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X-ray micro-computed tomography and tortuosity calculations of percolating pore networks

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Abstract

Synchrotron source X-ray micro-computed tomography was used for non-destructive three-dimensional (3-D) imaging of porous alumina structures, in which the porosity was induced by a granular porogen, added in amounts of 10–60 vol.%. Microstructural characteristics related to transport properties, including connectivity and tortuosity, were measured from the resulting 3-D data sets. Connectivity of 94.5–99.6% was measured for samples produced with 35–60% porogen (30.8–49.6% porosity). Two methods of calculating tortuosity, path length ratio and gas phase flux were compared, and the effect of sample volume on calculated tortuosity value and computational time was examined. Average sample tortuosity calculated using the two methods generally agreed, although significant directional anisotropy was detected in some cases for the gas phase flux calculation method. Tortuosity values as low as 1.5 were measured for alumina components with 49.6% porosity.

Keywords: Tortuosity; Porosity; Transport properties; X-ray computed tomography; Finite difference modeling

1. Introduction

Interconnected porosity has many roles in ceramic components, from modifying electrical, mechanical and thermal properties to serving as a transporting phase. A diverse range of applications require porous ceramic components, including filters, catalysts and catalyst supports, reaction chambers, heat exchangers, thermal insulators, electrical insulators, composite preforms and biomedical implants and devices [1]. In addition, many structural components rely on porosity for damage-tolerant coatings, strategically weak interlayers and tailored elastic properties [1,2]. Characterization of the volume fraction, size, morphology and connectivity of porosity in engineered components is vital for understanding its effects on material properties and for optimizing processing conditions to yield desired microstructures and associated effective properties.

1.1. Pore network characterization

Percolating pore networks can be characterized by a wide range of parameters, from simple volume fraction and connectivity measurements to more complex

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geometrical constructs, such as tortuosity [3,4], and topological or homological indices, such as Genus and Betti numbers [5,6]. Tortuosity, the measure of the "curviness" of a path, is a convenient parameter, as it captures the relative deviation from linearity of networked paths independently of any other geometrical characteristics such as pore shape and size scale. For many applications, the desired pore network structure can be dictated in terms of tortuosity. For example, controlled-release pharmaceutical delivery devices require high tortuosity to regulate diffusion [7], while high throughput catalyst supports, filters, heat exchangers and similar components generally benefit from low tortuosity and correspondingly high permeability [8]. Several mathematical definitions of tortuosity have been suggested in the literature, based on geometric constructs [3,4,9] as well as conductivity [9–11], diffusivity [3,12–16] and permeability [8,17] relationships. These definitions range from simple path length ratios (PLR) [3,4] to quantities calculated from results of simulations based on differential equations describing three-dimensional (3-D) fluid flow or diffusion [8,9,12,17].

1.2. Characterization techniques

A number of analysis techniques are available to examine aspects of pore networks, including the Archimedes buoyancy method, BET analysis, pycnometry, mercury porosimetry, microscopy and 3-D imaging. Three-dimensional imaging includes destructive techniques such as serial sectioning or polishing combined with optical microscopy [18] as well as focused ion beam milling combined with scanning electron microscopy (SEM) [8]; it also includes non-destructive methods such as X-ray microcomputed tomography (μCT) [19,20]. The resulting 3-D images can be used directly for measuring pore connectivity, size, surface area and phase volumes. Skeletal and bulk densities can be calculated based on volume fraction if the solid phase density is known. Tortuosity can be measured from skeletonized data or by simulating transport using finite element analysis or finite difference calculation on meshed data (see, for example, Ref. [8]).

In the present work, pore size, connectivity and tortuosity of tailored porous alumina samples are measured from 3-D data sets produced by μCT using synchrotron X-radiation. Two methods of calculating tortuosity are compared and their relative merits examined.

2. Procedure

2.1. Sample preparation

Porous alumina samples were produced by thermoreversible gelcasting [21–24]. Base gels of 6.3 wt.% PMMA–PnBA–PMMA copolymer (Kuraray America, Inc.) in isopropyl alcohol (IPA; ACS grade, BDH Chemicals) were produced by ultrasonic mixing in a 70 °C water bath. Alumina powder (HP-DBM, Baikowski Malakoff, Inc.), polypropylene (PP) granular porogen with 35 μm mean particle diameter (Polysciences, Inc.) and Aerosol AY-65 dispersant (Cytec Industries, Inc.) were added in four equal loadings to form a slurry with 50 vol.% solids (of which 40–90 vol.% was Al_2O_3 and 10–60 vol.% was PP) and 0.02 g dispersant per gram solids. The slurries were mixed ultrasonically in a 70 °C bath after each loading for ~10 min.

Cylinders up to 10.5 mm in diameter and 12.5 mm tall were cast by filling nylon molds. After the cast samples cooled and gelled, they were removed from their molds and slowly dried in a concentrated IPA atmosphere for 1–2 days before being allowed to dry fully in air. Once dry, the samples were burned out and pre-fired in a vented furnace by heating at a rate of 10 °C min^{-1} to 1000 °C with 1 h soaks at 600 °C and 1000 °C. Samples were then sintered in a high-temperature furnace by heating at a rate of 10 °C min^{-1} to 1600 °C with a 1 h soak, resulting in samples containing 9.6–49.6% porosity, depending on the volume fraction of the porogen. Cylindrical samples with a diameter of 1.5 mm or 3 mm and height $\geqslant$5 mm were machined from sintered disks using an ultrasonic drill press and a diamond-coated coring bit.

2.2. X-ray μCT

X-ray μCT was performed at station 2-BM of the Advanced Photon Source at Argonne National Laboratory [25] for several different cycles. As a result, imaging parameters varied, as discussed below, but did not affect the numerical results obtained. The 3-mm-diameter samples were mounted 75 mm from the detector, while the 1.5-mm-diameter samples were mounted 15 mm from the detector. Radiographs using 22, 27 or 29 keV photons captured ~3 mm (horizontal) × 1.5 mm (vertical) projections. Each sample was rotated 180° in 1/8° increments. At each step, up to four exposures were taken and averaged. The exposure time was ~0.25 s, which gave a total imaging time of ~20–40 min for a typical sample. A 2048 × 2048 pixel CCD detector was used in combination with a 4×, 5× or 10× optical lens, giving cubic voxels with edge lengths ranging from ~0.8 μm to 1.9 μm. The two-dimensional (2-D) cross-sectional images, oriented along the cylinder axis, were reconstructed using the 2-BM software [25]. The 2-D images were then stacked to reconstruct the 3-D microstructure, which was re-sliced in the two orthogonal directions for additional 2-D analysis of pore sizes and orientations using particle measuring tools built into ImageJ software [26].

In order to analyze the 3-D reconstructed volumes quantitatively, binary segmentation was performed using Adobe® Photoshop® CS2 software (Adobe Systems, Inc., San Jose, CA) by equalizing the grayscale images, applying built-in noise reduction filters, and thresholding to selected levels, resulting in pore fractions closely matching those of each sample as measured by Archimedes' buoyancy method [27]. Once segmented, the images were imported