

An overview on oxidation of metallic interconnects in solid oxide fuel cells under various atmospheres

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ABSTRACT

This study explores the emergence of oxidation-resistant alloys as potential replacements for traditional ceramics in Solid Oxide Fuel Cells (SOFCs), specifically Ferritic Stainless Steel (FFS). Despite its promise, FFS encounters challenges such as oxidation and corrosion. Most research on FFS interconnect damage has primarily focused on high-temperature oxidation in single atmospheres. However, in practice, the interconnect is exposed to both oxidizing and reducing atmospheres concurrently, leading to enhanced oxidation known as the dual atmosphere effect. The significance of this phenomenon is increasingly recognized by researchers and industry experts, yet understanding its implications for FFS degradation and protective measures remains a young discipline. This article provides an overview of the oxidation mechanisms of FFS under various conditions, proposed mechanisms, and potential protective strategies to address the effects of the dual atmosphere.

1. Introduction

To address the growing global demand for clean energy, environmentally friendly and highly efficient energy conversion devices are needed. As an all-solid-state chemical power source, Solid Oxide Fuel Cells (SOFCs) directly convert the chemical energy stored in fuels (including hydrogen and hydrocarbons) into electrical energy through catalytic reactions on electrodes [1]. SOFCs have many advantages, including high energy conversion efficiency (up to 75% [2,3]), fuel flexibility, low environmental impact, and all-weather distributed power generation [4,5].

Commonly, a single cell in SOFCs consists of a cathode, anode, and electrolyte, as shown in Fig. 1 (a). It can only generate potential at around 1 V [6]. This is insufficient to meet the energy and power requirements to support most real-world applications. To address this problem, multiple SOFCs will be connected in series as a whole stack to enhance the output power. To connect these SOFCs, an interconnect is needed, and it is the fourth vital component in modern SOFCs [7]. Solid Oxide Electrolysis Cells (SOECs) are similar to SOFCs in structure but operate in reverse, converting electrical energy into chemical energy to produce hydrogen and other valuable fuels [8]. In SOFCs, the main issue for interconnects is oxidation on the air side, which leads to the formation of oxide scales that reduce conductivity and affect long-term

durability [9]. In SOECs, the higher oxygen partial pressure during electrolysis accelerates degradation, causing oxygen buildup and increased material stresses, particularly at the interconnect-air interface [10]. These harsher conditions in SOECs make interconnects more susceptible to rapid corrosion and material failure. Thus, the behavior of the interconnecting material during SOFC and SOEC operation is different. There are various options for SOFC's interconnecting material including different kinds of ceramics and metal. The working temperature of the SOFCs has dropped from 1000 °C to a lower range (500–800 °C) because of the usage of solid-state materials for the cathode ($\text{La}_{x}\text{Sr}_{1-x}\text{FeO}_3$ and $\text{La}_{x}\text{Sr}_{1-x}\text{MnO}_3$ [11]) and electrolyte (yttria-stabilized zirconia (YSZ) [12]). This temperature reduction has made it possible to use interconnect components made of alloy materials rather than the original ceramic interconnect components. The usage of metal materials as interconnect shows a great advantage in cost-effectiveness because of less expensive manufacturing processes compared to ceramic material. Therefore, metallic alloys have become a popular choice for the interconnect in commercial SOFCs [9]. However, the susceptibility of metallic interconnects to oxidation and corrosion under SOFCs' specific operational conditions poses a substantial challenge to their durability and functionality [13–17]. To mitigate these issues, the development of protective antioxidant coatings when using these for alloys in this application has become a research focus [7,

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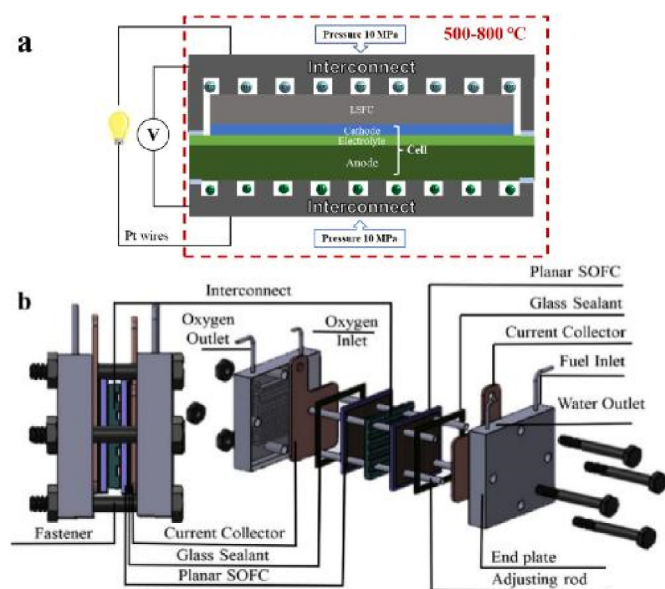


Fig. 1. (a) Single cell structure schematic diagram of a SOFC. (b) Stack structure schematic diagram of a SOFC [7].

17–20].

Over past decades, concerted global research efforts have led to substantial advancements in the stability and performance of SOFCs [1–5,7,17–20]. Critical to these advancements has been a deepened understanding of the interactions occurring at the interconnects and interfaces within SOFCs systems. This article provides an overview of the oxidation mechanisms of Ferritic Stainless Steel (FSS) for SOFC interconnects under various conditions, including existing theories, research methods, proposed mechanisms, and potential protective strategies to address the effects of dual atmosphere. In this paper, the degradation mechanisms of the metal interconnect under varying atmospheric conditions will be summarized. In particular, when the interconnect simultaneously encounters oxidising and reducing conditions on either side, the degradation of the alloy metal is more severe. This is defined as the "dual atmosphere" effect in the SOFCs. This working condition is common in the real SOFCs but a comprehensive discussion on the mechanism is missing in current literature.

The rest of the paper is organized in the following structure. Section 2 will revisit the basic concepts of interconnect materials used in current SOFCs, and the reason for the rise of FFS for the interconnect application. In addition, the oxidation and corrosion rates of common metallic interconnect alloys are listed and compared. In section 3, the mechanisms of degradation for the in dual atmospheres are described and summarized. In section 4, the various coating materials proposed for the interconnect protection will be summarized. As such, this paper provides a detailed overview of the mechanisms behind the currently proposed oxidation processes in different atmospheric conditions and introduces recent advances in protective coatings and their efficacy. We have systematically reviewed the literature from various studies, and have offered new insights into the oxidation mechanisms under dual atmospheres.

2. Interconnect materials

2.1. Introduction to interconnect material

The interconnect is located between the two electrodes in the stack, serving as both a bipolar plate and a separator. As such, it both conducts electricity and is necessary to separate the oxidizing and reducing gases, preventing them from mixing. This has implications for the required properties of the interconnect material, which must conduct electricity

while not degrading via electrochemical reactions. For long-term performance, the interconnect requires stable chemistry in both atmospheres. As a SOFC can be fueled with a wide range of fuels, including hydrogen, natural gas, and hydrocarbon fuels, this can be challenging, especially as all of these fuels may contain water and sulfide impurities that can damage the interconnect to varying degrees.

