. Elektrochem., 63 (1959) 772
- 2. J.-K. Park, Mater. Lett., 9 (1990) 313-316
- 3. R. L. Pastorek and R. A. Rapp, Trans. Metal. Soc. AIME, 245 (1969) 1969–1711
- 4. F. H. Stott et al., Solid State Ionics, **12** (1984) 365-374