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Role of site-disorder in energy materials: case of $Li_xNb_2O_5$ pseudocapacitor and β-Li_3PS_4 solid electrolyte[1] P. GANESH, ANDREW A. LUBIMTSEV, GOPI K.P. DATHAR, JONATHAN ANCHELL, PAUL R.C. KENT, ADAM J. RONDINONE, BOBBY G. SUMPTER, Center for Nanophase Materials Sciences, Oak Ridge National Laboratory — In this study, we will present computational studies to elucidate the importance of site-disorder in energy materials. We will specifically focus on two recently discovered materials: a Li-ion intercalation pseudocapacitor $Li_xNb_2O_5$ (Nature Materials, **12** 518 (2013)) and a Li-ion solid-electrolyte.(JACS, **135** 975 (2013)). A combination of theoretical methods, such as density functional theory (DFT) based cluster-expansion, basin hopping, *ab initio* molecular dynamics, and nudged-elastic-bands calculations were employed to understand the origin of intercalation pseudocapacitance in the niobate-system.(J. Materials Chem. **1**14951 (2013)). It was found that having multiple sites with similar energies for ion-adsorption, lead to a site-occupancy disorder that eventually lead to a capacitative slope in the voltage profile over the entire range of ion intercalation, as seen in experiments. A similar site-occupancy induced sublattice melting in the β-Li_3PS_4 solid-electrolyte, which when "frozen" to RT, lead to high Li-ion conductivity.(G.K.P.Dathar et al, submitted (2014)). Further, we will elucidate how to take advantage of this control over site-disorder to better engineer improved energy materials for batteries and fuel-cells.

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