MATERIALS ENGINEERING SEMINAR

"Materials Design Using First Principles Calculations: Investigating Halide Perovskites and Transition Metal Electrocatalysts"

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Purdue MSE Ph.D. Final Exam

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ABSTRACT

With increasing global renewable energy demands, there is a need for new materials with improved performance, lower cost, and less toxicity. One such application is photovoltaics, where halide perovskites (HaPs) represent the fastest growing market of absorbers owing to their impressive optoelectronic properties and excellent tunability from composition engineering and structural manipulation. However, the practically infinite composition-structure space of HaPs when considering cation and/or anion site mixing, octahedral distortion and rotation, and other forms of polymorphism, raise considerable challenges when comprehensively exploring their stability and optoelectronic properties. First principles calculations are powerful tools that can investigate large numbers of compounds and structures in a high-throughput fashion. In my thesis, I performed high-throughput density functional theory (DFT) computations to generate a HaP dataset within a wide chemical space covering ~500 unique chemical compositions in the (pseudo-)cubic phase, across a 14-dimensional ionic space. This work explored both pure and alloyed compositions, with the latter simulated using the special quasi-random structures approach. Many critical properties were computed using the semi-local GGA-PBE and hybrid non-local HSE06 functionals, including decomposition and mixing energies, electronic band gap, and spectroscopic limited maximum efficiency (SLME), which is a theoretical surrogate for the likely absorption efficiency of the compound when used in a single-junction solar cell. Property screening over this dataset yielded 32 stable perovskite candidates with attractive optoelectronic properties.

Polymorphism in HaPs is investigated by simulating larger supercell alloys with different ionic ordering, generating compounds with random octahedral distortions and rotations, and optimizing various compositions in non-cubic phases such as tetragonal and orthorhombic. Linear correlation analysis is performed to gain a critical understanding of how properties are influenced by specific cations and anions, their mixing fractions, the perovskite phase, ionic segregation, and amount of strain or distortion in the lattice. Finally, trends, design rules, and predictive insights achieved from the DFT datasets are applied over a much larger set of thousands of hypothetical compounds, resulting in identification of more promising materials, and understanding of the most important A-B-X combinations that yield multiple desired objectives.

Furthermore, a similar DFT workflow is applied for designing transition metal electrocatalysts. DFT simulations are performed to model Hydrogen adsorption, OH adsorption, and the water splitting reaction on Ni_3N/Ni and Co_2N/Co hybrid structures, to explore their likelihood in being used for Hydrogen Evolution Reaction (HER). The results reveal the excellent catalytic performance of transition metal and transition metal nitride hybrid structures.

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Place: ARMS 1021 or via link:
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