An Effective Machine Learning Frame for Materials Discovery Structured by a Chemical Concept

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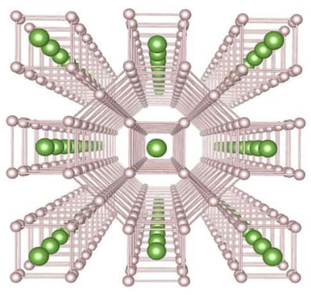
Abstract

Despite intensive work on hydrogen-rich binaries, large regions of composition space remain unexplored, especially systems with non-integer non-metal-to-metal ratios. We combine the chemical template concept with machine learning to build a targeted structure-discovery workflow that efficiently predicts new stable compounds. Applying this framework, we identify 13 new structural prototypes and 31 stable metal superhydrides, a 23% increase over previous datasets. Many of these phases feature 3D hydrogen clathrate cages and primitive cells with more than 50 atoms, a regime that is typically challenging to explore with conventional methods. Notably, 19 of the newly discovered superhydrides have predicted superconducting transition temperatures T_c above 100 K, underscoring the promise of chemical-template-guided machine learning for uncovering higher T_c materials in complex hydrogen-rich systems.

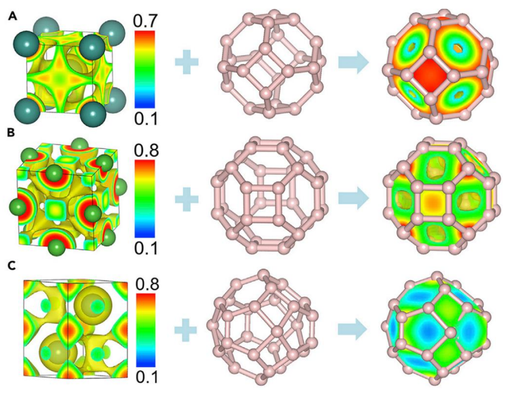
Background

High Pressure Chemistry

- High-pressure chemistry unveils unique material structures leading to discoveries like phase transitions and unusual stoichiometries
- It can induce superconductivity, transform the typical chemistry of elements, and stabilize compounds with distinctive electronic, mechanical, and optical characteristics

