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Published in:
Proceedings

Publication date:
2011

Document Version
Publisher's PDF, also known as Version of record

Determination of overpotential characteristics of reversible solid oxide cells via impedance spectroscopy and correlation with cell degradation

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Solid oxide electrochemical cells are a very promising technology for efficient, economical and sustainable energy conversion between electrical energy and chemical energy. They can be operated as fuel cells for electric power generation from fossil hydrocarbons, operated as electrolysis cells for hydrocarbon fuel production from renewable or nuclear electricity, and operated reversibly between the two modes for large-scale electricity storage (e.g. for intermittent renewable power). Today's state-of-the-art cells have high electrochemical performance. An important goal is improving the long-term durability.

The degradation of different cell components, e.g. the electrodes, is either certainly related to or suspected to be related to the overpotential across the component, depending on the cell operating conditions. For example, too high anodic overpotential for Ni-based anodes will cause Ni oxidation, dictated simply by thermodynamics. Too high anodic overpotential for oxygen-electrodes is suspected to be responsible for build up of oxygen partial pressure in electrolyte grain boundaries near the interface of the electrolyte and oxygen electrode [1]. It is therefore of great interest to determine the overpotentials during long-term operation of full cells operated as electrolyzers.

By careful measurement and analysis of impedance data measured on technological Risø DTU cells (anode-supported with thin electrolyte), we calculate current-overpotential curves without using a reference electrode. This involves analyzing impedance spectra measured at systematically varied experimental conditions – gas compositions independently varied at the fuel electrode and the oxygen electrode, temperature variation, gas flow rates and reactant utilization, and load/current density – to determine an appropriate cell model. The operating conditions for long-term testing are then applied, and the load is then incrementally stepped up to the desired potential, measuring impedance at each step. This data is fit to the model to obtain the differential resistance versus current density. This curve gives the instantaneous slope of an overpotential-current curve, and is simply integrated to obtain the overpotential-current curve. Overlaying these overpotential-current curves obtained from the AC impedance data measured under load on top of a DC polarization curve gives identical results. A similar technique was recently performed by Leonide et al [2] to calculate Butler-Volmer charge transfer coefficients. Here we use the method to study overpotential characteristics of reversible solid oxide cells and correlation between overpotentials and degradation rates.

1. R. Knibbe, M.L. Traulsen, A. Hauch, S.D. Ebbesen, and M. Mogensen, *J. Electrochem. Soc.* 157, B1209-B1217 (2010).
2. A. Leonide, Y. Apel, and E. Ivers-Tiffee, *ECS Transactions* 19, 81-109 (2009).