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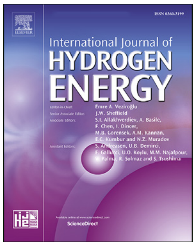

Short Communication

Degradation rate quantification of solid oxide fuel cell performance with and without Al_2TiO_5 addition

Clay Hunt [a,*], *Marley Zachariasen* [a], *David Driscoll* [a], *Stephen Sofie* [a], *Robert Walker* [b]

[a] *Department of Mechanical and Industrial Engineering, Montana State University, Bozeman, MT, 59717, USA*
[b] *Department of Chemistry & Biochemistry, Montana State University, Bozeman, MT, 59717, USA*

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ABSTRACT

Degradation rates of electrical current during constant voltage operation of SOFCs with anodes made using NiO precursor powders from two different manufacturers with and without the addition of aluminum titanate (ALT) added by either mechanical mixing or anode infiltration have been quantified using a novel MATLAB algorithm. Because the algorithm has been used to quantify degradation rates for many different SOFC tests, it is thought that the method can be applied to most measured SOFC data to quantify the instantaneous cell degradation rate as a function of time for the entire SOFC performance measurement. Degradation rates determined at different times have been plotted against varying concentrations of ALT addition, facilitating the estimation of optimum ALT concentration for SOFC anodes made with NiO from a specific manufacturer. The algorithm used to determine degradation rates is available upon request to the corresponding author.

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Introduction

Solid oxide fuel cell (SOFC) technology is improving every day. This promising means of energy conversion is fuel efficient, clean, and approaching commercialization [1]. When both electrical energy and heat are harvested from a solid oxide fuel cell (SOFC) stack, thermodynamic efficiency can be as high as 90% [2,3]. The main problem to be overcome before this technology becomes a viable source of electricity is the degradation occurring in the electrodes of the cell in the SOFC operating environment [4–12]. Because most SOFC degradation is attributed to the SOFC anode, great efforts have been made to stabilize traditional Ni yttria-stabilized zirconia (YSZ) anodes, and to develop entirely ceramic anodes [13–19].

Decreasing SOFC operating temperature is also a strategy in reducing cell degradation. This is pursued through the use of SOFCs made with the smallest possible electrode particles, and with metal-supported SOFCs [20]. The report of Ding and Hashida describes the synthesis of a nano-composite NiO-$Ce_{0.8}Sm_{0.2}O_{1.9}$ powder [21]. Anode stabilization against carbon coking and nickel coarsening through alloying the Ni in the SOFC anode with another transition metal has been reported by Kim et al. [22].

Electrodes made with small particles, or even from solution infiltration of electrode scaffolds are interesting because

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* Corresponding author.

E-mail address: clayhunt@glacigen.com (C. Hunt).

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the triple-phase boundary length of an electrode increases as electrode particle size decreases [23,24]. However, decreasing particle sizes are associated with more rapid nickel coarsening because the driving force of sintering is inversely proportional to particle size [25]. The work of Khan et al. has proposed a model for nickel-particle agglomeration occurring in Ni-YSZ anodes that is similar to equations concerned with grain size in sintering ceramics [26–28]. Cell degradation that is not consistent with Khan et al.'s predictions might be attributable to other means of cell degradation [29,30]. These results suggest that a means of quantifying degradation rate could be useful in identifying the way in which an SOFC is degrading.

Amazing techniques exist for modeling the effects on the triple-phase boundary of nickel migration in the anode of an SOFC [31,32]. However, a method of quantification of the instantaneous degradation rate of a SOFC is not the subject of much attention despite degradation being a subject of interest. The main challenge in the quantification of SOFC degradation rates is that measured electrical power output of a SOFC is difficult to differentiate in a way that provides meaningful results. The work of Wang et al., attempts to fit first-order polynomials to cell voltage that was measured during constant-current cell operation [33]. Although the lines used fit the data in the report by Wang et al., the shape of the such a voltage curve is, in general, not linear with respect to time. Only the recent report of Welander et al. uses a method of curve-fitting that can fit a larger set of fuel-cell degradation data [34]. This report elaborates on the design and merit of the technique used in the previous work and further applies the method of degradation quantification.

