



Does the conductivity of interconnect coatings matter for solid oxide fuel cell applications?



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HIGHLIGHTS

- Different Co coating thicknesses are exposed to air for 500 h at 600 °C.
- The coating thickness does not significantly affect the ASR.
- The contribution of the Co₃O₄ coating to the ASR is negligible.
- Theoretical calculations verify that the main contributor to the ASR is Cr₂O₃.
- The results can be generalized for most coating materials and temperatures.

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ABSTRACT

The present work aims to quantify the influence of typical interconnect coatings used for solid oxide fuel cells (SOFC) on area specific resistance (ASR). To quantify the effect of the coating, the dependency of coating thickness on the ASR is examined on Crofer 22 APU at 600 °C. Three different Co coating thicknesses are investigated, 600 nm, 1500 nm, and 3000 nm. Except for the reference samples, the material is pre-oxidized prior to coating to mitigate the outward diffusion of iron and consequent formation of poorly conducting (Co,Fe)₃O₄ spinel. Exposures are carried out at 600 °C in stagnant laboratory air for 500 h and subsequent ASR measurements are performed. Additionally the microstructure is investigated with scanning electron microscopy (SEM). On all pre-oxidized samples, a homogenous dense Co₃O₄ top layer is observed beneath which a thin layer of Cr₂O₃ is present. As the ASR values range between 7 and 12 mΩcm² for all pre-oxidized samples, even though different Co₃O₄ thicknesses are observed, the results strongly suggest that for most applicable cases the impact of the coating on ASR is negligible and the main contributor is Cr₂O₃.

1. Introduction

Interconnects are an essential part of solid oxide fuel cells (SOFC), where they connect multiple cells to form a fuel cell stack, thus increasing the overall potential of a fuel cell [1]. Therefore, one important factor of any interconnect material is its electrical resistance at operating temperatures. In theory, the resistance should be negligible when using ferritic stainless steel interconnects, which is the norm today [1,2]. However, due to corrosion throughout the operation of a fuel cell, a continuously growing oxide layer occurs on the interconnect, resulting in an increase in resistivity. Issues such as Cr-evaporation and other corrosion-related problems can easily be decreased by applying coatings to the steel. In recent years, plenty of studies have been published on highly conductive coatings, which are thought to lower the area specific resistance (ASR) of the interconnect significantly even

after long-term operations [3–14]. The most prominent suggestions are compounds with spinel structure as these are easily deposited onto the steel. Besides applying the spinel coating directly to the steel by, for example, screen printing or physical vapor deposition (PVD), a different, more cost-efficient approach is to apply a metallic coating instead. The metal or metals will then oxidize to form M₃O₄ [15–17] during fuel cell operation. An extensive list of different spinel conductivities at 800 °C can be found in Ref. [18]. Especially Cu_{1.3}Mn_{1.7}O₄ and MnCo₂O₄ stand out with high conductivities, $\sigma < 225 \text{ S cm}^{-1}$ at 750 °C and $\sigma < 60 \text{ S cm}^{-1}$ at 800 °C, respectively. To increase conductivity even further, dopants such as Cu or Ni have been suggested as additives to the (Co,Mn)₃O₄ spinel [7,9]. However, the contribution of this coating to the overall ASR remains unknown. This is especially important as a continuously growing Cr₂O₃ layer is always present below the coating. With regard to the low conductivity of Cr₂O₃, which

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Table 1
Composition of Crofer 22 APU in weight%.

Crofer 22 APU	Fe	Cr	Mn	La	Ti	Al	Cu	Si	P	C	S
W.-Nr. 1.4760	Bal.	22.92	0.38	0.09	0.06	0.01	0.01	0.01	0.005	0.004	< 0.002

