

Getting Solid Predictions: Solid-State Thermodynamics from Quantum Chemistry

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The absence of reliable experimental data for the thermodynamic properties of many solids is a major challenge in fields ranging from electrolyte solutions to materials science. Being able to quickly obtain accurate and reliable values for these properties is important in several chemical contexts including electronic waste recycling, thermal energy storage, carbon capture and storage and pharmaceutical production. In recent decades, electronic structure models originating in quantum chemistry (QC) have paved the way for highly predictive simulations of solid-state thermodynamics [1]. Despite this, the broad application of QC-based models is hindered by their complexity and high computational cost, while novel atomistic models still lack the desired chemical accuracy [2].

In our work, we seek to investigate and develop frameworks for fast and accurate prediction of the thermodynamic properties of solids. Firstly, we show that the quantitative performance of the harmonic approximation for the prediction of the temperature-dependence of the thermodynamic properties of solids is excellent in many cases; thus, paving the way for less complex and considerably more efficient predictions of the thermodynamic properties using QC-based models [3]. Secondly, aided by QC calculations, we have developed a framework based on reaction networks which accurately predicts the experimental enthalpy of formation of solids (Fig. 1). Upcoming work includes application of predicted thermodynamic properties to investigation of the solution and thermodynamic properties of thermal energy storage materials.

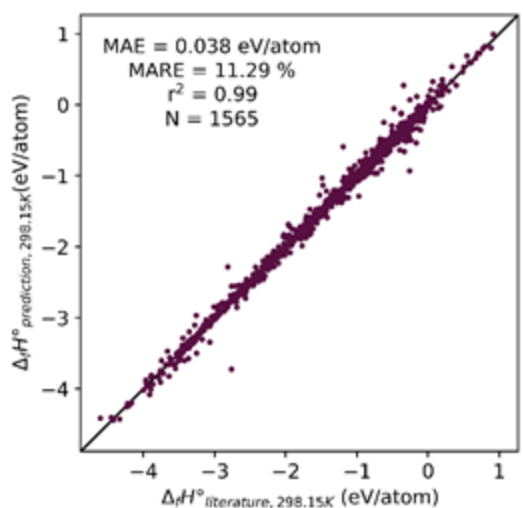


Figure 1: Parity plot of the enthalpy of formation at 298.15 K predicted using reaction networks, for 1565 compounds, against literature values. Also included are the mean absolute error (MAE), mean absolute relative error (MARE) and the coefficient of determination (r^2).

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