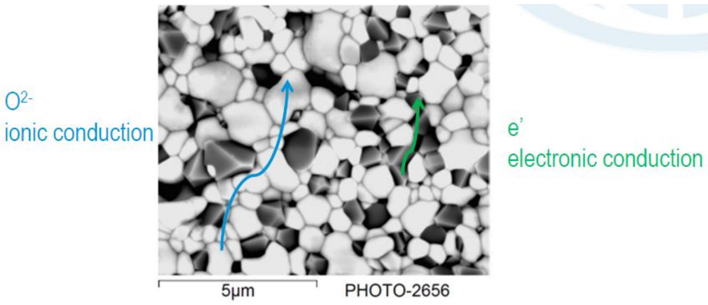
11th World Congress on Computational Mechanics (WCCM XI)
5th European Conference on Computational Mechanics (ECCM V)
6th European Conference on Computational Fluid Dynamics (ECFD VI)
July 20 - 25, 2014, Barcelona, Spain

CONFORMAL COMPUTATION OF OXYGEN FLUX IN HETEROGENEOUS MIXED-CONDUCTOR MATERIALS

Fazle Rabbi,[1] Kyle Brinkman,[2] Ken Reifsnider[1]

[1] University of South Carolina, Columbia, SC 29209, Reifsnider@sc.edu, www.HeteroFoaM.edu
[2] Clemson University, Clemson, SC 29634,   E-mail address and URL

Key Words: *Instructions, Multiphysics Problems, Applications, Computing Methods.*



$$JO_2 = \gamma \frac{RT}{16F^2 L}\int_{\ln PO_2'}^{\ln PO_2''} \frac{\sigma_e \sigma_i}{\sigma_e + \sigma_i} d\ln PO_2$$

Figure 1

Brinkman, et al., have addressed the microstructural design and characterization of 3D inter-connected microstructural net-works in mixed ionic and electronic conducting ceramic composite membranes.[1-6] That work has recently focused on mixtures of conductive phases that clearly show the dependence of oxygen flux on microstructure features as well as constituent and interface behavior (Figure 1). An interesting result of this effort is the fact that a systematic, monotonic change in the volume fractions of the electronic and ionic conducting phases does not necessarily produce a corresponding order of observed flux changes (see example in Figure 2). This anomalous behavior presents an obstacle to the understanding of the fundamental physics of the behavior and to the development of design concepts and analyses that enable the design and development of such materials and membranes. Wagner's equation, as shown in Fig. 1 for example, is clearly not sufficient for this purpose.

