Symposium | A. Advances in Materials Theory for Multiscale Modeling

[SY-A10] Symposium A-10

Chair: Steve Fitzgerald(University of Leeds, UK)
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[SY-A10]Mesoscale understanding of ionic conduction in yttria stabilized zirconia

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Yttria-stabilized zirconia (YSZ) is widely used as an electrolyte in solid oxide fuel cells (SOFCs). Much of past research on ionic conductivity in YSZ has focused on understanding diffusion process atomistically, e.g., migration barriers for O^{2-} ion movement and vacancy trapping behavior, in an attempt to explain the bulk O^{2-} transport. How the mesoscale structure of YSZ, i.e., structures at length scales lying between the atomic/micro-scale and the macroscale, influences ionic conduction remains poorly understood. An improved understanding of mesoscale factors that affect ionic conduction in YSZ can potentially benefit our efforts to search for new SOFC electrolyte materials.

Two mesoscale aspects will be covered in this talk. The first part of the talk relates to the percolation network structure formed in YSZ. It is well known that Y3+ ions in the YSZ structure block the movement of O2- ions. Other parts of YSZ that locally do not contain the Y3+ ions form a fast ion conducting percolation network. The network topology is determined by the underlying cation arrangement. Insights into the percolation network composition, topology and O^{2-} ion conduction rates are provided using kinetic Monte Carlo simulations. A relationship between the ionic conductivity and the topological details of the network is derived.

The second part relates to free energy of finding O^{2-}-vacancy (O^{2-}-vac) pairs in a local environment, which again is a mesoscale property of the material. Higher probability of O^{2-}-vac pairs in some regions of YSZ can result in greater number of transitions in those regions. O^{2-} ion movement in bulk YSZ is studied using multiple independent short molecular dynamics (MD) trajectories. Analysis of the MD trajectories yields free energy of O^{2-}-vac pairs in 42 different local cation (Y^{3+}/Zr^{4+}) environments, as well as effective O^{2-} hopping rates and Arrhenius parameters. Based on the free energies we conclude that it is possible that ionic movement is hindered in some environments not just because of high migration barriers or vacancy trapping as believed earlier, but also because O^{2-}-vac pairs are destabilized by these environments. Increasing the temperature and/or decreasing the dopant composition stabilizes O^{2-}-vac pairs in these environments, which in turn affects the YSZ conductivity.