



Coated stainless steel 441 as interconnect material for solid oxide fuel cells: Oxidation performance and chromium evaporation[☆]



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HIGHLIGHTS

- We exposed coated AISI 441 in a SOFC cathode side atmosphere.
- The oxidation and chromium volatilization were monitored.
- RE coated 441 showed improved corrosion resistance and no oxide scale spallation.
- RE/Co-coated 441 showed superior corrosion behaviour and low chromium evaporation.

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ABSTRACT

Reactive Element (RE) and RE/cobalt-coated stainless steel AISI 441 was exposed at Solid Oxide Fuel Cell (SOFC) cathode conditions (850 °C in air with 3% water content) for up to 500 h. The chromium evaporation was measured by applying the denuder technique. Uncoated material exhibited severe spallation which could be successfully prevented by using cerium or lanthanum coatings. By applying double layer coatings of cerium or lanthanum in combination with cobalt the oxidation rate was decreased and the chromium volatilisation was also about 90% lower than the uncoated material.

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1. Introduction

Solid Oxide Fuel Cell (SOFC) technology is considered to be promising in the fields of decentralised electricity and heat production. To achieve high voltage output, planar fuel cell elements so-called PEN elements (positive electrode, electrolyte, negative electrode) consisting of an anode, an electrolyte and a cathode which are stacked together [1]. So called interconnectors are needed to stack these PEN elements. The functions of interconnects

are not only the electrical connection between two PEN elements but also the separation of the anode atmosphere of one cell from the cathode atmosphere of the following cell. The requirements for interconnects, among others, are an expansion coefficient similar to the ceramic components of the PEN element, high temperature stability, high electrical conductivity, good gas tightness and low production costs.

Recent developments, such as a thinner electrolyte thickness and improved cathodes, have resulted in lower operating temperatures of about 600–800 °C [2,3]. An advantage of these lower operation temperatures is the larger choices of potential interconnect materials. At high temperatures mainly ceramic materials have been applied, whereas at lower temperatures ferritic steels can also be used for an interconnect application. The advantages of these steels are a similar expansion coefficient, good durability, relatively high electronic conductivity, mechanical durability and they are also more economical. Most promising for an interconnect

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application are ferritic steels with a chromium content between 18 and 25%.

These steels form a protective chromia layer when exposed to an SOFC cathode atmosphere. However, the protective chromia scale of the steel leads to degradation of the fuel cell due to the volatilization of $\text{CrO}_2(\text{OH})_2$, which reacts at the cathode/electrolyte/atmosphere triple points, leading to so called cathode poisoning [4,5]. Early studies on conventional commercial ferritic stainless steels have shown that they lack sufficient corrosion resistance for most interconnect applications. This leads to the development of tailor-made steels not only to increase creep strength and corrosion resistance but also to decrease chromium evaporation. Some examples of these steels are Crofer 22 APU and Crofer 22 H from ThyssenKrupp VDM, Sandvik Sanergy HT from Sandvik Materials Technology AB or ZMG 232 G10 from Hitachi Metals [6,7]. All of the steels have additions of about 0.5 wt.% manganese, leading to the formation of an outer chromium manganese spinel layer during exposure. This phase reduces chromium evaporation by about 60–70% [8].

In order to improve corrosion resistance, the above-mentioned steels contain so called Reactive Elements (RE), e.g. cerium, lanthanum, yttrium, neodymium or zirconium. The beneficial effect of REs has been discussed for several decades and different mechanisms have been proposed, which is beyond the focus of this paper [9–11]. Besides small additions of REs to the steel it has been shown that RE coatings can also have a beneficial effect on the corrosion performance of the steel in SOFC relevant atmospheres [12–14].

Chromium evaporation can be decreased by thin film coatings, which can be classified into two main groups; first perovskite structure and second spinel structure coatings.

Up to now various perovskite coatings for corrosion protection and the prevention of chromium evaporation have been applied; Stanislawski et al. have applied various lanthanum-based ceramics such as LSC (LaScCrO_3) or LSM (LaSrMnO_3) in thicknesses between 32 and 45 μm [8]. The coated samples exhibited lower chromium evaporation, but some of the coatings suffered spallation. Kurokawa et al. have also studied LSM (LaSrMnO_3) and LSCF (LaSrCoFeO_3) and found a significant decrease in chromium evaporation [15]. A common drawback of perovskite coatings are often difficulties in the adhesion of the coatings [16].

