

# REDUCTION AND RE- OXIDATION KINETICS OF NICKEL-BASED SOLID OXIDE FUEL CELL (SOFC) ANODES

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## ABSTRACT

Typical solid oxide fuel cells (SOFC) consist of a dense layer of yttria-stabilized zirconia (YSZ) electrolyte and a two phase, porous mixture of Ni + YSZ as anode. In an anode-supported SOFC design, a thin film of YSZ is slurry-coated on a preform of NiO + YSZ, and the assembly is co-sintered. The as-sintered NiO + YSZ support of an anode-supported cell can be nearly fully dense, or can have some porosity. During the reduction step, NiO converts to Ni, and additional porosity is created. If the support is fully dense, the only porosity created is due to the reduction of NiO to Ni, which is usually sufficient for efficient operation of SOFC, depending upon the composition. In the actual SOFC operation, accidental stoppage of fuel flow at temperature is a likely event. It is known that should this occur, air may enter the anodic chamber, and reoxidation of the anode can occur. It is also known that cracking of the anode can occur during reoxidation, leading to the destruction of SOFC. Currently, efforts are underway at several laboratories to develop redox-tolerant anodes. Most of this work is on developing entirely new anode compositions, and in some cases metal-free anodes. In such cases, however, it is necessary to develop entirely new fabrication processes, and design around whatever shortcomings are inherent in such materials. The present approach, by contrast, is based on improving redox behavior of Ni-based anodes, so that they become more redox tolerant than the state-of-the-art anodes. The present work was undertaken with this objective.

In the present work, both reduction and re-oxidation kinetics of Ni-based anodes was investigated over a range of temperatures between 650 and 800°C. The experimental part of the investigation consisted of first fabricating NiO + YSZ two-phase composites of essentially full density. The fully dense samples were subsequently reduced in a hydrogen-containing environment. The reduced layer thickness was measured as a function of time at temperature. Subsequently, the reduced samples were re-oxidized in air at various temperatures, ranging between 650 and 800°C. In this set of experiments, the re-oxidized layer thickness was measured as a function of time at temperature. These two types of experiments were used for determining the kinetics of reduction as well as that of re-oxidation. It was observed that the reduction kinetics was linear (interface-controlled), while the re-oxidation kinetics was parabolic (diffusion-controlled). Figure 1 shows plots of reduced layer thickness vs. time at several temperatures. Figure 2

shows a plot of the square of the reoxidized layer thickness vs. time at 700°C. A simple theoretical model was developed to describe the two processes. Interface control of the reduction process was attributed to the rapid gaseous transport through the pores formed which occurs upon reduction of NiO to Ni. By contrast, diffusion control of the re-oxidation process was attributed to the existence of a very small amount of porosity, which occurs due to a slight shape change of nickel particles, when Ni re-oxidizes to NiO. The overall objective of the work is to identify compositions and/or microstructures such that the porosity created is as small as possible, in order to prevent/suppress cracking of the anode during re-oxidation.

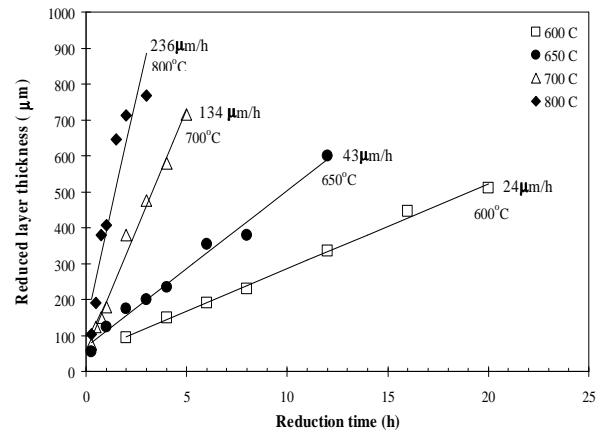


Figure 1: Plots of reduced layer thickness vs. time at various temperatures.

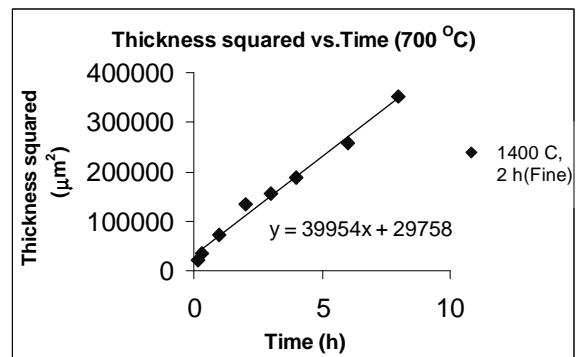


Figure 2: A plot of the square of the reoxidized layer thickness vs. time.