The impetus for developing this tool was the need to quantitatively ascertain optimum doping levels of any coarsening inhibitor for Ni-based anode catalysts [23,24,35]. Wang et al.'s method of fitting measured voltage to a line demonstrates the idea of fitting measured data to a curve to determine degradation rates from measured voltage. The method discussed in this report is simply an extension of that idea. This method can be used to quantify degradation rates of most fuel cells with data sets that record changing voltage at constant current or changing current at constant voltage. This work considers the application of this algorithm to measure the effect of the addition of Al_2TiO_5 (ALT) to the anodes of SOFCs made with ~3·μm (J.T. Baker) and ~4·μm (Alpha Aesar) initial particle size NiO on fuel cell performance.

Original application of ALT to the standard SOFC anode material, the Ni-YSZ cermet, was done to engineer the CTE of said cermet. Increased resistance to fracture of cermet bars with ALT was casually observed. This observation has been the subject of the recent work of Driscoll et al. [35]. The casual observation of anode cermet resistance to fracture also motivated the 2013 study by Driscoll et al. [24]. Results of this work suggest ALT might act as a sintering inhibitor for nickel in the anode cermet under typical SOFC operating conditions. Results of the 2013 Driscoll et al. study motivated the in-depth analysis of ALT in the anode cermet reported by Driscoll et al., in 2016 [23]. Results of this work detail the effects of thermal treatment on ALT in the cermet as studied with electrical impedance spectroscopy, X-ray diffraction, electron microscopy, and voltammetry. These results suggested the effects of ALT on cell performance might be an interesting application of the degradation-rate algorithm.

Experimental procedure

Anodes

Anodes were applied to 32·mm Fuel Cell Materials electrolytes by aerosol application of homogenized suspensions of appropriate quantities of the spherical grade 8YSZ (8YS) (8YSZ (YS) Tosoh Corp., Tokyo, Japan), NiO with initial particle size of about 3·μm (JT Baker NiO) or NiO with initial particle size of about 4·μm (Green NiO, Alfa Aesar), and corn starch, used as a pore forming agent to yield an equal-volume mixture of 8YSZ, and nickel metal after anode reduction in the fuel cell operating environment. The anode spray consisted of about 32.4 wt% YSZ, 62.2 wt% NiO, and 5.4 wt% pore former, where total powder mass is given by the sum of the masses of the pore former, YSZ, and NiO. Sprays with ALT were made by adding into the mix sufficient quantities of ALT to make 1, 2.5, 5, and 10 wt% ALT-doped (ALT, Alfa Aesar) anode mix powder.

Each suspension was thoroughly mixed in a 35 wt% xylene, 35 wt% ethanol solvent mixture using 2 wt% of copolymer dispersant (KD-1). Additionally, 2 wt% of 200 molecular weight polyethylene-glycol (PEG 200), 3 wt% of polyvinyl butyral (B98), and 1.5 wt% of butyl benzyl phthalate (S-160) were added as binders and plasticizers. The dispersant was dissolved in the solvent ultrasonically (Branson Sonifier 450) before the addition of any other ingredient. The addition of the appropriate amount of ALT to the spray mixture occurred after dissolution of the dispersant, and before the addition of any other powder. The 8YSZ, NiO, and pore former were ultrasonically mixed in the suspension after the dispersant was dissolved. Binders and plasticizers were then added, the suspension was ultrasonically mixed a third time, then ball milled in a high-density polyethylene bottle with cylindrical YSZ milling media for 12 hours to facilitate complete homogenization of the mixture.

Cell anodes were applied via airbrush (Badger air-brush Model No. 360-7) onto weighed (Sartorius CPA 225D), and appropriately masked electrolytes. The anode-electrolyte bi-layers were then allowed to dry in air overnight before being sintered at 1250 °C for two hours with heating and cooling rates of 10 °C per minute. Following sintering, the mass of each bi-layer was recorded for ALT-infiltration calculations.

ALT was added to the anode by either infiltration of an ALT solution, or by mechanical mixing of ALT powder into the anode spray, as described above. The ALT solution used for infiltration was prepared by mixing stoichiometric amounts of aluminum nitrate, and titanium lactate, both dissolved in deionized water, to produce Al_2TiO_5. The resulting ALT solution was added to the anode by pipetting a drop of the ALT solution onto the anode, allowing the water to evaporate, and removing organics by placing the infiltrated cell into a 400 °C furnace for about two minutes. The mass of the doped anode-electrolyte bilayer was then recorded to determine dopant mass. This process was repeated until desired ALT-doping concentrations were approximated. Doped and non-doped cells were then heated to 1400 °C with heating and cooling

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