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Study of the tortuosity factors at multi-scale for a novel-structured SOFC anode

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Abstract. Gas transport properties are closely related to the tortuosity of the pore network within porous materials. For the first time, this study explores a multi-scale imaging and modelling method to measure the tortuosity of an Solid Oxide Fuel Cell (SOFC) electrode material with pore sizes spanning over hundreds of orders of magnitude. This analysis is normally challenging using image-based techniques, as pores of different sizes may not be easily resolved at the same time using X-ray computed tomography (CT). In this study, a tubular SOFC anode, fabricated by a phase inversion technique, is used to illustrate this approach. A heat flux analogy is used to simulate mass transport and the results show that the embedded large-scale finger-like pores can significantly improve mass transport by providing less tortuous pathways.

1. Introduction

Gas transport through porous anodes is critical to the electrochemical performance of solid oxide fuel cells (SOFC) as the partial pressures of the reactants and products are closely related to the anode polarization, which significantly contributes to the loss of operating voltage. The porosity (ε) and tortuosity of the gas transport path each play a significant role in determining the diffusivity of the fuel gas in the porous anode [1]. The tortuosity factor (τ) is a material parameter used to characterise gas diffusion resistance due to the tortuous pore volume [2, 3]. This effective transport parameter can be obtained by the modelling of mass/heat flux through a 3D porous phase using computational fluid dynamics (CFD) methods [4].

A newly developed tubular SOFC fabricated by phase inversion technique has attracted considerable attention [5]. Apart from the controllable pore size in the spongy layer, the exchange of solvent and non-solvent in the hollow fiber generates micro-channels which grow in the radial direction, with a diameter of approximately 20 μm. This design can significantly improve gas transport in the anode with little sacrifice of mechanical robustness. In this study, non-destructive X-ray CT is used to image the SOFC anode at micrometer and nanometer resolution for the whole anode and the porous phase in the spongy layer, respectively. The tortuosity factor extracted from the high resolution scan was subsequently used for the simulation of the entire anode.

2. Methodology

2.1 Tortuosity Factor Measurement

The analogue between heat and mass transfer is well established [6] and is regularly used to measure the tortuosity factor. The heat flow driven by a temperature gradient in a porous material is described using modified Fourier's law as

$$Q_p = -A\frac{\varepsilon}{\tau}k\frac{\Delta T}{L} \tag{1}$$

Where A is the cross sectional area of the flow volume, k is the thermal conductivity, ΔT is the temperature difference and L is the length of flow volume, ε is the porosity and τ is the tortuosity factor. As ε can be measured by image analysis, by dividing Q_p by the heat flow in a fully porous volume Q (i.e. $\varepsilon = 1$, $\tau = 1$), τ can be obtained. For simplicity, the thermal conductivity of the micro-channels are set to unity (i.e. $k = 1$), so that the thermal conductivity is equivalent to the effective transport parameter ε_s/τ_s for the spongy layer.

2.2 X-ray Computed Tomography

The entire tubular anode and the spongy layer were imaged using a Versa 520 and Ultra 810 X-ray microscope (Zeiss Xradia, Carl Zeiss, CA, USA) respectively [7]. Detailed scanning parameters are shown in Table 1. FDK and filtered-back projection algorithms were used for the reconstruction of the full anode and the spongy layer scans respectively [5, 8]. The reconstructed sample volumes were segmented using the Avizo V9.0 software (VSG, Bordeaux) package.

Table 1. Scanning parameters for the spongy layer and entire anode

	Tube target	Voltage (kV)	Voxel size (μm)	Field of view (μm^2)	Projections	Exposure time (s)	Camera binning
Spongy	Cr	35	0.032 (binned)	16×16	1201	60	2
Anode	W	140	1.07	2000×2000	2001	18	1

2.3 Computed Dynamics Fluid (CFD) Simulation

The segmented porous phase of the spongy layer was imported into the CFD software Star-CCM+ (CD-adapco Inc.) for meshing followed by heat flux simulation. ε_s/τ_s was then used to define the effective thermal conductivity of spongy layer in the simulation of the entire anode on a control volume with full radial thickness, an axial depth of 700 μm and 90° arc angle (see Figure 1). The detailed simulation parameters are shown in Table 2.

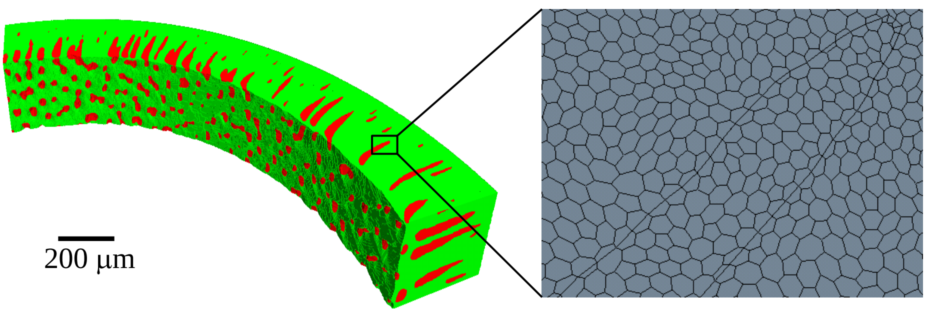

Figure 1. The effective thermal conductivities of the micro-channels (red) and spongy layer (green) are assigned as unity "1" and ε_s/τ_s respectively. Conformal meshes are generated between the two regions.

