



Model-based prediction of the ohmic resistance of metallic interconnects from oxide scale growth based on scanning electron microscopy



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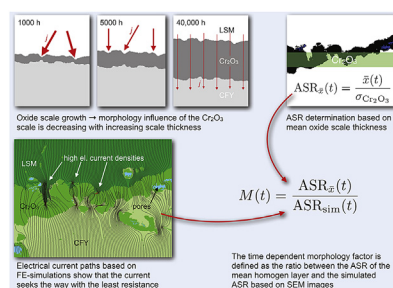
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HIGHLIGHTS

- FE-modeling using oxide scale morphology to predict ohmic degradation of SOFC stacks.
- Oxide scale morphology has major impact on ohmic resistance of metallic interconnects.
- Using SEM images to simulate el. current distributions in metallic interconnects.
- New method to assess the electrical conductivity of thermal grown oxide scale.
- Model-based method for reliable extrapolation of degradation data of SOFC stacks.

GRAPHICAL ABSTRACT



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ABSTRACT

The increase of ohmic losses caused by continuously growing Cr₂O₃ scales on metallic interconnects (MICs) is a major contribution to the degradation of SOFC stacks. Comparison of measured ohmic resistances of chromium- (CFY) and ferritic-based alloy (Crofer) MICs at 850 °C in air with the growth of mean oxide scale thicknesses, obtained from SEM cross section images, reveals a non-trivial, non-linear relationship. To understand the correlation between scale evolution and resulting ohmic losses, 2D finite element (FE) simulations of electrical current distributions have been performed for a large number of real oxide scale morphologies. It turns out that typical morphologies favor nonhomogeneous electrical current distributions, where the main current flows over rather few “bridges”, i.e. local spots with relatively thin oxide scales. These current-“bridges” are the main reason for the non-linear dependence of ohmic losses on the corresponding oxide scale morphology. Combining electrical conductivity and SEM measurements with FE simulations revealed two further advantages: it permits a more reliable extrapolation of MIC-degradation data over the whole stack lifetime and it provides a method to assess the effective electrical conductivity of thermally grown Cr₂O₃ scales under stack operation.

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1. Introduction

Commercially available stationary combined heat and power (CHP) systems based on solid oxide fuel cells (SOFCs) [1–4] are currently expected to reach lifetimes of > 40,000 h with a maximum power degradation rate of < 1% per 1000 h [5]. It is obvious that extensive performance testing over such a long time period is not feasible. Consequently, to obtain reliable lifetime and degradation predictions, advanced experimental methods to perform short- and medium term tests combined with simulation tools providing accurate models are needed [6–10]. In this study we present methods for model based predictions of the losses arising from oxide scale growth on metallic interconnects.

In SOFC devices cells are generally stacked and connected in series using ceramic or metallic interconnects in between two cells. Interconnects typically have a fluted structure and act as a gas separator to provide the anode with fuel and the cathode with air [11–14].

Chromium containing metallic interconnects (MICs) are among the most favored interconnect materials [12,13,15]. They represent a reasonable compromise between the numerous requirements (e.g. chemical, mechanical and electrical properties) imposed on such components [11,12,16,17]. In applied research, the degradation rate by oxide formation on MICs recently gained greater attention due to improvements of the “cycling-stability” (reduction-oxidation plus temperature (redox)) of SOFCs that was previously the major degradation source [2,18,19]. Especially for long-term degradation (>10,000 h), the continuous growth of oxide scales on MICs has been identified as the main contribution [20]. Methods are therefore needed to accurately quantify the power losses caused by thermally grown oxide layers during stack operation. Specifically, these methods should incorporate information about oxide scale growth, intrinsic material properties such as the electrical conductivity of the oxide scale and the corresponding increase of ohmic losses (area specific resistances (ASRs)) in a consistent manner. To include the effect of oxide growth as accurately as possible, the morphology of oxide scales should be taken into account as well.

