



Review on Oxidation Behavior and Chromium Volatilization of Fe-Cr-Based Interconnects at High Operation Temperatures of Solid Oxide Fuel Cells

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ABSTRACT

Reduction of operating temperature to the range of 600 - 800 °C allows metallic alloys to be considered as solid oxide fuel cells (SOFCs) interconnects. Of all SOFC components, interconnects have the most stringent criteria as they are required to work in both oxidizing and reducing conditions. To date, Fe-Cr alloys are the most promising candidates owing to their good oxidation resistance, electrical conduction, and matching thermal expansion coefficient with SOFC components. Unfortunately, the alloys are susceptible to the formation of oxide scales which are $(\text{Mn,Cr})_3\text{O}_4$ and Cr_2O_3 at elevated temperatures. Moreover, thermodynamic instability in the cathode environment causes Cr volatilization from the scales, which subsequently cause cell degradation. The volatilization can be retarded with the use of protective coating or specially designed alloys with tailored compositions that enhances Laves phase precipitation. The oxidation behavior of the metallic interconnects, Cr volatilization, and their relationship with the composition of metallic alloys will be discussed.

Keywords:

SOFC; Interconnect; ferritic stainless steel; oxide scale; Cr volatilization

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1. Introduction

Solid oxide fuel cells (SOFCs) produce electrical energy through electrochemical reactions between air and fuel (e.g., H) and thus potential sustainable power sources [1–3]. SOFCs have three main components, namely, anode, cathode, and electrolyte, each of which uses different materials and has a distinct function. Cells are connected by interconnects, which build stacks to fulfill the power requirement of an application (Figure 1). Interconnects conduct electricity between cells and act as barriers to separate air and fuel in cathodes and anodes.

The successful reduction of the high thermal operating temperatures of SOFCs to 600–800 °C has enabled the replacement of conventional ceramic interconnects with metal alloys, which demonstrate better mechanical strength, electrical conductivity, and manufacturability than

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ceramics [4–6]. SOFCs must have at least 40 000 h of lifetime for them to be used in an application [7,8]. Therefore, interconnect materials should have good chemical stability in both oxidizing and reducing conditions, excellent oxidation resistance at high temperatures, excellent electrical conductivity, and good thermal conductivity, and compatible thermal expansion coefficient (TEC) with adjacent components [9,10].

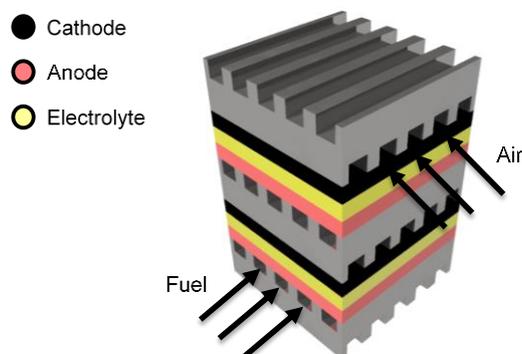


Fig. 1. Illustration of Solid Oxide Fuel Cell (SOFC) components

Among the potential candidates, Ni-Cr-based alloy and Fe-Cr-based alloy are of particular interest and widely studied in the past decade. Figure 2 shows the number of publications related to Fe-Cr and Ni-Cr-based alloys for the past 10 years. Fe-Cr is preferred compared to Ni-Cr because of its economic cost and better TEC matching with other SOFC components because of its body-centered cubic structure [11]. The material forms dual-layer oxide scales, when exposed to high thermal operation for a long period namely, Cr_2O_3 and $(\text{Mn,Cr})_3\text{O}_4$, which provide oxidation resistance and conduct electricity [12]. However, the incompatibility of TEC of the scales with metal alloys lead to cracks and spallation, which increase the electrical resistance (Figure 3) [13]. The scales also undergo Cr volatilization, which blocks the active area of cathode and degrade the cells. Thus, the Fe-Cr material with low oxide scale growth rate is preferred to be used as interconnects.