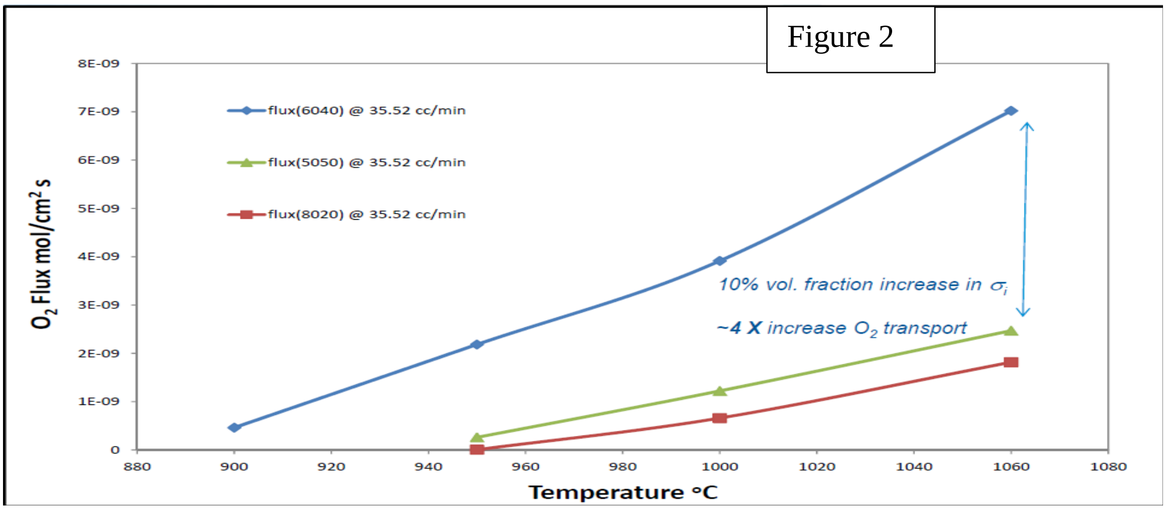


Figure 2

Over the last four years, we have constructed a general approach to the prediction and observation of charge distributions in mixtures of conductors and dielectric materials. We have successfully predicted permittivities for a control material that was anisotropic, as a function of volume fraction and orientation of meso-constituents, as well as the interfacial surface charge for this system at that scale.[5] During that time, we have also observed anomalous oxygen transfer in MIEC membranes wherein the microstructure is at the sub-micron scale, that is a strong function of PO_2, temperature, and micro-morphology. In the present paper, we will apply our methodology to understand emergent oxygen flux in a micro-mixture.

First principles modeling of real microstructures with 3D models capable of solving Maxwell's equations to calculate impedance, currents, and voltage distributions of a given heterogeneous domain have been developed by our team using COMSOL. For impedance measurements and voltage distributions of regular geometrical meso-structures, the model shows agreement with experimental work.[5] To model 3D irregular shaped *nano-structures* at the conformal level, 3D domains were created from x-ray nano-tomography images using the commercial code Simpleware. The code generates 3D meshes from image slices generated from x-ray nanotomography data. In our continuing work, this model is further developed to incorporate a modified Wagner's equation at the local, conformal level, to predict collective flux through a membrane. This modified equation will consider constituent conductivity as well as interfacial interactions between the constituents that give rise to emergent properties that are different from the sum of the individual constituent properties. Our approach also has the capability to assign different impedance and conduction properties to boundaries (or finite boundary regions) between two different constituent materials, which can be used to represent interfacial effects. The resulting individual and global properties i.e., ion flux, conductivity, permittivity etc., can be measured over a wide frequency and temperature range.

REFERENCES

1. Q. Liu, "Directly resolving particles in an electric field: local charge, force, torque, and applications," *Int. J. Numer. Meth. Engng.*, vol. 90, pp. 537-568, 2012.
2. Q. Liu, "Physalis method for heterogeneous mixtures of dielectrics and conductors: Accurately simulating one millionparticles using a PC," *J. Comput. Phys.*, vol. 230, pp. 8256-8274, 2011.
3. Q. Liu and K. L. Reifsnider, "Heterogeneous Mixtures of Elliptical Particles: Directly Resolving Local and Global Properties and Responses," *Journal of Computational Physics*, vol. 235, pp. 161-181, 15 February 2013.
4. K. L. Reifsnider, W. K. Chiu, K. S. Brinkman, Y. Du, A. Nakaja, F. Rabbi and Q. Liu, "Multiphysics Design and Development of Heterogeneous Functional Materials for Renewable Energy Devices: The HeteroFoaM Story," *J. Electrochemical Society*, vol. 160, pp. F470-F481, 2013.
5. Jeffrey Baker*, Jon Michael Adkins, Fazle Rabbi, Qianlong Liu, Kenneth Reifsnider, and Rassel Raihan, "Meso-Design of heterogeneous dielectric material systems: structure property relationships," JOURNAL OF ADVANCED DIELECTRICS, Vol. 3, No. 1 (2013) (accepted)
6. Brinkman K., Iijima T., Takamura H., Tuller H., *"The Preparation and Oxygen Permeation Properties of Nanocrystalline CeO2 Thin Films on Porous Cerium Oxide Substrates*", **Journal of the Electrochemical Society** Vol. 157 Issue No. 12 page B1852-B1857 (2010).