Table 2. CFD simulation parameters for spongy layer and entire anode

	Spongy layer	Full anode
Number of cells	1.2 million	1.6 million
Inlet temperature (℃)	1000	1000
Outlet temperature (℃)	300	300
Thermal conductivity	ε_s/τ_s	1
Sample dimension (μm^3)	9×6×4	800×800×800

3. Results and discussion

A virtual slice of the anode is shown in Figure 2a. The radially aligned micro-channels are clearly resolved. The porous phase in the spongy layer, which appears as solid phase in Figure 2a, is segmented from the CT data and imported to Star-CCM+ to simulate the heat transfer driven by the temperature difference from 1000 K (LHS) to 300 K (RHS) (Figure 2b). The porosity and the mean diameter of the porous phase in the spongy layer are measured to be 0.19 and 200 nm respectively by image analysis. The heat flux in the 3D microstructure is shown in Figure 2c. It is observed that the tortuosity and constrictivity of the pore network generate a heterogeneous distribution of the flux, and the local flux maxima appear at the position where the transition of the pore thickness is sharp in the pathway. By integrating the heat flux on the cross-sectional plane, and dividing it by the heat flow on the cross-sectional plane of the empty volume, the material parameter ε_s/τ_s is equal to 0.015, which is subsequently used as the thermal conductivity of spongy layer in the anode simulation.

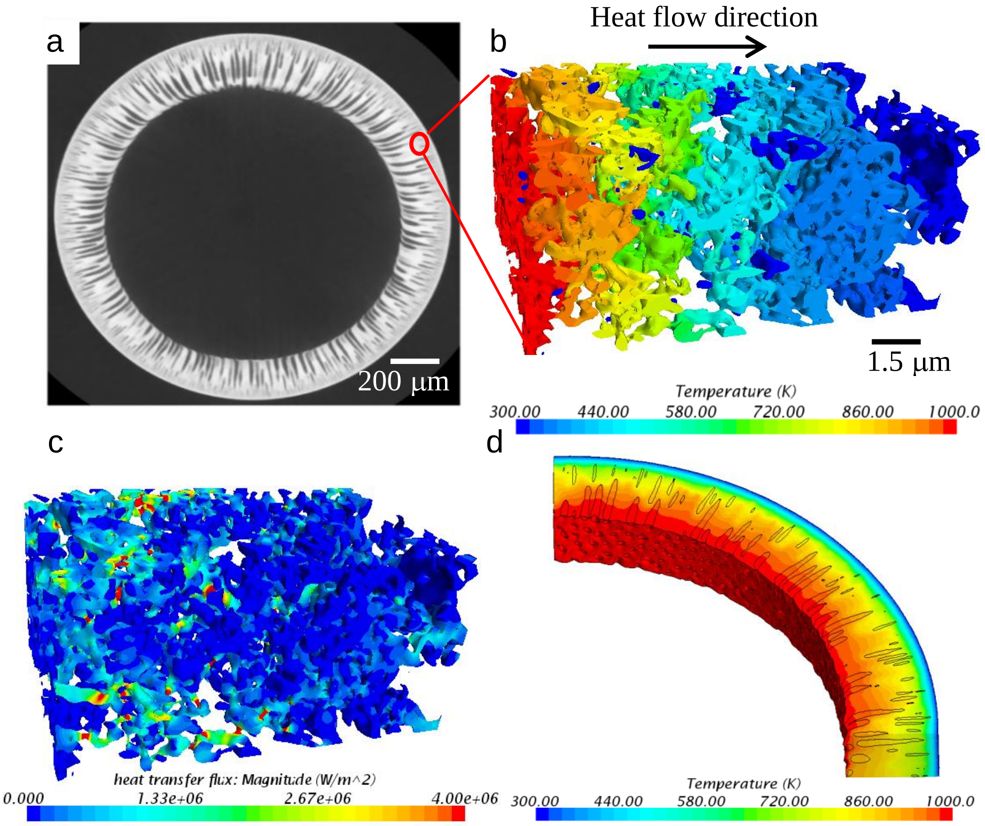

Figure 2. (a) Virtual ortho slice of the anode; (b) simulation of heat transfer in the spongy layer; (c) heat flux in the spongy layer; (d) Heat transfer simulation on the anode with different thermal conductivity defined in the micro-channel and spongy layer.

The result of the heat transfer simulation for the whole anode is shown in Figure 2d. The temperature contour displays a non-uniform distribution with a ragged edge where there are isolated micro-channels. These are caused by the discrepancies of the effective thermal conductivity between the micro-channels and the spongy layer. As the heat flows from one micro-channel to the other, temperature gradient occurs at the spongy layer between them due to the low thermal conductivity.

The microstructure parameters extracted from the anode heat transfer simulation are compared with those in the spongy layer in Table 3. With the micro-channels embedded in the anode, the macroscopic material parameter for the whole anode ε_a/τ_a is approximately 4 times as large as ε_s/τ_s, indicating a 300 % increase in terms of heat flow.

Table 3. Comparison of the material parameters measured in spongy layer and full anode

	ε	τ	ε/τ
Spongy	0.19	12.1	0.015
Anode	0.33	5.2	0.063

4. Conclusion

Multi-scale imaging and hierarchical simulation is an effective method to extract the material parameters (i.e. porosity and tortuosity) in the samples with distinctly different pore sizes spanning over several orders of magnitude, under which circumstances the porous phases cannot be resolved at the same time. These geometrical parameters are critical for effective diffusivity estimation in macroscopic electrochemical simulations. Results show that mass transfer in novel-structured tubular SOFC anodes embedded with radially aligned micro-channels is significantly improved four-fold due to the decrease of global tortuosity compared to the traditional homogeneous tubular SOFC anode with the pure spongy layer.

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