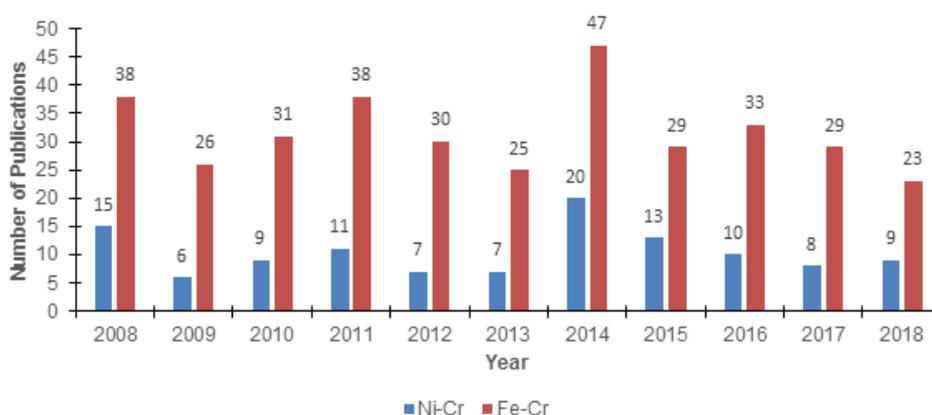


Fig. 2. Number of publications on different types of metallic interconnect in the past 10 years (Keywords: “SOFC,” “interconnect,” “Ni-Cr,” and “Fe-Cr” in <http://www.sciencedirect.com>, March 2018)

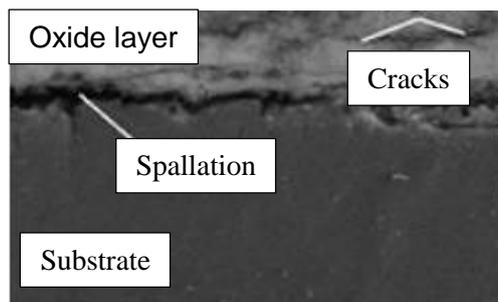
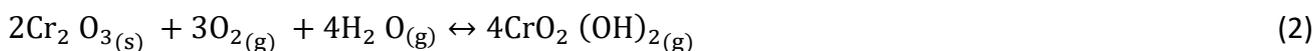


Fig. 3. Scanning electron microscope image of Fe-Cr specimen after 61 h of oxidation at 800 °C. The specimen shows cracks and spallation of oxide scale. Reproduced with permission [14]

2. Oxidation Behavior and Cr Volatilization

Oxide scales and Cr volatilization are closely related to each other. Cr volatilization occurs when gaseous Cr species forms due to thermodynamic instability of O at the cathode environment. Given that reactions depend on the O partial pressure and water content [15], Cr poisoning at the anode can be neglected due to rich H gas and low O partial pressure [16]. The reactions between the scales and O gas can be represented through the following equations [17].



The volatilization of Cr in oxide scales causes Cr migration to cathode which can be hastened by increasing partial pressure [18,19]. The migration causes cathode poisoning because of gaseous Cr species deposition at the triple phase boundary, which blocks the cathode active area and leads to cell degradation [20,21]. The volatilization is also closely related to the oxide scales microstructure where Cr_2O_3 is the inner layer, while the $(\text{Mn,Cr})_3\text{O}_4$ is the outer layer (Figure 4). The $(\text{Mn,Cr})_3\text{O}_4$ layer undergoes less Cr volatilization than the Cr_2O_3 layer, which is beneficial in enhancing oxidation resistance and preventing Cr migration from oxide scales [22–24]. Nevertheless, decreasing the growth rate of both layers may reduce Cr volatilization.

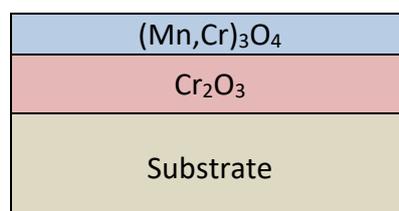


Fig. 4. Schematic illustration of oxide scales

The growth rate of the scales can be measured by determining the oxidation kinetics of the material. The value of oxidation kinetics value can be calculated using Eq. (4), as follows