The interconnects make direct contact with the cathode and anode materials but are also connected to sealants between the cell and conductive materials for collecting and transmitting electrons, necessitating good compatibility with the surrounding environments. To enhance ionic conductivity through the solid electrolyte, actual working SOFCs stacks must operate at a high temperature (500–800 °C) for thousands of hours and withstand more than a hundred thermal cycles caused by the shutdown and startup processes [22].

Based on the SOFCs working environment, essential interconnect engineering/materials requirements include:

- Withstanding high temperatures (500–1000 °C) and possessing good oxidation and corrosion resistance (parabolic rate constant k_p value about 10^{-14} – 10^{-11} $\text{g}^2\text{cm}^{-4}\text{s}^{-1}$) at these temperatures [23];
- Maintaining high electrical conductivity (area-specific resistance (ASR) below $0.1 \Omega \text{ cm}^2$) even when oxide layer growth occurs on the surface during high-temperature operation [24];
- Having thermal expansion coefficient ($9\text{--}11 \times 10^{-6} \text{ K}^{-1}$) similar to those of electrode materials, sealing materials, and conductive materials to minimize stresses when thermally cycling [25];
- Providing good overall and interfacial thermo-mechanical reliability and durability [23];
- Exhibiting good chemical compatibility with surrounding materials to avoid chemical reactions [9];
- Minimizing the volatility of components that could damage the electrode material and so reduce cell stability and efficiency [9].

Ceramic materials; traditional alloy materials, especially FSS; and new high-temperature alloys are, to varying degrees, widely used for SOFC interconnects as these can fulfill the above requirements.

2.2. Alloy interconnect material

Additional desirable attributes for interconnect materials include low cost, wide availability, and simplicity of processing and manufacturing. These factors make metals more attractive than ceramic materials for SOFC interconnects. Although several metallic materials have been proposed, including Fe-based, Cr-based, and Ni-based oxidation-resistant alloys, as shown in Fig. 2, only a few, particularly FSS, have been used in real SOFC stacks [21]. Among these materials,

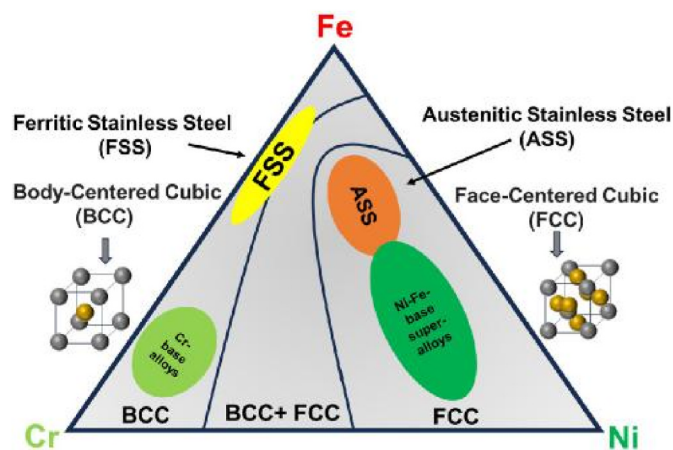


Fig. 2. Schematic showing chemistry dependence of possible phases in the Fe–Cr–Ni based alloys used as metallic interconnect materials [21].

Plansee's CFY alloy, a Cr–Fe–Y-based material, has seen real-world application in SOFCs interconnects due to their excellent oxidation resistance and durability under SOFC operating conditions [34]. Other alloy systems, such as austenitic stainless steels and Ni–Fe–Cr-based alloys, were initially explored, but they were not widely used in commercial SOFCs stacks due to higher costs and poor oxidation resistance [35].

Alloys used for SOFC interconnects, such as CFY and FSS, contain active elements like Cr, Al, and/or Si. As the resulting active element oxides form an additional layer, potentially inhibiting the electrochemical reaction, they must be understood. Cr₂O₃, for instance, acts as a semiconductor with an electrical conductivity ranging from 1.0 to 5.0 × 10^{−2} S cm^{−1} between 500 and 800 °C in air [36,37]. In contrast, Al₂O₃ and SiO₂ are insulators, but offer superior thermodynamic stability and exhibit much lower growth rates compared to Cr₂O₃, resulting in enhanced oxidation resistance [38].

The initial use of Cr-based alloys as alternatives to ceramic materials in SOFC interconnects was primarily due to their compromise of oxidation resistance high-temperature mechanical properties and conductivity. However, chromium diffusion towards the surface can create cavities at the oxide scale/metal interface, reducing the oxide scale's adhesion [39,40]. This challenge can be mitigated by incorporating active elements such as Y, La, Ce, and Zr, which slow down the chromium oxide growth rate and enhance adhesion by restricting outward chromium migration [41]. Nonetheless, chromium-based alloys, similar to other Group VIB metals, exhibit a ductile-brittle transition temperature significantly higher than SOFC's operating temperatures and are vulnerable to gaseous impurities; issues not addressable by adding active elements [38].

FSS, sharing a body-centered cubic (BCC) structure with Cr-based alloys, are currently the most widely used material because of their excellent oxidation resistance, compatible coefficient of thermal expansion (CTE) with ceramic materials, and lower production costs [42]. However, alloy CTE values are only readily available from manufacturers around room temperature. For temperatures above 500 °C, the range of interest for SOFCs, data is limited. Further results in this area are needed.

When considering manufacturability, oxidation resistance, compatibility, production challenges, and cost, ferritic stainless steel emerges as the most promising material for SOFCs interconnects. Table 1 presents a composition of commonly used ferritic stainless steel materials for SOFC metal interconnects.

2.3. Oxidation rate of common FFS

As diffusion through the oxide layer is the rate-controlling step, the growth of the oxide layer tends to follow a parabolic rate law (although other kinetics rates exist). This means that the growth rate slows down over time as the oxide layer becomes thicker [43]. A lower parabolic constant value is obtained for a lower rate of oxidation, meaning the oxide layer is more protective. The parabolic rate law is defined by the following equation:

$$(\Delta m)^2 = k_p t \quad (1)$$

where Δm is the mass gain per unit area, k_p is the parabolic rate constant, units are typically expressed as g²cm^{−4}s^{−1} or μm²s^{−1}, and t is time.

The k_p values of different interconnect materials in various atmospheres are compared in Tables 2 and 3. To facilitate data comparison, all values in μm²s^{−1} from the literature were converted to g²cm^{−4}s^{−1} using the 100% density of Cr₂O₃ (5.22 g/cm³ [61]). The data marked with an asterisk (*) in Table 2 and all values in Table 3 were converted using this method.

SUS430, a common stainless steel alloy, has gained widespread attention in SOFC interconnect and coating research. At 800 °C, SUS430 exhibits the formation of a dense Cr₂O₃ oxide film, enriched with MnCr₂O₄ and a minor presence of FeCr₂O₄, in both air and fuel atmospheres [45]. In a single atmosphere, specifically air at 800 °C, the k_p value for SUS430 is 2.09 × 10^{−13} g²cm^{−4}s^{−1} after 168 h of exposure (Table 2). When exposed to different gas atmospheres, such as pH₂/pH₂O = 97/3, the k_p values show slight variations but remain within the same order of magnitude. Under dual atmosphere conditions at 800 °C, with air on the oxidation side and a mixture of H₂ and 3% H₂O on the reduction side, the k_p value increases to 5.72 × 10^{−12} g²cm^{−4}s^{−1} (Table 3). In this instance, the performance of 430 in dual atmospheres shows increased oxidation rates compared to single atmosphere conditions. This suggests that, while SUS430 can be effective in single atmosphere environments, its long-term stability may be compromised under dual atmosphere conditions, making it less suitable for applications used in such environments.

