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Three dimensional electrochemical simulation of solid oxide fuel cell cathode based on microstructure reconstructed by marching cubes method

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HIGHLIGHTS

- 3D numerical model based on marching cubes geometry is conducted.
- Partial bounce-back scheme is applied to maintain MC geometry details.
- Local effective conductivity is applied in each grid to maintain MC geometry details.
- The proposed model improves computational accuracy by better illustrate the topology.
- The proposed model works better with larger ratio of partial filled grids.

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ABSTRACT

In the present study, a model is introduced to correlate the electrochemical performance of solid oxide fuel cell (SOFC) with the 3D microstructure reconstructed by focused ion beam scanning electron microscopy (FIB-SEM) in which the solid surface is modeled by the marching cubes (MC) method. Lattice Boltzmann method (LBM) is used to solve the governing equations. In order to maintain the geometries reconstructed by the MC method, local effective diffusivities and conductivities computed based on the MC geometries are applied in each grid, and partial bounce-back scheme is applied according to the boundary predicted by the MC method. From the tortuosity factor and overpotential calculation results, it is concluded that the MC geometry drastically improves the computational accuracy by giving more precise topology information.

1. Introduction

Solid oxide fuel cell (SOFC) is a very promising technology for power generation because of its high efficiency and fuel flexibility [1–4]. On the other hand, high operation temperature (700–1000 °C) of SOFC significantly limits the choice of the component materials. Reduction of the SOFC operation temperature is attractive for its ability to reduce the system cost and to enhance cell durability [5,6]. However, operation at intermediate temperatures, e.g. 500–700 °C, causes deteriorations of the ionic conductivities and polarization characteristics [7–9]. Therefore, it is important to develop SOFC electrode materials which are efficient under such intermediate operation temperatures. Mixed ionic and electronic conductor (MIEC) cathode materials such as $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_3$ (LSCF) have attracted large attention because of its high conductivities and electro-catalytic properties even at low operation temperatures [10–14].

The electrodes are multiphase porous structures in which gas, ion and electron transfer, and electrochemical reaction take place. Numerical models are often used to predict the electrochemical performance, and to investigate the intrinsic mechanisms of the electrode reaction [15–18]. Accurate and effective numerical tools are of great help for developing new designs and manufacturing processes of the SOFCs [15,16]. Focus ion beam scanning electron microscope (FIB-SEM) [19–22] attracts much attention since it enables direct three dimensional (3D) reconstruction of real SOFC electrode microstructures. The 3D reconstruction provides detailed information to obtain important electrode microstructure parameters. For instance, Gostovic et al. [19] calculated cathode surface area, triple phase boundary length, and tortuosity based on FIB-SEM reconstructed structure. Iwai et al. [20] evaluated triple-phase boundary (TPB) density and tortuosity factor in a Ni-YSZ 3D microstructure reconstructed by FIB-SEM.

In addition, the 3D microstructures can provide more valuable information to evaluate the electrochemical properties when combined with the numerical simulation tools. Among the electrochemical simulation tools, lattice Boltzmann method (LBM) [21–23] is attractive for its capability to solve flows in complex geometries. Suzue et al. [21]

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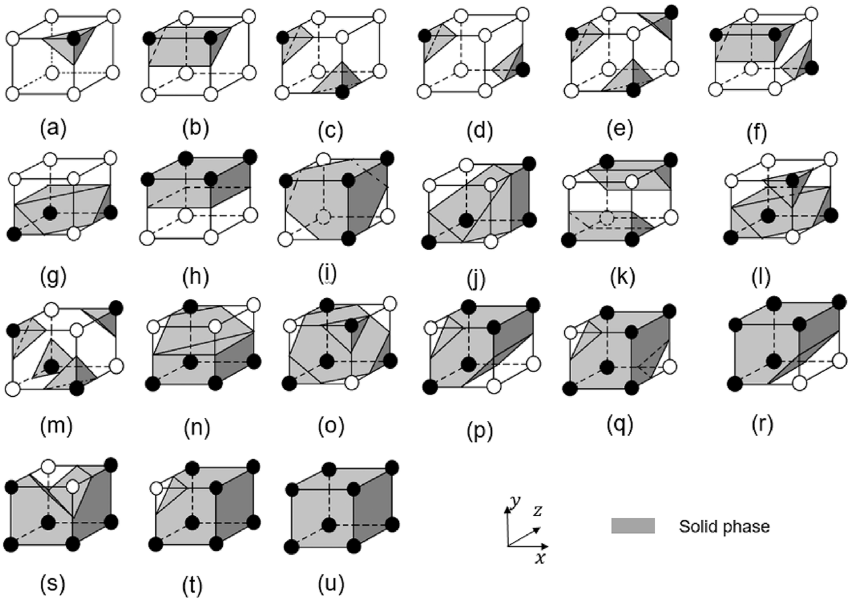

Fig. 1. Twenty-one patterns used in the marching cubes method. Gray is the estimated solid phase, and the transparent part is the pore.

have conducted 3D LBM simulation to assess SOFC anode performance. Shikazono et al. [22] used LBM to solve transport equations and electrochemical reactions inside 3D microstructures reconstructed by FIB-SEM.