$$\left(\frac{\Delta W}{A}\right)^2 = K_p t \tag{4}$$

where ΔW is the weight gain, A is the sample area, t is the oxidation time, and K_p is the oxidation kinetic. Weight gain shows how much scales were formed after several hours of oxidation. Figure 5 shows the oxidation kinetics for different Fe-Cr materials according to previous studies. FeCrCo exhibits the lowest oxidation kinetics of $1.42 \times 10^{-15} \text{ g}^2\text{cm}^{-4}\text{s}^{-1}$ [25], followed by Crofer 22H with $5.29 \times 10^{-15} \text{ g}^2\text{cm}^{-4}\text{s}^{-1}$ [25] and SUS 430 with $17.2 \times 10^{-15} \text{ g}^2\text{cm}^{-4}\text{s}^{-1}$ [26]. The large difference in weight gain between the three materials is due to the different chemical compositions, as tabulated in Table 1.

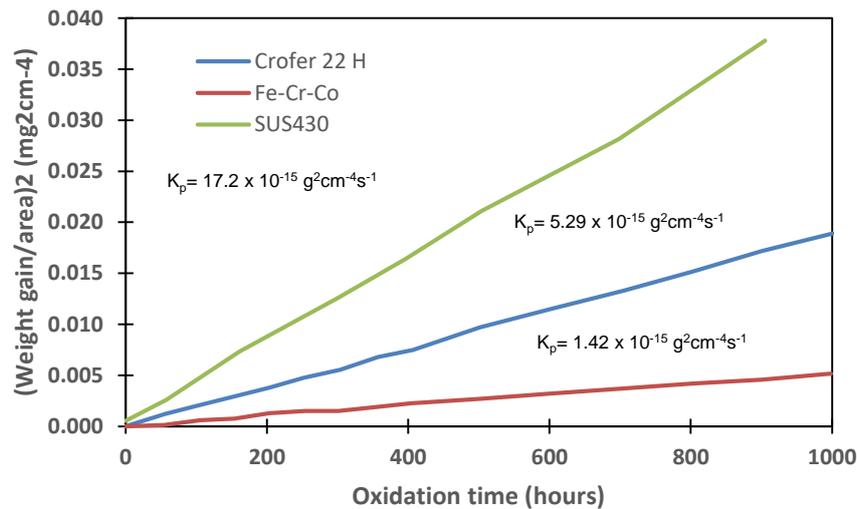


Fig. 5. Oxidation kinetic for different Fe-Cr materials at the temperature of 750 °C in air [25,26]

Table 1

Chemical compositions of FeCrCo, Crofer 22 APU, and SUS 430 (wt%) [25,26]

Material	Cr	Mn	Co	Nb	Zr	La	Y	
FeCrCo	12.44	0.57	9.68		0.024	0.0074	0.0014	
Crofer 22 H	20–24	0.3–0.8		0.2–1.0		0.04–0.20		
SUS 430	16.76	0.69						
Material	C	S	P	Si	N	W	Ti	Cu
FeCrCo	0.0031	0.0016	0.0088	0.032				
Crofer 22 H	<0.03	<0.006	<0.05	0.1–0.6	<0.03	1.0–3.0	0.02–0.20	<0.5
SUS 430	0.12			0.75				

Elements such as Mn, Y, La, Zr, and Ti promote oxidation resistance and contribute to the growth of a second oxide layer, that is, a $(\text{Mn,Cr})_3\text{O}_4$ layer that inhibits Cr volatilization [27–31]. As shown in Table 1, FeCrCo containing the aforementioned elements has lower oxidation kinetic than the other materials that do not have these elements or have lower content of these elements than FeCrCo. Meanwhile, materials that contain Nb, Ti, W, or Mo such as Crofer 22 H are considered as Laves-type materials due to the Laves phase precipitation of the elements near the oxide scales. Precipitation prevents O diffusivity into the oxide scales, which reduces the oxidation rate [32–35] (Figure 6). However, excess amounts of these elements can contribute to some disadvantages. For example, excess amounts of Mn and Ti causes faster oxidation rate and increases area-specific resistance [36–38]. Meanwhile, excess Laves phase elements lead to the formation of brittle intermetallic secondary phases, which degrade the impact toughness and corrosion resistance of alloys [14,39,40].

