Chemical Short-Range Order in Hollandite Type Phases for Nuclear Waste Form Applications

Robert J. Koch,[1] Jake Amoroso, Kyle Brinkman,[3], and Scott T. Misture[1]
[1]Kazuo Inamori School of Engineering, Alfred University, Alfred, NY 14802, [2]Savannah River National Laboratory, Aiken, SC 29801, [3]Department of Materials Science and Engineering, Clemson University, Clemson, SC 29634

The hierarchical tunnels structure and wide chemical flexibility of Hollandite-type phases offers an interesting opportunity for nuclear waste forms, particularly for ^{137}Cs entrapment, which has limited solubility in current waste form glasses. The nature and extent of structural disorder in these types of materials is still however poorly understood, particularly within the tunnel sublattice. This lack of understanding is a disadvantage for waste form applications, as it is likely that this disorder directly underpins radionuclide sorption and desorption rates, thus dictating waste form efficiency and long-term stability.

We present a study of the local tunnel structure of Ba, Cs, Mg, and Na containing Hollandite phases through combined synchrotron X-ray pair distribution function (PDF) and Rietveld analysis of synchrotron data. The impact on structural disorder of both tunnel and network chemistry are discussed, with focus on cation ordering in the tunnels and consequently nanoscale disorder that is unveiled using PDF studies.