THERMODYNAMIC AND SIMS ANALYSES ON CORROSION OF SOFC METAL INTERCONNECTS

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Corrosion of metal interconnects has to be surmounted for the establishment of the intermediate temperature SOFCs with a variety of applications. We have recently started the investigation on corrosion of metal interconnects by using the SIMS technique to know the mass transfer of intentionally added components or impurities elements in ferritic alloys. Focus is placed on behavior in the early stage of the oxide scale formation to examine more precisely the mass transfer of elements. For this purpose, well-polished surface was used and oxidized in a mixture of simulated anode gases.

Typical experimental results are given in Fig. 1. Significantly fast diffusion and oxidations are observed for manganese, lanthanum, silicon and alumina. Oxide scale formed in the early stage was in a layer structure and was stably grown keeping the same layer type. Alumina appeared as the internal oxidized grains. Silica appeared between the layered oxide scale and the alloy matrix containing alumina internal grains.

Those behaviors have been analyzed from the thermodynamic point of view in several manners.

First attempt is to compare with the oxygen partial pressure for oxidation of respective elements. As shown in Fig.2, the elemental distribution can be roughly interpreted in terms of the sequence of oxidation; that is, alumina appeared as deepest part, then silica appeared and Cr, Fe, Mn forms the outer oxide scale. There are some exceptional elements. For example, La moved to the surface region although it is quite active metal. On the other hand, nickel stayed inside.

Second attempt was made to take into account the tendency of forming double or complex oxides. The most striking feature in Fig.1 is that respective elements behave differently. To account for this behavior, chemical equilibrium calculations were made to simulate the effects of oxygen partial pressure by making sequential calculations by introducing a small amount of water stepwisely. Result equilibrium reaction products are plotted as a function of equilibrated oxygen partial pressure in Fig. 3. This shows that alumina and silica can be formed separately without forming aluminosilicate. In addition, double oxides between transition metal oxides and silica/alumina can be formed; this is not realized in Fig.1.

These features strongly suggest that the elemental distribution cannot be properly interpreted without consideration on diffusion. For such a purpose, the chemical potential diagram is the most appropriate. However, it is impossible to construct those for the 9 element system. Instead, Fig. 4 shows the chemical potential diagram for the Cr-Fe-Mn-O system in the three dimensional space. By taking account of the feature that the reactive diffusion path should be simple in the chemical potential diagram, plausible reactive diffusion path can be drawn in Fig. 4 for the elemental distribution in the outer oxide scale by adopting that Mn and Fe are faster than Cr in the spinel phases.

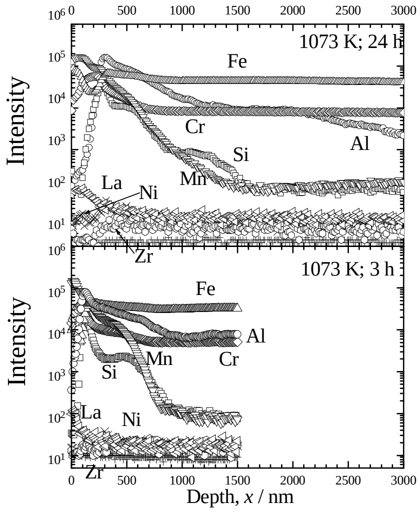

Fig.1 Elemental distribution of alloy elements after oxidation of ZMG232 alloy in anode atmospheres.

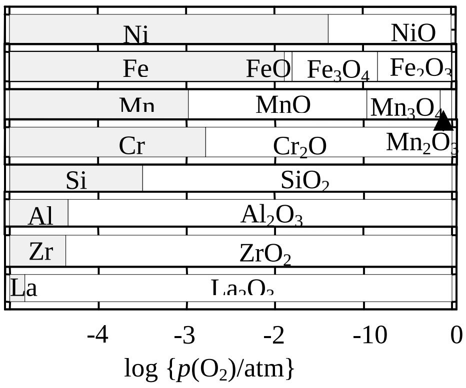

Fig. 2 oxidation potential of respective elements

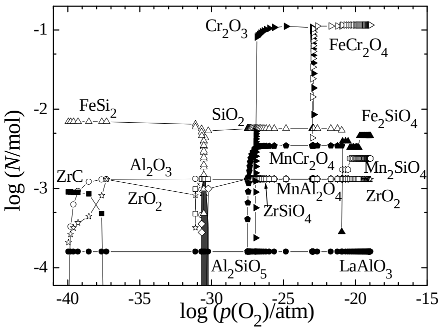

Fig. 3 Results of a series of chemical equilibrium calculations by adding water in step-wisely increasing amount.

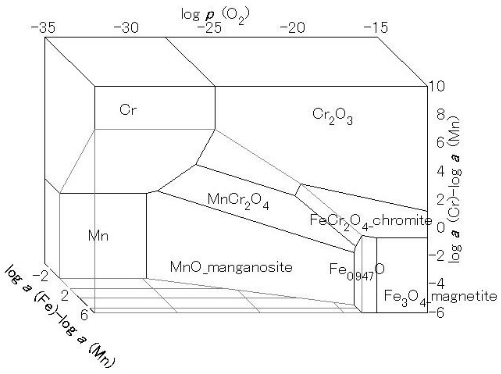

Fig. 4 Generalized chemical potential diagram for the Cr-Fe-Mn-O system at 1073 K.