Crofer 22 APU, a ferritic stainless steel alloy commercialized by ThyssenKrupp [62], forms a distinctive (Mn,Cr)₃O₄ spinel top layer and chromium oxide sublayer upon exposure [63]. In a single atmosphere, at 800 °C, Crofer 22 APU has a k_p value of 9.70 × 10^{−14} g²cm^{−4}s^{−1} or 3.56 × 10^{−7} m²s^{−1} after 500 h of exposure in air (Table 2). This demonstrates a lower oxidation rate compared to SUS430 without preoxidation under similar conditions. In dual atmosphere conditions at 800 °C, the k_p values for Crofer 22 APU are 1.23 × 10^{−12} g²cm^{−4}s^{−1} for non-preoxidized samples (Table 3). These values indicate better performance than SUS430 under dual atmosphere conditions. Crofer 22 APU exhibits superior high-temperature corrosion resistance in both single and dual atmospheres compared to SUS430. The pre-oxidation treatment further enhances its performance under dual atmosphere conditions, apparently making it a more reliable choice for SOFCs interconnect applications.

Developed by Hitachi Metals Co., Ltd., ZMG232 has shown promising performance as SOFCs interconnect material [64,65]. It is available in different grades: standard ZMG232 for high-temperature use in SOFCs, ZMG232L for increased strength, and ZMG232U for harsher conditions. In a single atmosphere, specifically air at 800 °C, ZMG232 has a k value of 1.76 × 10^{−13} g²cm^{−4}s^{−1} after 1000 h of exposure (Table 2). This value is comparable to Crofer 22 APU but higher than SUS430 under similar conditions. While ZMG232 shows good performance in single atmosphere conditions, the lack of data for dual atmosphere conditions limits the ability to fully assess its suitability as a SOFC

Table 1
Typical compositions of commonly used alloys for SOFCs interconnects.

Material	Concentration/wt.%														Ref.
	Fe	Cr	Mn	Mo	Ni	Si	Al	Ti + Nb	La	Zr	N	C	P	S	
SUS430	Bal.	16.14	0.209	–	0.044	0.279	0.11	–	–	–	–	0.048	0.028	0.0006	[26]
AISI430	Bal.	17.4	0.92	–	–	–	0.85	–	–	–	–	0.12	0.03	0.02	[27]
AISI441	Bal.	17.8	0.3	0.3	–	0.6	–	0.65	–	–	0.015	0.015	–	–	[28]
AISI444	Bal.	17.63	0.24	1.87	–	0.54	–	0.48	–	–	<0.02	0.013	<0.03	<0.02	[29]
Crofer 22 APU	Bal.	22.8	0.44	0.1	0.03	0.5	–	0.3	0.1	–	–	–	–	–	[30]
Sanergy HT	Bal.	22	0.5	1.0	–	0.3	–	0.75	–	–	–	0.001	0.02	0.02	[31]
ZMZ232	Bal.	22	0.5	–	0.26	–	0.21	–	0.04	0.22	–	0.02	–	–	[32]
E-brite	Bal.	26	0.01	–	–	0.025	–	–	–	–	–	0.001	0.02	0.02	[33]

Table 2

Comparison of k_p values for alloy materials in single atmospheres as interconnect steel in SOFCs.

Interconnect	$\text{g}^2\text{cm}^{-4}\text{s}^{-1}$	m^2s^{-1}	T/ °C	Exposure time/h	Atmosphere	Ref.
SUS430	4.03×10^{-14}		750	0–200	Air	[44]
	1.54×10^{-13}		750	200–500	Air	[44]
	$2.09 \times 10^{-13*}$	7.7×10^{-7}	800	168	Air	[45]
	8.50×10^{-14}	3.0×10^{-6}	800	100	Air	[46]
	$2.23 \times 10^{-13*}$	8.2×10^{-7}	800	168	$\text{pH}_2/\text{pH}_2\text{O} = 97/3$	[45]
	3.40×10^{-14}	1.2×10^{-6}	750	100	$\text{pH}_2/\text{pH}_2\text{O} = 94/6$	[46]
	8.80×10^{-14}	3.8×10^{-6}	800	300	$\text{pH}_2/\text{pH}_2\text{O} = 97/3$	[46]
	2.90×10^{-14}	1.1×10^{-6}	850	300	$\text{pH}_2/\text{pH}_2\text{O} = 94/6$	[46]
AISI430	8.88×10^{-12}		800	0–150	Air	[27]
	8.33×10^{-13}		800	150–500	Air	[27]
	1.25×10^{-11}		800	50	Air	[47]
	6.92×10^{-13}		800	100	Air	[48]
	5.26×10^{-12}		800	200	Air	[49]
	2.2×10^{-13}		700	200	Air	[50]
	1.41×10^{-13}		800	960	Air	[51]
Crofer 22 APU	$9.70 \times 10^{-14*}$	3.56×10^{-7}	800	500	Air	[52]
	$1.83 \times 10^{-13*}$	6.65×10^{-7}	850	500	Air	[52]
	2.94×10^{-14}	10^{-7}	800	280	Air	[53]
	1.11×10^{-13}		850	500	Ar + 3% H_2 + 40% H_2O	[54]
	1.89×10^{-13}		850	1000	Air + 1% H_2O	[23]
	6.49×10^{-14}		800	1000	Air	[55]
	1.45×10^{-14}		800	500	Air	[56]
	4.8×10^{-14}		800	100	Air	[57]
	2.8×10^{-14}		800	600	Air	[58]
ZMG232	1.76×10^{-13}		800	1000	Air	[55]
	2.7×10^{-13}		800	1000	Air	[58]
	$1.20 \times 10^{-12*}$	4.42×10^{-6}	800	1032	$\text{CH}_4 + \text{H}_2\text{O}$	[59]
DIN 500049	$2.86 \times 10^{-13*}$	1.05×10^{-6}	800	478	Air	[60]
	$1.40 \times 10^{-13*}$	5.16×10^{-7}	825	478	$\text{pH}_2/\text{pH}_2\text{O} = 94/6$	[60]

interconnect material compared to Crofer 22 APU and SUS430. However, based on its single atmosphere performance, it can be considered a viable option, pending further investigation.

From the comparison of k values, it is evident that dual atmosphere conditions generally result in higher oxidation rates for interconnect materials. Crofer 22 APU demonstrates superior performance under

both single and dual atmospheres, making it a highly suitable material for SOFCs interconnect applications. SUS430, while effective in single atmosphere conditions, may face challenges in dual atmospheres, and ZMG232 shows promise but requires further data for a comprehensive evaluation.

