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Graves, Christopher R.; Sudireddy, Bhaskar Reddy; Mogensen, Mogens Bjerg

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Molybdenum Based Perovskite Electrodes for Solid Oxide Cells

Christopher Graves^{1,2}, Bhaskar Sudireddy²,
Mogens Mogensen²

¹ Lenfest Center for Sustainable Energy, and Department of Earth & Environmental Engineering, Columbia University, New York, NY, USA

² Fuel Cells and Solid State Chemistry Division, Risø National Laboratory for Sustainable Energy, DTU, Roskilde, Denmark

crg2109@columbia.edu

Recently, molybdenum-based double-perovskites, e.g. $\text{Sr}_2\text{MgMoO}_6$, were shown to have high performance for H_2 and CH_4 oxidation as anodes for solid oxide fuel cells, due to high electrocatalytic activity of the mixed valence $\text{Mo}^{5+}/\text{Mo}^{6+}$ and/or mixed ionic and electronic conductivity [1-3]. In this work, these materials are further studied, along with new compositions of molybdate double-perovskites and other perovskites with high B-site Mo content which have not been previously studied as electrode materials. We have also included W instead of Mo for a few compositions; based on the similarity of these elements, tungsten is expected to exhibit similar behavior for many properties. In addition to investigating basic material properties, the materials were studied in solid oxide cell negative-electrodes – both for fuel cell operation and for electrolysis operation.

The compounds were synthesized by solid state reaction. The phase and microstructure were characterized by X-ray diffraction, scanning electron microscopy, and energy dispersive X-ray spectroscopy. Using sintered bars, the electrical conductivity of the materials was measured with varying temperature and isothermally with varying oxygen partial pressure ($p\text{O}_2$). When changing the $p\text{O}_2$ and then going back to the prior $p\text{O}_2$, the reversibility of the conductivity measurements gave an indication about the stability of the materials (e.g. see Figure 1). Phase and microstructural analyses were also carried out after the conductivity measurements. Finally, the most promising compounds were tested as electrodes on yttria-stabilized zirconia electrolytes. Electrochemistry measurements included impedance spectroscopy and anodic and cathodic polarization curves (vs a reference electrode) in mixtures of $\text{H}_2/\text{H}_2\text{O}$ and CO/CO_2 at varying temperature from 650-900 °C.

Our results explain some unexplained phenomena from literature about the double perovskites. In general, Mo needs a partner to be stabilized against reduction to metal in the reducing atmospheres present at the negative-electrode, but it does not necessarily need to be a double-perovskite – we found other Mo-rich compounds which are also stable. Redox stability of the various B-site Mo compounds will also be discussed. This study has identified some promising new materials for use in solid oxide cell negative-electrodes.

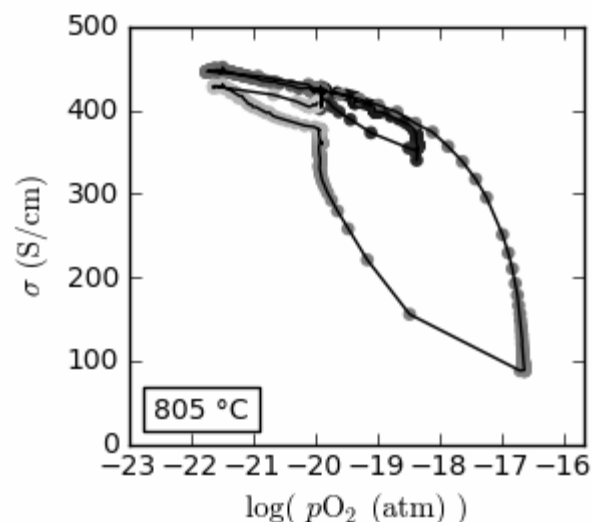


Figure 1. Isothermal electrical conductivity measurement with varying $p\text{O}_2$ of one of the compositions. One point was measured every 3 minutes and the color of the points changes from black to light gray with time.

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