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Oxygen ionic transport in La_2NiO_4-based membranes: thermodynamic and phenomenological modelling

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Mixed conductors derived from the K_2NiF_4-type lanthanum nickelate, $La_2NiO_{4+\delta}$, attract significant attention as promising materials for solid oxide fuel cell cathodes and ceramic membranes for oxygen separation and partial oxidation of light hydrocarbons. Important advantages of La_2NiO_4-based compositions include relatively high oxygen-ionic and p-type electronic conductivities, moderate thermal and chemical expansion, and high electrocatalytic activity under oxidizing conditions. The oxygen permeation through dense nickelate ceramics is essentially limited by kinetics of redox processes at the surface, which prevents bulk decomposition and enables stable operation under large oxygen chemical potential gradients in the intermediate temperature range. This work was focused on the analysis of steady-state oxygen transport through $La_2NiO_{4+\delta}$ membranes having different architecture. The gas-tight membranes with and without porous layers were prepared via the glycine-nitrate process and tape-casting, followed by sintering. The characterization methods included X-ray diffraction, scanning electron microscopy combined with energy-dispersive spectroscopy, thermogravimetry, dilatometry, coulometric titration, and measurements of total conductivity and steady-state oxygen permeation fluxes in a wide range of temperature and oxygen partial pressure. The data on equilibrium oxygen nonstoichiometry, conductivity and oxygen permeability were used to develop a model describing the bulk ambipolar diffusion and interfacial oxygen transfer through the membrane/gas boundary. The model was validated employing experimental data on the oxygen permeability of $La_2NiO_{4+\delta}$ membranes with different thickness, architecture and ceramic microstructure.