In addition to traditional FSS alloys, high-entropy alloys (HEAs) such as FeCoCrNi and FeCoCrNiMn show potential as SOFCs interconnects due to their low electrical resistivity and conductive oxide formation [66]. SUS445, produced through powder metallurgy, also exhibits high corrosion resistance [67]. The current reported testing used relatively short exposure times compared to SOFC lifespans. However, other studies with thousands of hours of data show significant long-term effects on material stability [68–70]. Over time, oxidation kinetics may shift due to breakaway oxidation leading to increased degradation, impacting interconnect performance [71,72].

3. Oxidation and corrosion effects in different atmospheres

Interconnects in SOFCs are exposed to both anodic (oxidizing) and cathodic (reducing) atmospheres during operation. The interaction of solid and gaseous components under these conditions poses significant challenges to the stability and performance of the metallic components. This interaction can also lead to the deterioration of other components, such as cathode catalysts. The oxidation and corrosion behavior of interconnect alloys has been extensively studied in various environments including cathodic moist air and anodic reducing gas. However, in actual conditions, the interconnect is exposed to both oxidizing and reducing environments simultaneously, resulting in intensified oxidation referred to as the dual atmosphere effect. While research on this phenomenon in the context of mechanisms is scarce, there is a growing interest in exploring its implications for interconnect deterioration and lifespan extension strategies.

3.1. Cathodic (moist air side) effects

In practical SOFCs applications, the "air" side often comprises of more than just pure O_2 . In SOFC design, steam is sometimes used for fuel processing, but using it on the air side is not practical due to the high energy cost of drying the air. In contrast, steam plays a critical role in SOECs for hydrogen production. In recent studies, the atmosphere on the cathode side has typically been a mixture of air or O_2 with water vapor to simulate an atmosphere closer to real working environment of SOFCs (refer to Tables 2 and 3). However this mixture can have detrimental effects. When exposed to high temperatures in air atmosphere, chromium, the active element, preferentially oxidizes forming an oxide scale on the surface through the following reaction:



During the growth of this oxide scale, metal cations move outward from the metal base, and oxygen anions inward from the oxidized scale. Graham et al. [39] employed oxygen isotope tracer methods to discover that the predominant transport mechanism involves the outward diffusion of metal cations (such as chromium). This outward transport of chromium tends to create pores and cavities at the interface between the oxide scale and the metal substrate, potentially reducing the adhesion of the oxide scale [40,86]. In contrast, the inward transport of oxygen contributes less to this process [86]. The oxide scale formed at this stage serves as a protective layer, shielding the interconnects from further corrosion.

There exists a direct correlation between the chromium content, the composition of the oxide, and its antioxidation properties during oxygen exposure [87–90]. Within the same matrix metal, higher chromium content correlates with enhanced oxidation resistance [88]. The chromium content in the matrix must surpass a critical threshold that marks the transition from external to internal oxidation. Adequate chromium

Table 3Comparison of k_p values for alloy materials in dual atmosphere as interconnect steel in SOFCs.

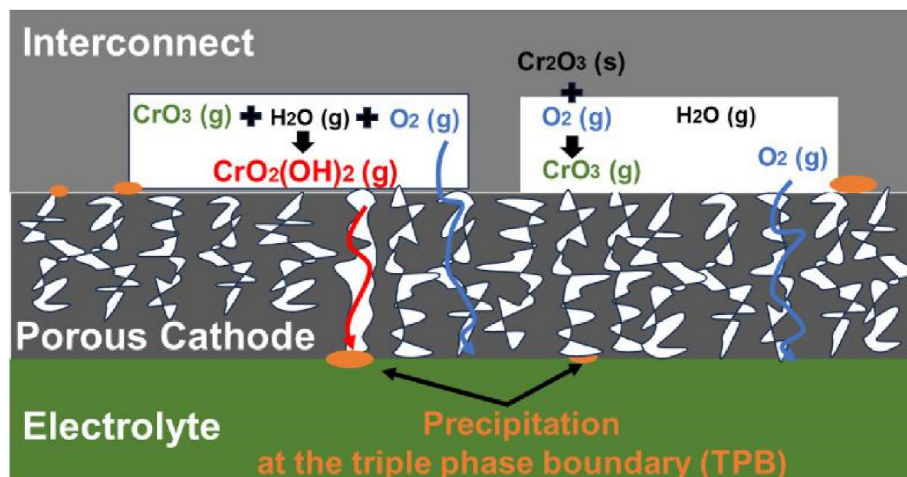
Interconnect	$\text{g}^2\text{cm}^{-4}\text{s}^{-1}$	m^2s^{-1}	T/°C	Exposure time/h	Oxidation Gas	Reduction Gas	Ref.
AISI430	$5.72 \times 10^{-12*}$	2.1×10^{-5}	800	300	Air	$\text{H}_2 + 3\%\text{H}_2\text{O}$	[73]
	$5.72 \times 10^{-12*}$	2.1×10^{-5}	800	300	Air	$\text{H}_2 + 3\%\text{H}_2\text{O}$	[74]
	$1.66 \times 10^{-12*}$	6.1×10^{-6}	800	300	Air + 1 to 3% H_2O	$\text{H}_2 + 3\%\text{H}_2\text{O}$	[63]
AISI441	$2.46 \times 10^{-11*}$	8.9×10^{-5}	800	200	Air + 3% H_2O	$\text{H}_2 + 3\%\text{H}_2\text{O}$	[75]
	$1.93 \times 10^{-11*}$	7.1×10^{-5}	800	200	Air + 3% H_2O	Ar + 3% H_2O	[76]
	$2.02 \times 10^{-12*}$	7.4×10^{-6}	800	336	Air + 3% H_2O	Ar + 5% $\text{H}_2 + 3\%\text{H}_2\text{O}$	[77]
	$6.81 \times 10^{-13*}$	2.5×10^{-6}	700	1000	Air + 3% H_2O	Ar + 3% $\text{H}_2 + 3\%\text{H}_2\text{O}$	[78]
	$1.72 \times 10^{-12*}$	6.3×10^{-6}	800	100	$\text{O}_2 + 5\%\text{H}_2\text{O}$	$\text{H}_2 + 90\%\text{H}_2\text{O}$	[79]
Crofer 22 APU	$8.72 \times 10^{-13*}$	3.2×10^{-6}	800	300	Air	$\text{H}_2 + 3\%\text{H}_2\text{O}$	[74]
	$1.23 \times 10^{-12*}$	4.5×10^{-6}	800	300	Air + 1 to 3% H_2O	$\text{H}_2 + 3\%\text{H}_2\text{O}$	[63]
	$1.23 \times 10^{-12*}$	4.5×10^{-6}	800	300	Air + 3% H_2O	$\text{H}_2 + 1\% - 3\%\text{H}_2\text{O}$	[80]
	$3.82 \times 10^{-12*}$	1.4×10^{-5}	800	200 (Preoxidized)	Air + 3% H_2O	$\text{H}_2 + 3\%\text{H}_2\text{O}$	[81]
	$8.72 \times 10^{-12*}$	3.2×10^{-5}	800	200 (Non-preoxidized)	Air + 3% H_2O	$\text{H}_2 + 3\%\text{H}_2\text{O}$	[81]
Sanergy HT	$2.21 \times 10^{-12*}$	8.1×10^{-6}	850	300	Air + 2.5% H_2O	Ar + 5% $\text{H}_2 + 2.5\%\text{H}_2\text{O}$	[82]
	$3.82 \times 10^{-12*}$	1.4×10^{-5}	850	500	Air + 2.5% H_2O	Ar + 5% $\text{H}_2 + 2.5\%\text{H}_2\text{O}$	[82]
	$8.45 \times 10^{-13*}$	3.1×10^{-6}	800	100	Air	Ar + 5% $\text{H}_2 + 3\%\text{H}_2\text{O}$	[31]
	$2.73 \times 10^{-13*}$	1.0×10^{-6}	800	300	Air	Ar + 5% $\text{H}_2 + 3\%\text{H}_2\text{O}$	[31]
	$3.82 \times 10^{-13*}$	1.4×10^{-6}	800	500	Air	Ar + 5% $\text{H}_2 + 3\%\text{H}_2\text{O}$	[31]
Fe–10Cr	$6.26 \times 10^{-10*}$	2.3×10^{-3}	900	72	Ar + $\text{O}_2 + 7\%\text{H}_2\text{O}$	Ar + $\text{H}_2 + 7\%\text{H}_2\text{O}$	[83]
Fe–20Cr	$8.18 \times 10^{-11*}$	3.0×10^{-4}	900	72	Ar + $\text{O}_2 + 7\%\text{H}_2\text{O}$	Ar + $\text{H}_2 + 7\%\text{H}_2\text{O}$	[83]
Fecralloy	$8.45 \times 10^{-14*}$	3.1×10^{-7}	800	300	Air + 1 to 3% H_2O	$\text{H}_2 + 3\%\text{H}_2\text{O}$	[63]
Haynes230	$2.04 \times 10^{-13*}$	7.5×10^{-7}	800	300	Air	$\text{H}_2 + 3\%\text{H}_2\text{O}$	[84]
	$2.67 \times 10^{-13*}$	9.8×10^{-7}	800	300	Air + 1 to 3% H_2O	$\text{H}_2 + 3\%\text{H}_2\text{O}$	[63]
	$2.21 \times 10^{-13*}$	8.1×10^{-7}	800	300	Air + 3% H_2O	$\text{H}_2 + 3\%\text{H}_2\text{O}$	[85]
HastelloyS	$1.17 \times 10^{-13*}$	4.3×10^{-7}	800	300	Air	$\text{H}_2 + 3\%\text{H}_2\text{O}$	[84]
Haynes242	$2.99 \times 10^{-11*}$	1.1×10^{-4}	800	300	Air	$\text{H}_2 + 3\%\text{H}_2\text{O}$	[84]

