Study of the influence of crystal structure on ion migration energy from high throughput bond valence calculations

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Progress in energy-related technologies demands new and improved materials with high ionic conductivities. Na- and Li-based compounds have high priority in this regard due to their importance for batteries. Recent developments in bond valence (BV) theory have made it possible a fast determination of ion diffusion energy barriers and ion diffusion paths [1,2]. An immediate application of these advances is the identification of potential high ionic conductors from the screening of large crystal structure databases. In this communication, we present a high-throughput exploration of the chemical space for such compounds. We show that there are significantly fewer Na-based conductors with low migration energies, as their favorable properties hinge on exceptional combinations of properties. To perform this analysis, we introduce a methodology based on bond-valence theory, graph percolation and a combined geometric and topological analysis of each structure. Specifically, we combine short-range descriptors based on a Voronoi construction, with a search for the global bottleneck in the ion transport path based on a hard-sphere model. A machine-learning analysis reveals that the ion migration energy depends mainly on the bottleneck, the coordination number of the cation and the volume fraction of the mobile species. We have implemented this workflow in the open-source Crystallographic Fortran Modules Library (CrysFML) [3].

[1] S. Adams and R. P. Rao, Struct. Bond. 158, 129-160 (2014)
[2] I. D. Brown, Chem. Rev. 109, 6858-6919 (2009)
[3] https://forge.epn-campus.eu/projects/crysfml

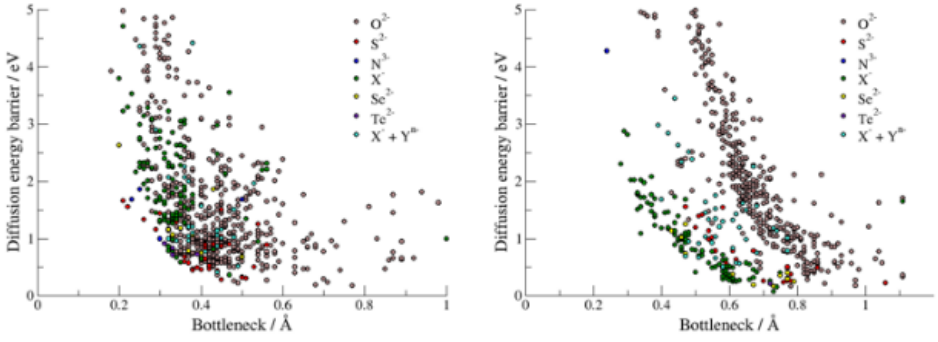

Figure 1 : High-throughput screening results for Li (left) and Na (right) compounds of the ICSD.