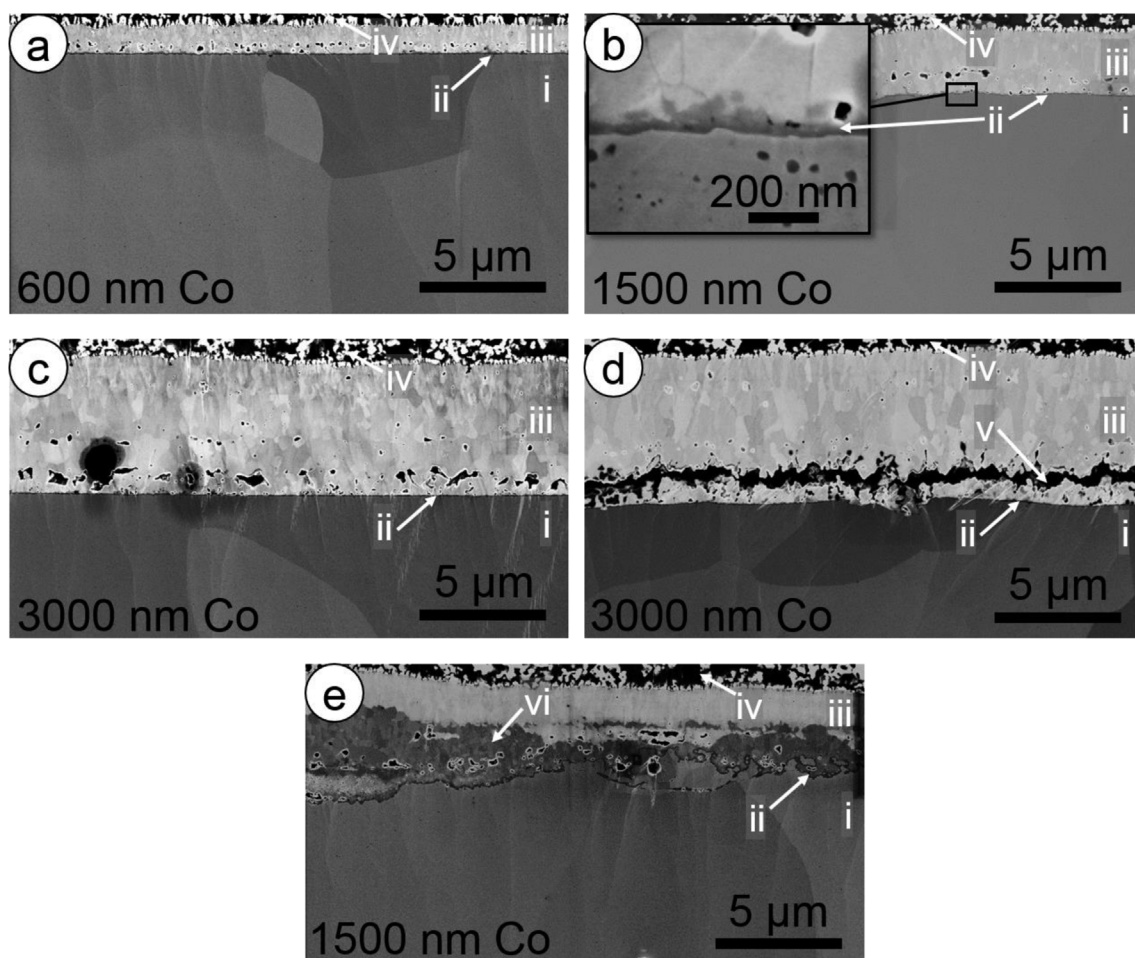


Fig. 1. SEM images of BIB milled cross-sections of Co coated Crofer 22 APU exposed for 500 h at 600 °C in stagnant, laboratory air. All samples were Co coated according to the following specifications a) a pre-oxidized sample with 600 nm Co coating, b) a pre-oxidized sample with 1500 nm Co coating, c) a pre-oxidized sample with 3000 nm Co coating, d) a pre-oxidized sample with 3000 nm Co coating, which showed signs of spallation after exposure, and e) a non-pre-oxidized sample with 1500 nm Co coating. All phases were identified as follows: i) steel, ii) thin Cr_2O_3 layer, iii) Co_3O_4 , iv) Pt electrode, v) spallation crack, and vi) $(\text{Co,Cr,Fe})_3\text{O}_4$.

ranges between 0.001 and 0.05 S cm^{-1} [19–26], the main contributor to the overall ASR is expected to be Cr_2O_3 in most practical cases.

To investigate the contribution of all oxides to the overall ASR, the moderately conductive Co_3O_4 ($\sigma = 6.7 \text{ S cm}^{-1}$ at 800 °C [18]) coating was applied to Crofer 22 APU with different thicknesses and, subsequently, exposed at 600 °C. For well adherent and dense coatings, it can be assumed that, if the coating significantly affects the ASR, thicker coatings will result in higher ASR values. Therefore a metallic Co conversion coating was prepared on Crofer 22 APU with three different Co coating thicknesses, 600 nm, 1500 nm, and 3000 nm, and the ASR was measured after 500 h of exposure in stagnant laboratory air at 600 °C. This temperature is typical for the lower end of SOFC operating conditions and is expected to result in a very thin Cr_2O_3 layer, and, thus, any effect of the Co coating thickness on the ASR can be expected to be the most pronounced.

2. Materials and methods

For this study, $15 \times 15 \times 0.3 \text{ mm}^3$ large coupons of Crofer 22 APU

were used. The composition of this material is shown in Table 1. The as-received and cut-out samples were ultrasonically cleaned in acetone and ethanol. Except for the reference case, all samples were pre-oxidized for 3 min at 900 °C in air prior to coating. Coatings were applied by Sandvik Materials Technology using PVD. Chosen Co coating thicknesses were 600 nm, 1500 nm, and 3000 nm. After coating, all samples were again ultrasonically cleaned in acetone and ethanol, and subsequently, exposed for 500 h at 600 °C in stagnant laboratory air.

After exposure, ASR measurements were performed by sputtering a defined area of 1 cm^2 with a thin Pt layer, followed by painting with the Pt paste Metalor 6926. Sintering of the Pt paint was achieved in a two-step process, a drying step for 10 min at 150 °C and a sintering step for 1 h at 600 °C. The ASR values were then measured according to the 4-point probe method in DC mode using the NorECs Probestat (Norway) and a Keithley 2400 source meter. The applied current was set to 100 mA cm^{-2} , and the ASR values were measured directly at exposure temperature, as well as during the cooling down phase, to verify semi-conductive behavior. More details on the ASR measurements can be found in Refs. [27] and [28]. Cross-sections were prepared using a Leica

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