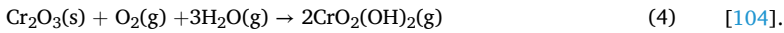
Note: * Data mined by Gongmei et al. assumed parabolic kinetics.

reserves ensure that the alloy maintains self-healing capabilities, preventing the formation of Fe-oxides and reducing the risk of accelerated oxide layer detachment [90]. At chromium levels below 5 wt%, the oxide is predominantly composed of iron oxides and offers little protection [87,89]. With 17–20 wt% chromium, the alloy's surface forms a protective oxide with a lower growth rate, consisting of spinel and chromium oxide [87,89]. The quantity of chromium stored also influences the stability of the interconnect, and depletion of chromium stores can lead to the detachment of oxides, especially on thin Fe–Cr substrates [55,91]. The addition of rare earth elements and yttrium can inhibit the outward diffusion of chromium, thereby increasing the adhesion of the oxide [92,93].

However, especially in high-temperature atmospheres with water vapor, the chromiumoxide scale tends to evaporate through specific

reactions. Gaseous CrO_3 and $\text{CrO}_2(\text{OH})_2$, as described by equations (3) and (4), migrate towards the electrodes or the interface between the electrodes and the electrolyte. There, they undergo a deposition reaction at the triple phase boundary (TPB). These deposits react with the electrolyte material to form phases with higher resistance, which reduces the conductivity of the cell thus leading to a rapid deterioration of SOFC performance, which is also known as "chromium poisoning" [94]. As shown in Fig. 3, the study of the chromium poisoning phenomenon is essential to improve the reliability and lifetime of SOFCs, and the volatilization of chromium can be effectively prevented by using a coating technique, which has been widely applied to the protection technology of interconnects, and is described in detail in section 4.

**Fig. 3.** Schematic representation of chromium evaporation from the interconnect leading to chromium poisoning of SOFCs [24].



3.2. Anodic (fuel side) effects

Solid oxide fuel cells have fuel flexibility and can use a variety of fuels, including hydrogen or hydrocarbon fuels such as natural gas and biogas [95]. However, the use of different types of fuels can pose different requirements and challenges for the operation and maintenance of the cell. For example, the use of hydrocarbon fuels may lead to carbon build-up or other chemical reactions at the anode, and the high water vapor partial pressure generated by water and potential residual sulfides are likely to lead to corrosion of metal interconnections on the anode side [96]. In the context of FSS interconnects, research conducted by Horita et al. [97–99] and Brylewski et al. [100] has indicated that in anode scale's growth rates are faster in humid hydrogen environments compared to the same alloy on the cathodic side, or when oxidized in air i.e. the scales formed on both sides of the interconnect possess similar phases but exhibit distinct microstructures and compositions [98].

When Fe–Cr-based alloys are used in high-water vapor environments, there is a tendency to form Fe or Fe oxides, which does not lead to breakaway oxidation. Breakaway oxidation is influenced by water vapor content and chromium content; higher water vapor content and/or lower chromium content accelerates the onset of segregation oxidation [101]. The impact of hydrogen on the degradation of AISI441 interconnects (Cr% = 17.5–18.5 wt%) was investigated over 336 h at temperatures ranging from 500 to 800 °C by Gunduz et al. [77]. According to this paper, hydrogen appears to impede chromium diffusion, potentially at grain boundaries, thus undermining the integrity of the protective oxide film. This effect was most pronounced at 600 °C and decreases at lower or higher temperatures. At high temperatures, Cr diffusion was enhanced, while at low temperatures, with a reduced amount of Cr, a protective oxide layer was formed.

In contrast, Ni–Fe–Cr-based compounds demonstrate superior oxidation resistance to high temperatures, exhibiting improved oxide adhesion and inhibition of NiO formation in the oxide layer under high H₂O vapor conditions [102]. Similarly, Quadakkers et al. [103] observed that an argon environment (Ar 4%H₂–2%H₂O) enhances the adhesion of chromium oxide scales. Pre-oxidation in a reducing environment positively impacts the formation of adherent oxide scales

3.3. Dual atmosphere effects

In practical applications of Solid Oxide Fuel Cells, interconnects are simultaneously exposed to air and steam on the cathode side, and to hydrogen-rich fuel on the anode side. Consequently, the interconnects experience different hydrogen or hydrocarbon partial pressures leading to chemical potential gradients from the fuel side to the air side. This exposure can lead to unusually rapid corrosion and premature failure of the metal interconnects, a degradation mechanism known as the dual atmosphere effect. Research in the dual atmosphere effect is diverse and includes various topics as shown in Fig. 4. In this section, relevant mechanisms, factors for influencing the dual atmosphere effect, effects of hydrogen on defects, recent debates, and the introduction of pre-oxidation to protect against this effect will be summarized with discussions. An effective preventive method for the dual atmosphere effects using coatings will be illustrated in the next section.

