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Phase relationships, electrical transport properties and redox behavior of oxides in the PrVO4-Ca2V2O7 system for SOFC applications

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Abstract:

Rare-earth and alkaline-earth vanadates attract attention as prospective materials for electrochemical applications, in particular, as redox-reversible components for fuel electrodes of solid oxide fuel cells (SOFC). An essential advantage of (Ln,A)VO_x-derived components of SOFC anodes is their anticipated resistance to carbon deposition and sulfur-containing impurities, which is critical for hydrocarbon- and biogas-fueled SOFCs. The present work was focused on the oxides of the $PrVO_4$-$Ca_2V_2O_7$ system as fuel electrode precursors, with an emphasis on phase formation, redox and thermomechanical behavior, and electrical properties.

$PrVO_4$, $Ca_2V_2O_7$ and the ceramics with the nominal composition $Pr_{1-x}Ca_xVO_{4-\delta}$ (x = 0.02-0.20) were prepared by the conventional solid-state route. Ceramics samples were sintered at 1000°C for $Ca_2V_2O_7$ and 1300°C for other materials. XRD demonstrated the formation of phase-pure $Pr_{1-x}Ca_xVO_{4-\delta}$ solid solutions with the tetragonal zircon-type structure for up to 5 at.% of calcium in Pr sublattice. At the same time, SEM/EDS suggest a lower solubility indicated by the presence of Ca-V-O phase impurities. Doping by calcium increases mixed ionic-electronic conductivity of Pr(Ca)VO_4 ceramics under oxidizing conditions. The electronic contribution is p-type and decreases with reducing $p(O_2)$. The reduction of $Pr_{1-x}Ca_xVO_{4-\delta}$ ceramics in a 10%H_2-N_2 atmosphere at 800°C leads to phase separation and formation of perovskite-like $PrVO_3$ and $CaVO_3$ phases. The redox behavior of $PrVO_4$-$Ca_2V_2O_7$ ceramics on isothermal cycling between air and 10%H_2-N_2 was studied by impedance spectroscopy, thermogravimetry, dilatometry and post-mortem XRD analysis.

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