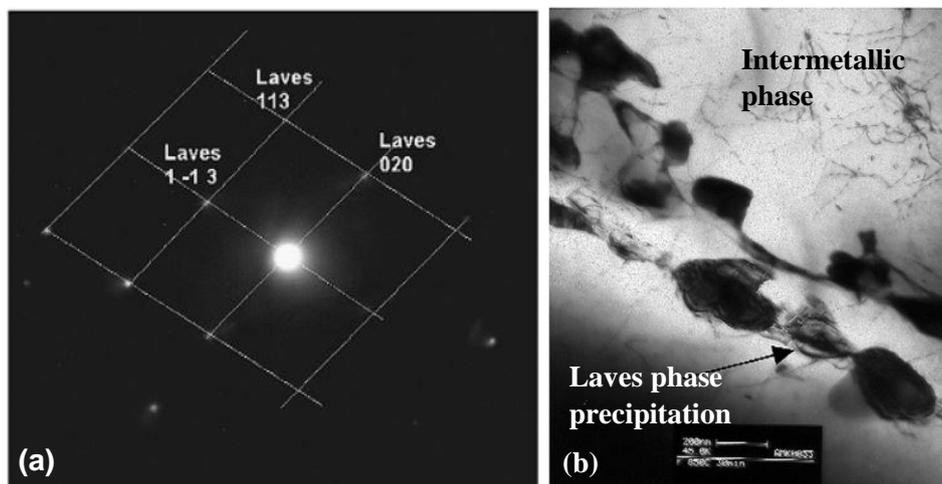


Fig. 6. (a) Selected area electron diffraction and (b) transmission electron microscopy of the Laves phase precipitation in 28% Cr–4%Ni–2%Mo–Nb superferritic stainless steel aged at 850 °C for 30 min. Reproduced with permission [39]

3. Recent Achievements

In previous years, researchers attempted to adopt spinel oxide properties into coating, which not only increases oxidation resistance and prevents Cr migration but also increases electrical conductivity with the presence of Co or Cu [41]. For example, Kumar *et al.*, [42] sputtered $(\text{Mn},\text{Co})_3\text{O}_4$ coating on SS 430, which not only reduces electrical resistance and increases oxidation resistance but also prevents inward O diffusion that promotes Cr volatilization. Talic *et al.*, [30] doped Fe into $(\text{Mn},\text{Co})_3\text{O}_4$ to further match the TEC of the coating with Fe-Cr materials. The results also show that denser coating can further reduce oxide scale growth and Cr volatilization. Sun *et al.*, [43] had coated $(\text{Mn},\text{Cu})_3\text{O}_4$ on Crofer 22 H, where the coating uses a Cu element instead of a Co element. In addition to having lower cost than $(\text{Mn},\text{Co})_3\text{O}_4$, this coating exhibits better electrical conductivity and eliminates the usage of toxic element such as Co [41].

Another method that also gains much attention is alloy design, which involves the alteration of the chemical composition of Fe-Cr materials, which can enhance oxidation resistance and electrical conductivity as well as reduce Cr volatilization [44–48]. Safikhani *et al.*, [14] found that the low Ti amount of 0.04 wt% and high Nb amount of 0.18 wt% in the Fe-Cr material can significantly increase oxidation resistance. The results of the study of Niewolak *et al.*, [49] showed that a high W content in Fe-Cr materials can strengthen the intermetallic phase, which increases the mechanical strength of the material. Ali-Löyty *et al.*, [50] added the reactive element Si into a Ti–Nb stabilized ferritic stainless steel (EN 1.4521) to promote Laves phase precipitation, which reduces Cr volatilization.

4. Conclusions

The formation of oxide scales and its growth rate is closely related to the composition of Fe-Cr-based interconnect. Cr migration is dictated by the behavior of oxide scales. Thus, the selection of right composition of Fe-Cr-based interconnect is the key factor in achieving high performance and excellent long-term stability. Apart from tailoring the composition of alloy to enhance oxidation resistance and electrical conductivity and effectively mitigate Cr migration, progressive efforts have been reported on the developments of coating materials to serve as a mass barrier, thereby retarding Cr migration in commercially available Fe-Cr-based alloys.

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