A detailed summary of various experimental exposure conditions for metallic interconnect alloys in dual atmospheres has been listed in Table 3. The gases appearing in the table are usually bottled gas mixtures. The pioneering work by Yang et al. [73] demonstrated that the oxide scale formed on the AISI430 alloy, when exposed to dual atmospheres, was markedly distinct from the scale that developed under exposure to air alone. This anomalous corrosive behavior appeared to be related to hydrogen transport. Further, Yang et al. conducted studies under air/hydrogen dual-exposure conditions on Fe–Cr-based [74,80,84,105] and Ni–Fe-based alloys [63], finding that the dual-atmosphere effect was particularly significant on the composition and microstructure of the oxide scales grown on the air side, with a lesser impact on the oxidation/corrosion on the hydrogen fuel side. As shown in Fig. 5, the oxide scale on the air side forms due to the outward diffusion of iron and chromium and the inward diffusion of oxygen, initially forming a protective chromium oxide layer. Over time, as chromium depletes, iron oxides begin to form. If this protective layer deteriorates, oxidation may accelerate.

The dual atmosphere effect is also temperature dependent. Alnegren et al. [78], in their study of AISI441, found that oxidation at 600 °C was more severe than at 800 °C; an unexpected phenomenon that contradicts conventional understanding, wherein oxidation increases with temperature. This has been observed elsewhere and linked in other oxidation scenarios to silicon assisting a healing layer in [71]. At 600 °C,

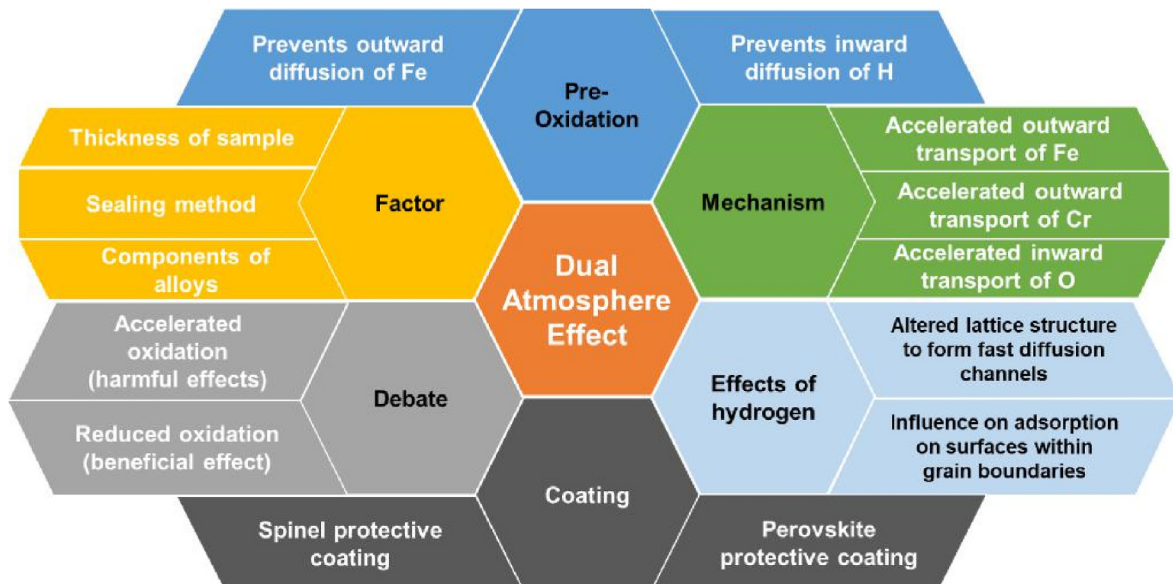


Fig. 4. Summary of research topics in dual atmosphere effect on interconnect in SOFCs.

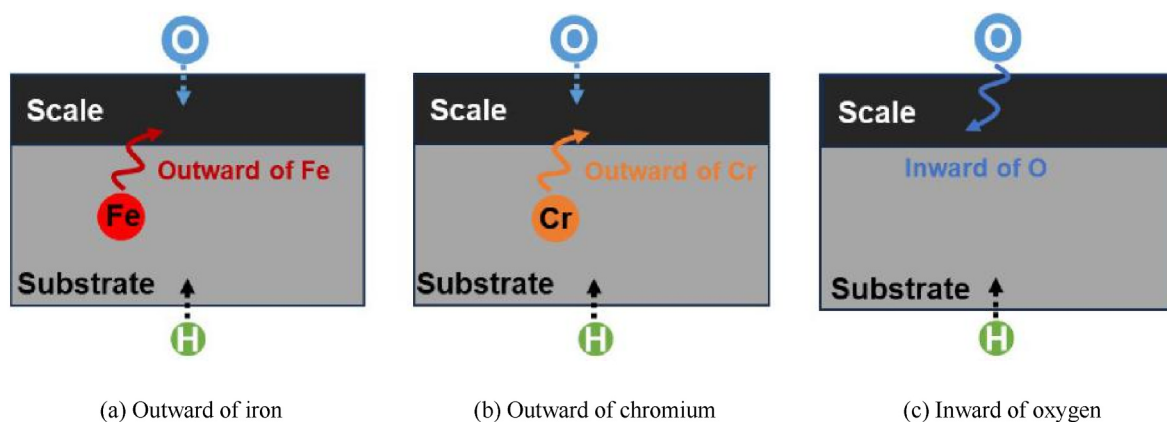


Fig. 5. Schematic diagram summarizing the transport paths of different ions during oxide scale formation.

catastrophic breakaway oxidation can happen, while at 800 °C, the oxidation process is more stable. They suggested that hydrogen diffusion, through the oxide and steel from the hydrogen side, effects the oxidation on the air side, making it harder for protective chromium oxide to form, especially at 600 °C. At 800 °C, although hydrogen still harms the chromium oxide layer, this is balanced by the faster diffusion of chromium at higher temperatures. This creates new challenges for anti-corrosion technology for interconnect components, as SOFCs are now being designed to operate at lower temperatures due to advancements in cathode catalysts [106].

Over the past two decades, researchers [31,60,73–75,78–80,84,105,107,108] have studied the dual atmosphere effect, but its severity varies greatly [60,75,78,108]. The mechanism by which hydrogen damages protective oxide films remains unclear. Kurokawa et al. [108] and Stygar et al. [60] did not observe significant dual atmosphere effects in the 800 °C exposure of FSS interconnects. In their study, thicker steel samples (2 mm) were used and, due to the use of high-temperature silicone sealant, the samples remained unsealed during the first 1.4 h of testing. This is consistent with the findings of Goebel et al. [107] that thicker AISI441 withstands the harmful effects of the dual atmosphere for a longer period of time. The use of thicker steel may have prolonged the hydrogen diffusion process to some extent, thus mitigating the effects of the double atmosphere. Skilbred et al. [31,82] observed only a slight dual atmosphere effect in their study of Sandvik Sanergy H alloy. They believed that at the initial stage of oxidation, hydrogen diffuses into the stainless steel and saturates it, and that this hydrogen dissolves in the form of protons in the oxide formed in the air, affecting its defect structure. Goebel [107,109], Alnegren [78,79], and Gannon [76] reported the most severe dual atmospheric effects. Amendola et al.'s study [81] showed that Crofer 22 APU in a single atmosphere forms an oxide layer of Cr_2O_3 , while an increase in Fe was observed in the surface oxide layer at the interface with the metal substrate in a dual atmosphere.