Oxide scale growth is in general extracted from data of weight gain change measurements or from scale thickness determinations based on scanning electron microscopy (SEM) cross-section images [21–26]. These quantification methods have in common that they neglect chemical composition and structural information, i.e. the oxide scale morphology and the presence of inclusions and pores. A typical SEM image of an oxide scale thermally grown on CFY at 900 °C in air together with the segmented oxide scale and its resulting average thickness is shown in Fig. 1 [10]. Note the strong geometric irregularity of the oxide scale, which is typical for thermally grown scales. Note also that oxide scale growth data available in the literature often covers only few testing hours [21,22]. Scale growth data, which covers a time period >1000 h is rarely available [10,27]. However, the observed time span can strongly influence the reliability of rate law parameters fitted to the experimental data [10,28]. For the investigated interconnect materials, this is because the kinetics of the individual oxide formation mechanisms, which contribute to the scale growth do change with time [10,28,29]. During the initial phase the growth rate is independent of the consumed amount of gas and metal and therefore comparable to a scale formation process, which mainly takes place in the outer scale region and on the surface, respectively. A scale formation that is mainly driven by this mechanism can be properly described by a linear rate law [28]. Some materials especially at temperatures below 300–400 °C form a dense and blocking oxide layer which results in negligible scale growth rates after a fast initial scale formation. For such a growth behavior the oxide scale formation often

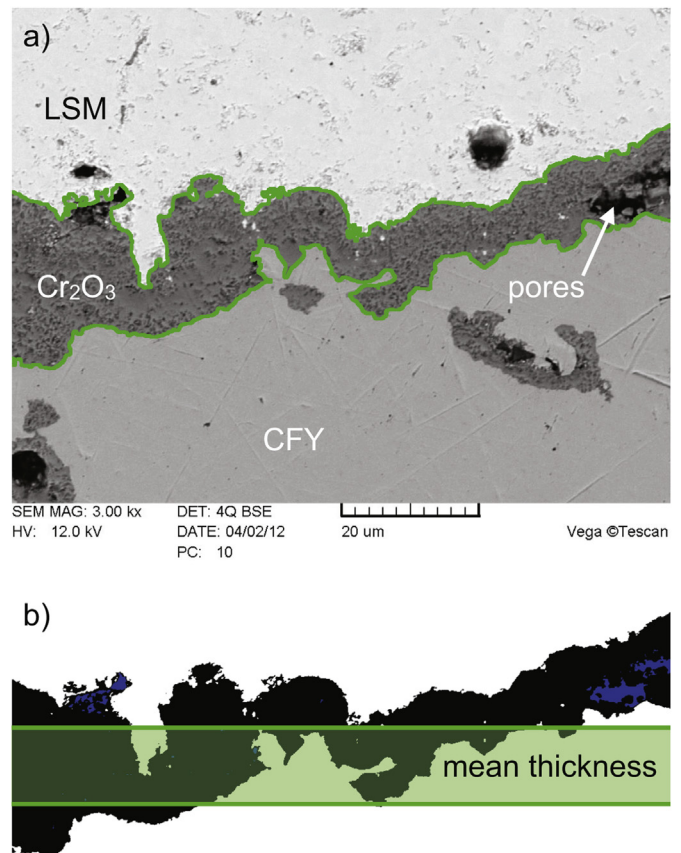


Fig. 1. a) SEM image of a metallic interconnect cross section after 10,700 h stack operation at 900 °C in air. The green perimeter marks the relevant oxide scale. b) Uniform distributed mean scale thickness (green bar) as defined from the segmented SEM image. (For interpretation of the references to color in this figure, the reader is referred to the web version of this article.)

follows a logarithmic time-dependence [28]. Kofstad [28] suggested a simple rate law that combines the different oxide formation stages, i.e.

$$\bar{x}(t) = k_e t^n. \quad (1)$$

Here, \bar{x} represents the mean scale thickness, k_e a scale growth parameter that depends on temperature and the applied atmosphere, i.e. partial pressure variation on cathode and anode side, t the operating or exposure time and n an adjustable exponent. This relation is a modification of the Wagner equation where n is 0.5 and which assumes a bulk diffusion process as rate limiting step [28]. In contrast to Wagners equation, the modified rate law is useful to describe the long-term oxide formation (>5000 h), which is usually a super position of different oxidation stages. These different oxidation stages are typically indicated by a time dependence of the fitted exponent n [10,29]. Investigations about the deviations from parabolic oxide growth usually report sub-parabolic behavior, i.e. $n < 0.5$ (see also Fig. 2) [10,13,21,29–32]. These deviations might be related to changes in chemical composition due to impurities, microstructure changes such as pore formation and grain size variation, diffusion and evaporation mechanism. Furthermore, the oxide formation process on MICs is influenced by the specific conditions during stack operation. As examples, both solid state hydrogen transport through the MIC from the anode to the cathode side [33–36], the local electrical currents resulting from the applied electrical load [25,26] and the presence of water vapor may influence the oxide growth behavior [35,37].

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