Spinel coatings are the second class of coatings commonly proposed. The ones most referred to in the literature are coatings based on cobalt and its oxides combined with other transition metals. Different thin film technologies have also been employed to apply spinel coatings. Stanislawski et al. have tested cobalt, nickel and copper coatings, which were applied via sputter coating on Crofer 22 APU, and found that all coatings decreased chromium evaporation significantly. Yang et al. have investigated $\text{Mn}_{1.5}\text{Co}_{1.5}\text{O}_4$ spinel coatings, prepared with a slurry-coating process, on AISI 441, and found that these coatings increased corrosion resistance and decreased electrical resistance [17].

Besides these coatings for the prevention of chromium evaporation, combinations of RE coatings with spinel structure coatings have been developed and investigated. Qu et al. have combined yttrium with cobalt coatings prepared via a sol–gel route and have investigated their corrosion and electronic properties. They found that the combined coating had lower resistances than single-coated samples [18]. Froitzheim et al. have shown a beneficial effect by combining cerium coatings with cobalt coating, where both the oxidation and the chromium evaporation could be lowered significantly [19].

The above cited studies exemplify that uncoated ferritic stainless steels are not suitable for almost all SOFC applications. Therefore, if coating steel is inevitable, the combination of a low cost steel

substrate with a coating that provides the required surface properties seems worthwhile investigating; particularly since the interconnect can, according to Hall et al. [20] comprise up to 45% of the total stack costs.

Type 441 stainless steel is, from an economic point of view, very promising but its corrosion performance in SOFC environments is not sufficient for a long-term operation [6]. This is mainly attributed to the higher silicon content and lower silicon to niobium ratio [6] and the lack of reactive elements. The aim of this investigation, therefore, is to determine if the corrosion resistance of type 441 stainless steel can be enhanced by applying so-called reactive element coatings, and if combinations of reactive element coatings and cobalt coatings can enhance the chromium evaporation performance of this stainless steel and additionally show an improvement in corrosion performance.

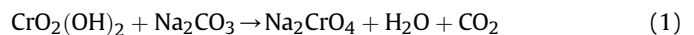
2. Experimental

2.1. Sample preparation

Pre-coated and uncoated steel strips of AISI 441 (compositions given in Table 1) received in sheets of 0.2 mm thickness from Sandvik Materials Technology AB were cut to coupons of 15×15 mm. The coatings were applied by physical vapour deposition and coating thickness was measured during the deposition process via a quartz crystal balance. Metallic targets were used; although in the case of the Ce and La coatings it might be expected that these form oxides during the deposition process. The samples were ultrasonically cleaned first in acetone and afterwards in ethanol for 20 min for each step. After cleaning, the samples were dried and weighted on a Sartorius MC5 scale.

2.2. Exposure

The samples were exposed using the denuder technique; described in more detail elsewhere [21]. The experiments were performed in a tubular furnace equipped with a flow restrictor of silicon carbide to ensure turbulent flow. A so-called denuder tube was placed at the outlet of the furnace, coated with sodium carbonate to collect evaporated chromic species. The reaction between the evaporated chromium-oxy-hydroxy species and sodium carbonate is shown in equation (1).



The denuder tubes were changed regularly and washed out with MQ water, the chromium concentration of the solution was measured using photo spectroscopy. By knowing the samples' geometry and by calibrating the photo spectrometer for chromium concentration it is possible to relate the amount of evaporated chromium to the exposed sample area.

Table 2 lists the different tested samples. To quantify chromium evaporation, the samples were exposed isothermally to a temperature of 850 °C for 500 h in air, which contained 3% water vapour. The tested environment was chosen to simulate a relatively harsh, due to high humidity and temperature cathode side environment. Since the focus of this study was concerning cathode side corrosion and the effect of chromium evaporation the samples were only exposed to a cathode side environment, which is in contrast to real

Table 1

Batch-specific values provided by the manufacturer, given in wt. %.

| | Fe | Cr | C | Mn | Si | S | P | Ni | Nb | Ti |
|-------|------|-------|-------|------|------|-------|-------|------|------|------|
| Wt. % | Bal. | 17.83 | 0.012 | 0.26 | 0.55 | 0.002 | 0.024 | 0.13 | 0.48 | 0.14 |

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