Crystal Site Disorder Analysis with Machine-Learned Atomic Potentials and Statistical Methods

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Crystal site disorder is a unique class of disorder in materials where particular crystallographic sites are randomly populated by two or more elemental species, including vacancies [1]. Site-disordered materials are ubiquitous in literature and experimental databases, and can occupy 30-60% the entries of certain materials classes. However, density functional theory (DFT)-based methods encounter additional challenges in performing computational analyses on site-disordered materials and they remain underrepresented in DFT-based databases such as Materials Project (MP) [2]. It is thus worthwhile to explore computationally economical options of analyzing site-disordered materials, as by doing so, we can accelerate the processes of (1) plugging the gaps of knowledge in DFT-based databases and (2) corroborating experimental and computational data involving inherently site-disordered materials and or fine stoichiometric tuning. Current methods of site disorder modelling employ the use of supercells and the generation and enumeration of virtual cell representations [1,3]. In this work, we explore the use of CHGNET machine-learned potentials [4] to expedite the high-throughput total energy evaluation of a set of virtual crystal unit cells. We propose that the massive configurational space of virtual cells, which can go up to billions of configurations, can be sampled with a reduced set of a few hundred virtual cells. Thirdly, we find that for site-disordered materials of certain classes, total energy calculations can be substituted with the evaluation of local coordination behavior according to Pauling's rules of ionic crystals [5], a much simpler process (Figure 1). With these, we can predict the physical and functional qualities of site-disordered materials and readily fill in a significant conceptual gap in DFT-based materials databases at a reduced computational cost. We explore further possibilities in tackling correlated disorder and machine learning prediction of crystal structures.

References

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Figures

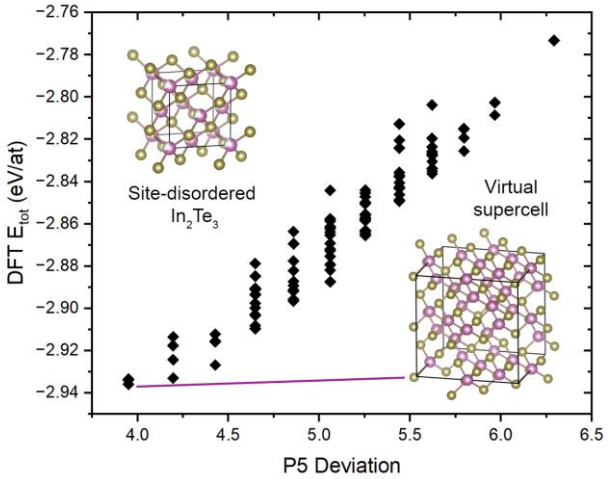

Figure 1. Correlation between DFT total energy and the deviation parameter of Pauling's 5th rule (P5 deviation) in site-disordered sphalerite indium telluride (In_2Te_3).