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Identification and assessment of three-phase boundaries in porous electrodes of solid-oxide electrolysis cells based on a 3D microstructure model

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Hydrogen energy is regarded as a promising energy carrier due to its high energy density and non-pollution. Solid oxide electrolysis cells (SOECs) have been studied extensively as a promising way for massive hydrogen production from renewable but unstable energy sources. The electrode microstructure of SOECs has a significant influence on their electrochemical performance. To better understand the relationship between their microstructures and electrochemical performance, the quantification of key microstructural parameters such as the three-phase boundary (TPB) density and phase connectivity are required. The density and activity of the TPB sites are crucial in determining the electrochemical performance of SOEC electrodes. Therefore, it is also important to find the quantitative relationships between the active TPB density and electrochemical performance. Many efforts in microstructural analyses of SOEC electrodes via focused ion beam-scanning electron microscopy (FIB-SEM) have provided a great opportunity to link the microstructural properties to the electrode performance and the active TPB density is usually evaluated from geometrical models. However, due to the influence of conductivity and mass transfer of the gas phase, not all TPB identified from the geometric models are electrochemically active. Also, it is hard to quantitively calculate the active TPB density linking to the charge transfer. Here, we developed a comprehensive method to distinguish the active and inactive TPB which combined the image-based method and finite-element (FE)-based method. A 3D pore-scale model based on the real three phases and TPB lines was built to capture mass transfer, electron/ion transfer, and electrochemical reaction processes. The TPB sites were assessed with their contributions to the total current of the electrodes. By comparing the active TPB densities calculated from geometrical models (image-based method) and that of the developed model in this study (combined image and FE method), the proposed model was more accurate in predicting electrode electrochemical performance. This provides an effective way to reduce experimental costs and time but also deepens our understanding of the microstructure of porous SOEC electrodes.

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