There is no consensus on the mechanism by which hydrogen affects the oxidation of interconnects. Ehlers et al. [110] believed that the growth of the oxide scale is controlled by the competitive adsorption of oxygen and water vapor molecules on the inner and outer surfaces of the oxide scale, as well as by the formation of volatile iron hydroxide during transient oxidation. Essuman et al.'s study [83] showed that using water vapor as the oxygen source, as opposed to O_2 gas, promotes the internal oxidation of Cr in Fe–Cr alloys. This could be due to hydrogen protons expanding into the iron lattice, forming a rapid diffusion pathway, thereby increasing the inward diffusion rate of oxygen [111]. The movement paths of iron, chromium and oxygen during the formation of the oxide scale are shown in Fig. 5. However, work by Chyrkin et al. [112,113], have shown that the inward diffusion of oxygen is not driven by hydrogen protons expanding into the iron lattice as previously hypothesized.

The dual atmosphere effect is difficult to explain solely by the

acceleration of the oxidation rate. Gunduz et al. [77] used experimental data from Skilbred et al. [31] and Alnegren et al. [78] to modify the Wagner formula, speculating that the increase in scale thickness in a dual atmosphere may be related to the rapid diffusion of Cr in the BCC lattice. Ani et al. [114] conducted interdiffusion experiments on Fe/Fe–16Cr diffusion couples in dry and wet environments, indicating that the diffusion coefficient of Cr is not affected by dissolved hydrogen, and that dissolved hydrogen promotes the transport of oxygen along the metal/oxide interface. However, this result only indicates that the lattice diffusion of Cr is not affected by hydrogen at 800 °C. Gunduz et al. [77] speculated that the dual atmosphere effect might be related to hydrogen's influence on chromium diffusion at grain boundaries, which could be due to the formation of hydrides or the adsorption of hydrogen on the inner surface of grain boundaries, impeding chromium diffusion. However, there is no direct experimental evidence to support this speculation. Isotope exposure and chromium tracer studies may be helpful in understanding the role of hydrogen in the dual atmosphere effect. Additionally, Density Functional Theory (DFT) calculations could reveal the possibility of hydride formation at grain boundaries.

Pre-oxidation may help mitigate the adverse effects of dual atmosphere response in FSS. Kurokawa et al. [108] studied the hydrogen permeation of pre-oxidized AISI430 steel, finding that Cr_2O_3 was an effective barrier against hydrogen permeation. Amendola et al. [81] showed that pre-oxidation significantly inhibited the outward transport of iron from FSS to the surface and reduced the thickness of the surface oxide layer on Crofer 22 APU. Goebel et al.'s work [109] further confirms that hydrogen can be prevented from penetrating AISI441 on the pre-oxidized skin. The dual atmosphere effect is not always negative; for example, air/hydrogen dual exposure has no adverse effect on the stability of the oxide skin of Ni–Cr-based alloys [84,85] and aluminum oxide-formed alloys [63]. Yang et al. [84] showed that dual atmosphere exposure did not significantly alter the oxidation behavior of Ni–Cr-based alloy Haynes 242, which still formed thick and defective double-layer oxide scales on its surface. The oxide skin on the air side caused by dual exposure is slightly thicker than that of the single atmosphere exposed sample, but the oxide skin/metal interface is smoother than that of the single atmosphere exposed sample [63]. The dual atmosphere effect is also influenced by the use of barrier coatings, as proposed in recent studies by Tomas et al. [115,116], Singh et al. [117] and Reiser et al. [118]. These coatings help protect the interconnect by preventing hydrogen from affecting the formation of the protective chromium oxide layer, thus improving the material's performance in dual atmosphere conditions [119].

The most interesting result from recent studies on operation SOFC stacks is that, even after long operational periods, dual atmosphere oxidation (such as accelerated or breakaway oxidation) has not been observed after 40,000 h [68] or 18,000 h [69]. This discrepancy between laboratory tests and real stack operations may be explained by

several factors. First, the operational environment in real stacks is more complex and dynamic, with fluctuating gas compositions and temperatures, which may reduce the formation of critical oxide layers that cause accelerated oxidation. Second, protective coatings, such as manganese cobalt spinel, applied in real stacks, effectively reduces chromium evaporation and hydrogen permeation, both of which are key drivers of dual atmosphere oxidation in lab settings [120,121]. Finally, materials in real stacks undergo gradual adaptation during long-term operation, leading to more stable oxidation behavior, whereas lab tests often simulate accelerated degradation over shorter periods. This will be an interesting area for future research.

4. Use of coatings for oxidation and corrosion protection

Although newly developed alloys, such as Crofer 22 APU, have shown significant performance improvements over traditional alloys (e. g. Fe–Ni–Cr-based alloys) and ceramic interconnects, there are still several challenges to the oxidation-resistance capabilities of the alloys working under dual atmospheres.

First of all, research suggests that the scale thickness caused by the oxidation reaction will increase with time, potentially following parabolic oxidation kinetics [137], leading to a growth in its resistance that can cut down the output power of the SOFCs. In addition, poisoning of the cathode material due to chromium evaporation can result in the degradation of the oxide exchange rate which may also lower the power of the SOFCs. Last but not least, there may be a mismatch in the coefficient of thermal expansion between the interconnect and other stack components (e.g., electrolyte and electrodes) causing material damage during temperature changes affecting the remaining useful life of the cell [138].

To address the mentioned issues, coatings are proven to be an effective protective method to alleviate the harmful impact brought by oxidation processes and prolong the interconnect's lifespan [7,18,120,122,124,139]. In short, a stable coating can act as a barrier to reduce the migration of these harmful components [7]. Also, appropriate coatings can provide a buffer layer to reduce the stresses caused by thermal expansion mismatches. Coating materials can be selected to improve the conductivity of the oxide scale on the interconnect surface; this is especially important as the oxide layer kinetics are greater at higher temperatures [139]. Coatings also improve the stability of interconnects in corrosive environments containing sulphur and water vapor. Therefore, a coating can ensure the long-term stable operation and efficient energy conversion of SOFC systems.

Fruitful results have been obtained on the constituent elements, components, and deposition techniques used in protective coating, as summarized in Table 4. Metal oxides are considered the most commonly studied type of interconnection coating, especially perovskite-structured metal oxides and spinel-structured metal oxides.

Metal oxides are widely used due to their excellent thermal and chemical stability. ABO₃ perovskite-type oxides, which demonstrate P-type electronic conduction (where holes act as the charge carriers), maintain stability under low partial pressures. However, they exhibit a weak shielding effect against chromium. Feng et al. [122] deposited LaCrO₃-based coatings on type 430 stainless steel by a high-energy micro-arc alloying (HEMAA) process to improve its properties and oxidation resistance, and the coatings demonstrated good protection with a three-layer structure and low contact resistance. Persson et al. [124] deposited six different structures of oxides on Crofer 22 APU, including fluorite, spinel, and perovskite oxides. The results showed that (La_{0.85}Sr_{0.15})CoO₃ + Co₃O₄ exhibited the best corrosion protection in humid air at 900 °C and had multiple interaction mechanisms with the grown oxide film. Feng et al. [123] prepared a deposited DyCrO₃ coating using high-energy micro-arc alloying technology, which can reduce the ASR of steel, especially reducing separation oxidation in wet air.

