

論文の内容の要旨

Three-dimensional numerical optimization of solid oxide fuel cell cathode microstructure
(固体酸化物形燃料電池空気極微細構造の三次元数値最適化)

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In the present study, the optimal structure of $\text{La}_{0.6}\text{Sr}_{0.4}\text{Co}_{0.2}\text{Fe}_{0.8}\text{O}_3$ (LSCF) phase inside the pure LSCF cathode is obtained using an adjoint method. Then, the electrochemical effects of $\text{Gd}_{0.1}\text{Ce}_{0.9}\text{O}_{2-\delta}$ (GDC) pillars inside the LSCF cathode is numerically investigated. Finally, the numerical optimization of the electrode-electrolyte interface shape is conducted.

Adjoint method is applied to optimize the structure of LSCF phase inside the pure LSCF cathode. The optimization is conducted until the total amount of reaction current density in the whole computational domain is maximized. During optimization, the FIB-SEM microstructure is optimized into the microstructure composed of LSCF particles with smaller particle sizes. The volume fraction of LSCF phase inside the optimized structure reduces along the thickness direction, and this gradient becomes larger for larger DPB reaction exchange current density ($i_{0,\text{DPB}}$). The LSCF is optimized into the microstructure composed of LSCF nano-particles even

starting from dense LSCF pillars which is completely different from porous LSCF microstructures. The structure transformation of LSCF-pore interface mainly takes place at the the electrolyte side. This is because the gradient of ionic potential is concentrated at the electrolyte side, and the regions near the electrolyte side is more important for the electrochemical performance than the region near the current collector side. This indicates that the porous LSCF microstructures are the best microstructure which can maximize the cell performance. What is more, the size of LSCF particles is the same even for different initial structures, which also implies that the particle size is crucial to enhance the cell performance.

In order to investigate the electrochemical effects of GDC pillars inside the LSCF cathode, three-dimensional pure LSCF and LSCF-GDC composite cathode microstructures with different particle sizes are reconstructed by fucused ion beam scanning electron microscopy (FIB-SEM), and the GDC pillars with different pillar lengths are inserted into these microstructures. According to the numerical simulation, it is shown that the GDC pillars can improve the cell performance by expanding the reactive thickness and by improving the ionic conductivities. The performance improvement is more pronounced for smaller particles, because the GDC pillars can expand the reactive regions more effectively for the microstructure with small particle size. In addition, the performance improvement is more pronounced for pure the LSCF cathode than the LSCF-GDC composite, because the GDC pillars more efficiently improve the effective conductivity for pure LSCF cathodes. In addition, if the GDC pillar is too long, the pillars replace the electrode region which is originally conductive, and the performance improvement effect is suppressed.

In order to optimize the shape of the interface between cathode and electrolyte, the computational domain is divided into the electrolyte region and electrode region. In the electrode region, electrode phase and pore phases are assumed to be homogeneously distributed. The electrolyte region is composed of electrolyte materials with high ionic conductivity.

Electrochemical reactions are assumed to take place at both electrode region and electrode-electrolyte interface. The interface shape between the pure LSCF cathode and GDC electrolyte is first optimized. It is found that the cathode with optimized interface structure has better electrochemical performance than the microstructure with flat interface. In addition, the volume of optimized electrolyte region structure is independent from the computational domain and the initial microstructure shapes.

In order to give guidance to the microstructure design with various electrolyte materials and with various electrode microstructures, the influence of dimensionless numbers on the optimized microstructure is investigated. It is analyzed that the distribution of ionic electrochemical potential is controlled by three dimensionless numbers, k , $\Gamma_{\text{electrode}}$, and $\Gamma_{\text{interface}}$. The change in the dimensionless number k is realized by changing electrolyte ionic conductivity ($\sigma_{\text{O}^{2-}, \text{electrolyte}}$). The optimized electrolyte region size becomes smaller with larger $\sigma_{\text{O}^{2-}, \text{electrolyte}}$. The change in the dimensionless number $\Gamma_{\text{electrode}}$ is realized by changing the electrode region reaction coefficient ($\beta_{\text{electrode}}$). The optimized electrolyte region size becomes larger with larger $\beta_{\text{electrode}}$. This is because the reactive thickness of the microstructure becomes thinner with larger $\beta_{\text{electrode}}$. The change in the dimensionless number $\Gamma_{\text{interface}}$ is realized by changing the interface reaction coefficient ($\beta_{\text{interface}}$). The optimized electrode-electrolyte interface area becomes larger with larger $\beta_{\text{interface}}$.