(P-4.40) OXYGEN DEFICIENCY, VACANCY ORDERING AND IONIC TRANSPORT IN (La, Sr)$CoO_{3-\delta}$

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Perovskite-like (La, Sr)$CoO_{3-\delta}$ possess very high level of mixed oxygen-ionic and electronic conductivity, and are thus of great interest for high-temperature electrochemical applications, including the SOFC cathodes and oxygen separation membranes. This work was centered on modeling of the steady-state oxygen permeation through dense $La_{1-x}Sr_xCoO_{3-\delta}$ (x = 0.3-0.7) membranes. The oxygen vacancy-ordering processes at reduced $p(O_2)$ were assessed by the Mössbauer spectroscopy analysis of 1% ^{57}Fe-doped cobaltites. The equilibrium $p(O_2)$-T-δ diagrams of $La_{1-x}Sr_xCoO_{3-\delta}$, collected at 973-1223 in the $p(O_2)$ range 10^{-5} – 1 atm, were adequately described using the rigid-band approach for delocalized holes and accounting for cobalt disproportionation, formation of pair clusters by Co^{2+} and oxygen vacancies, and site-exclusion effects near the positive point defects. Modeling of the permeation processes using the experimental data on the oxygen fluxes and nonstoichiometry, Wagner equation for the membrane bulk and various phenomenological expressions for the interfacial exchange showed that the surface exchange rate at the membrane feed-side surface is substantially affected by the oxygen vacancy concentration, at least for x = 0.5-0.7. At 1123-1223 K, the bulk ionic conductivity can be described neglecting the long-range vacancy ordering and Coulomb repulsion.

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