In these simulations [21,22], the microstructures are composed of voxel cubes, since the microstructures obtained from the 3D tomographic scans are directly applied in the simulations. However, microstructure made up of voxel cubes poorly represents the real geometry by exhibiting a stair shaped structure [24]. Thus, in order to improve the computational accuracy, efforts should be made to improve the topology properties of the 3D microstructure geometry.

Marching cubes (MC) method [25] considers 21 patterns to reconstruct the geometry from the phase information of neighboring 8 voxels as shown in Fig. 1. It draws attention because the MC topology of the porous microstructure is more illustrative. Accordingly, it is expected that by using the MC reconstructed structure in the numerical simulation, e. g. LBM, the computational accuracy can be improved.

Matsuzaki et al. [23] introduced a numerical model to predict the electrochemical performance of LSCF cathode. They computed the double phase boundary (DPB) density based on the microstructure reconstructed by the MC method, and the accuracy of electrochemical reaction simulation is enhanced. However, the diffusions and conductions in Matsuzaki et al. are solved based on the microstructure with voxel cubes, i.e. their model did not make the best use of the MC geometry.

In most of the conventional LBM simulation solving porous microstructures [21–23], halfway bounce-back scheme is applied based on the microstructure formed by voxel cubes. In the simulation by Ahrenholtz et al. [24], a partial bounce-back scheme with the boundary reconstructed by the MC method is applied, which improved the calculation precision. Halfway bounce-back and partial bounce-back schemes are applicable only when the conduction or diffusion in a grid is completely prevented in the direction of interest. However, many of the MC geometry patterns as shown in Fig. 1 do not completely block the conduction or diffusion through the grid, which means that applying partial bounce-back scheme is not preferable in such partly conductive grids. For example, partial bounce-back is helpful to represent the geometry (b) in Fig. 1. This is because if the gray phase is conductive, the conductions through the grid in y and z directions should be zero in this MC pattern. On the other hand, the partial bounce-back is not suitable to represent the geometry patterns such as Fig. 1 (q), (s), and (t), because in these patterns the conductions through the grid exist in all directions. Therefore, additional schemes are required to fully represent the MC patterns.

Recently, He et al. [26] introduced a novel electrochemical model by applying effective conductivities in each computational grid, and the effective conductivities are obtained based on sub-grid geometric information. According to the numerical results, they succeeded to maintain the sub-grid geometric information even when coarser computational grid is used. It is expected that the MC geometric information can be maintained by applying the effective conductivities and diffusivities in each computational grid.

In this study, a numerical model to correlate the electrochemical properties of the SOFC cathode with the 3D microstructure reconstructed by the MC method is introduced. During the simulation, local diffusivities and conductivities in each computational grid are obtained by solving the diffusion equation based on the MC geometry. For verification, tortuosity factor and overpotential of a LSCF cathode are calculated using microstructures composed of voxel cubes and the marching cubes method.

2. Sample preparation and reconstruction process

In the present study, LSCF cathode microstructure which was reconstructed by FIB-SEM in our previous work [27,28] is used. Cathode was sintered at 1150 °C for 1 h. The sample was infiltrated with epoxy resin (Marumoto Struers KK) under vacuum so that pores can be easily distinguished during the post processing of the images. An Ar-ion beam cross section polisher (JEOL Ltd., SM-09010) was used to polish the sample to make the cross-section for FIB milling. Three-dimensional microstructure reconstruction of cathode sample was conducted by FIB-SEM (Carl Zeiss, NVision 40). EsB Detector was applied with acceleration voltage of 1.5 kV. After repeating FIB milling and SEM imaging processes, a series of cross section images with a pixel size of 25.9 nm and an interval of 49.6 nm was obtained. LSCF and pore phases are segmented based on the brightness values of the images. After filtering, the gray scale of the pore phase infiltrated by epoxy resin becomes 0 and the gray scale of the solid phase becomes 102–120, which makes it possible to distinguish the solid and pore boundary during binarization [23]. As the cubic computational grid is required for LBM and the cross-section images have a pixel size of 25.9 nm and an interval of 49.6 nm, the reconstructed 3D structure is coarsened to a voxel size of 50.0 nm to