Chemical Engineering Dissertation Defense

Unlocking Efficient Thermochemical Energy With Computational Materials Design Through The Compound Energy Formalism

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Abstract

Cyclical chemical looping involves the thermal reduction of metal oxide to release O2 at high temperatures, followed by its oxidation using O-containing molecules like O2, H2O, or CO2. This process is a promising method for converting solar heat to fuels, oxygen separation, and thermal energy storage. The efficiency and economic viability of this process hinge on the thermodynamics of metal oxide reduction. This dissertation presents innovative methods to enhance the performance of these processes, with a specific focus on solar thermochemical water splitting (STCH), by advancing thermodynamic characterization and screening of potential metal oxides. These advancements aim to identify more efficient solar-to-fuel materials, thereby reducing STCH H2 energy costs.

The study utilizes advanced quantum mechanical simulations and density functional theory (DFT) to gain insights into material and chemical behavior in thermochemical energy cycles. A novel CALPHAD approach, the CrossFit Compound Energy Formalism (CEF), integrates theoretical (DFT) and experimental (thermogravimetric) data to develop thermodynamic models for desired materials. Comparisons with the traditional van ‘t Hoff thermodynamic extraction technique reveal that the cross-fit CEF method significantly outperforms conventional methods. For instance, the CEF method was employed to extract thermodynamic data for the CaFexMn1-xO3 materials family to identify optimal heat storage materials.

To expedite data collection and reduce the required data points, an active data selection process is implemented. Bayesian Inference, when combined with the CrossFit CEF method, assesses the likelihood that the current model accurately describes the data, providing confidence intervals for the model. This approach reduces the amount of data needed for an accurate thermodynamic model by 50%.

Furthermore, the CrossFit CEF and Bayesian methods are integrated into a system-level STCH model to optimize materials for specific plant operating conditions and accelerate material design processes. Overall, this dissertation introduces methods that yield more accurate thermodynamic models with reduced data requirements compared to the standard van ‘t Hoff analysis. The time saved in data collection enables screening of more materials, expediting material identification and optimization. The materials identified through these techniques are expected to enhance chemical looping cycles, leading to increased H2 production efficiency and reduced costs.

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