AB₂O₄ spinel coatings can serve as an effective mass transfer barrier for the outward migration of Cr cations. Reddy et al. [18,120] have

Table 4

Comparison of coatings and preparation methods applied to interconnects for use in SOFCs.

Coating in cathode side	Structure	Methods	Thickness	Ref.
LaCrO ₃ , (Ni,Fe)Cr ₂ O ₄	Perovskite, spinel	HEMAA	25 pm	[122]
DyCrO ₃ , NiFe ₂ O ₄	Perovskite, spinel	HEMAA	15 pm	[123]
La _{0.85} Sr _{0.15} MnO ₃ , Mn ₂ O ₃	Perovskite	Spray	7/15/30 pm	[124]
La _{0.85} Sr _{0.15} CoO ₃ , Co ₃ O ₄	Perovskite	Spray	7/15/30 pm	[124]
La _{0.75} Sr _{0.25} Cr _{0.5} Mn _{0.5} O ₃	Perovskite	Sol-gel	2–3 pm	[125]
CuFe ₂ O ₄	Spinel	Magnetron Sputtering	5 pm	[126]
CuFe ₂ O ₄	Spinel	Glycine Nitrate Process (GNP)	1.5 pm	[127]
CuCo ₂ O ₄	Spinel	Sol-gel	10 pm	[128]
MnCo ₂ O ₄	Spinel	–	–	[129]
MnCo ₂ O ₄	Spinel	SP	–	[130]
MnCo ₂ O ₄	Spinel	Spray	7/15/30 pm	[124]
MnCo ₂ O ₄	Spinel	Inkjet Printing	50 pm	[20]
MnCo _{1.8} Fe _{0.2} O ₄	Spinel	Inkjet Printing	50 pm	[20]
Mn _{1.0} Co _{1.9} Fe _{0.1} O ₄	Spinel	APS	–	[131]
NiCo ₂ FeO ₄	Spinel	Sol-gel	600 nm	[132]
Mn _{1.5} Co _{1.5} O ₄	Spinel	Sol-gel	5 pm	[133]
Mn _{1.5} Co _{1.5} O ₄	Spinel	Sol-gel	5 pm	[134]
Mn _{1.5} Co _{1.5} O ₄	Spinel	Thermal evaporation	2 pm	[121]
Mn _{1.5} Co _{1.5} O ₄	Spinel	Electrophoretic	20 pm	[135]
Mn/Co	Spinel	Electroplating	20 pm	[13]
Mn/Co	Spinel	EPD	20 pm	[136]
Mn/Co, Mn/Cu	Spinel	Thermal Spray	1 nm	[17]
Ce/Co	Spinel	PVD	Ce 10 nm, Co 640 nm	[16]
Ce/Co	Spinel	PVD	Ce 10 nm, Co 600 nm	[120]
Ce/Co, Ce/Cu, Ce/MnCu	Spinel	PVD	1 nm	[17]
Co ₃ O ₄	Spinel	Electroplating	9 pm	[15]
Co ₃ O ₄	Spinel	Spray	7/15/30 pm	[124]
ZrO ₂	Fluorite	Spray	7/15/30 pm	[124]
Al ₂ O ₃	Corundum	Spray	7/15/30 pm	[124]

reported that (Mn,Co)₃O₄ spinel coatings can reduce chromium evaporation by 60 fold, making these a potential interconnect coating. Hua et al. [129] have shown that the conductivity of MnCo₂O₄ spinel is improved by two orders of magnitude compared to MnCr₂O₄ spinel, and pre-oxidation (800 °C air for 2 h) after calcination in a reducing atmosphere is the optimal process for forming this protective coating. Mn_{1.5}Co_{1.5}O₄ spinel contains a binary structure of both cubic and tetragonal phases, exhibiting good thermal stability and electronic conductivity, making it a potential application for SOF interconnect [133].

In addition to Mn–Co based spinel coatings, Cu–Fe coated spinel layers are also used for interconnects [7]. Hosseini et al. [127] demonstrated that CuFe₂O₄-coated Crofer 22 APU prepared by screen printing, reduced Cr evaporation by approximately 92% and significantly reduced ASR. The ASR after 600 h of oxidation at 800 °C was 14.3 mΩ cm². Geng et al. [126] reported that the CuFe₂O₄ spinel layer can improve the electrical properties of SUS430 by inhibiting Cr outward migration and reducing oxidation rate. Hassan et al. [7] compared Mn–Co and Cu–Fe based coatings in different literature and reported that CuFe₂O₄ is the best reported spinel coating to date. Moreover, in low-cost ferrite interconnects, SUS430 exhibits equivalent performance to Crofer 22 APU based interconnects after appropriate spinel coating.

It is worth mentioning that the deposition of spinel materials can be

achieved using a variety of techniques, including Physical Vapor Deposition (PVD) [16,17], Sol-Gel Dip Coating [128,132], Electrophoretic Deposition (EPD) [19,136], Electroplating [13,15], Screen Printing [125,133,134], Spray Pyrolysis (SP) [130], Inkjet Printing [20], Thermal Evaporation [121], and Atmospheric Plasma Spraying (APS) [131]. Reddy et al. [140] applied eight distinct techniques to deposit various coatings on Crofer 22 APU, subjecting it to a 3000 h exposure at 800 °C. The Co–Mn-based coatings on the steel resulted in a chromium evaporation rate that was over 50 times lower and a Cr₂O₃ oxide layer that was at least three times thinner than uncoated steel. The coated steel samples demonstrated lower ASR values (15–25 mΩ cm²) compared to their uncoated counterparts (70 mΩ cm²), irrespective of the coating's thickness, composition, or method of deposition.

5. Conclusion

This review has systematically examined the recent advances in the understanding of corrosion and oxidation behaviors of SOFCs' interconnects, with a specific focus on the impacts of dual atmosphere conditions on FSS. It is evident from the study that, while there have been significant strides in characterizing and mitigating degradation mechanisms, substantial research gaps remain, particularly in the long-term effects of dual atmospheres on an interconnect's durability. Addressing these gaps is crucial for the further development of SOFCs technologies, as robust interconnects are key to enhancing the overall efficiency and lifespan of SOFCs systems. The implications of this research extend beyond academic interest and hold substantial potential for advancing SOFC deployment in the energy sector. As the global energy landscape shifts towards more sustainable solutions, the development of reliable and efficient SOFCs represents a critical pathway toward achieving lower carbon emissions and fostering a greener energy future. Future work should therefore not only continue to explore new materials and coatings but also consider the integration of these advances into full SOFC systems to assess their practical viability and performance in real-world applications.

CRedit authorship contribution statement

Gongmei Yang: Writing – original draft, Methodology, Investigation, Formal analysis, Conceptualization. **Andrew Potter:** Writing – review & editing. **Joy Sumner:** Writing – review & editing, Supervision.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Gongmei Yang reports financial support was provided by Chinese Scholarship Council. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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