We report our research on the roles of oxygen defects on oxygen reduction reaction (ORR) kinetics of solid oxide fuel cell (SOFC) cathode. For perovskites, we developed a multi-domain 1-D physical model incorporating multi-step charge transfer to examine the competitive behaviors between the paralleled 3PB and 2PB kinetic pathways. Analyses identified the limitation of surface oxygen ion diffusion as the mechanism for 3PB-to-2PB transition. The model also proved surface reactions are driven predominantly by electrochemical forces at the 3PB, while being controlled by oxygen vacancy concentration variation at regions away from 3PB. For Ruddlesden–Popper (R–P) phases, the governing factors of the ORR are identified as oxygen adsorption and incorporation. The incorporation rate is drastically dependent on the amount of interstitial oxygen. Since the unfilled interstitial sites serve to accommodate the adsorbed oxygen during incorporation, more oxygen interstitials would seem to suppress the kinetics of this process. We proposed a physical model to reconcile the discrepancy between the experimental results and intuitive reasoning. This model illustrates a possibility of how oxygen interstitials works to regulate the exchange rate, and how the contradiction between oxygen vacancies and oxygen interstitials is harmonized so that the latter in the R–P structure also positively promotes the incorporation rate in the ORR.

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