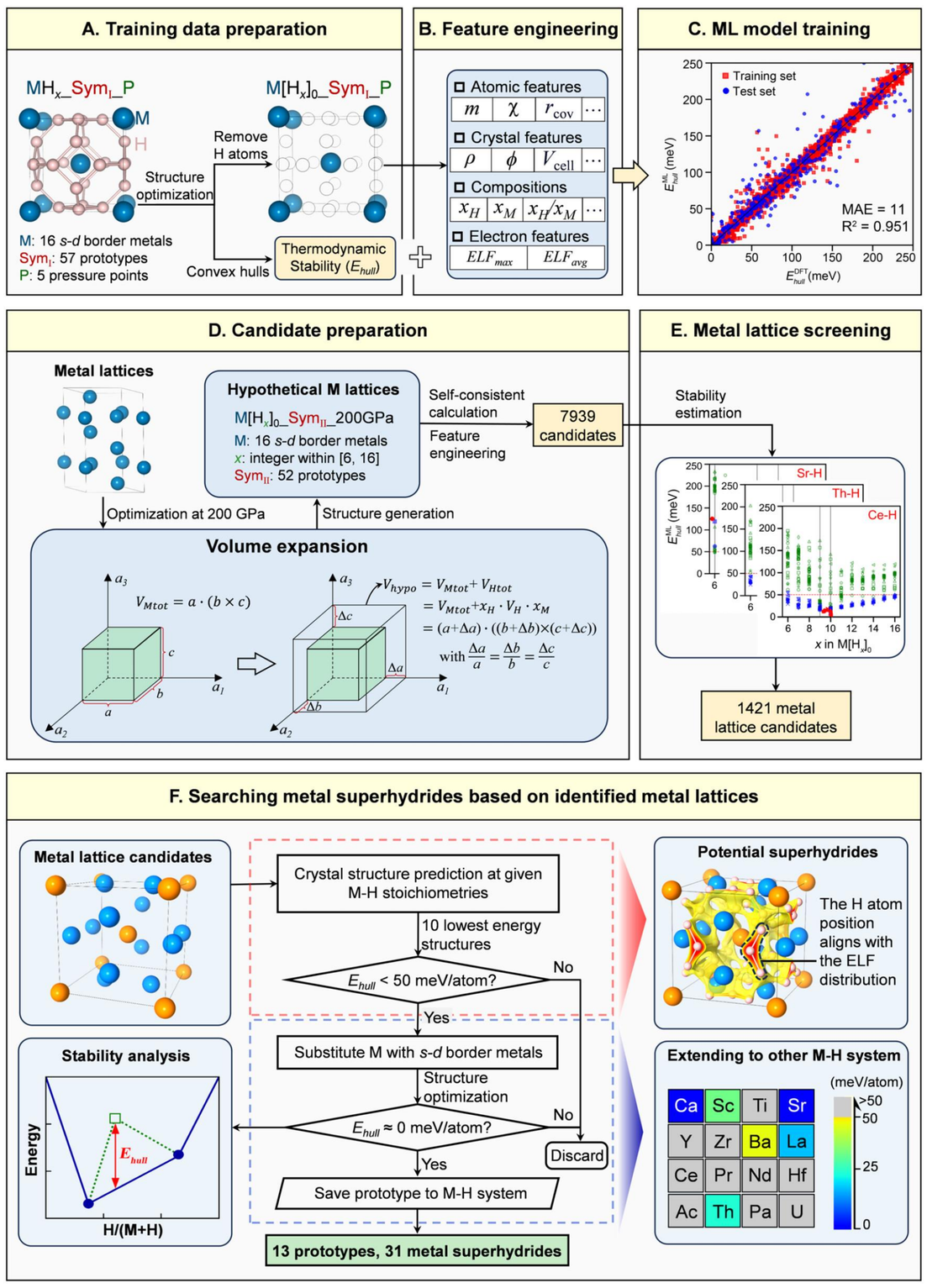


Chemical Template Effect



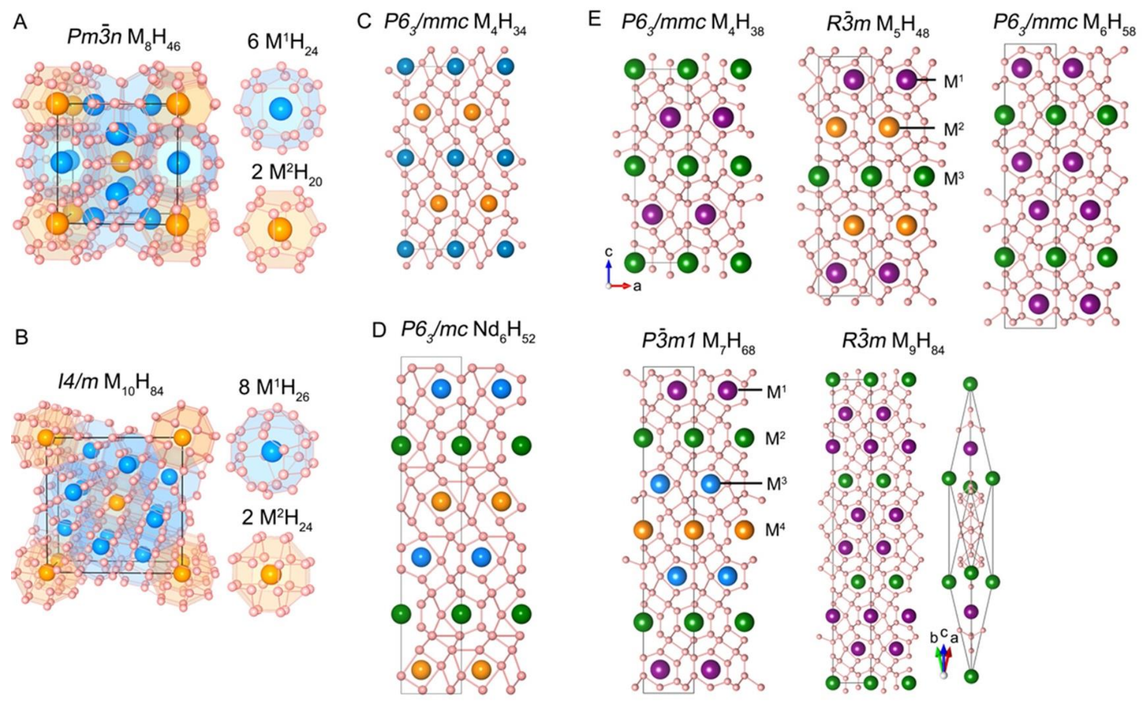
- Electrons occupying the quantum orbitals located at the interstitials of metal sublattices (quasi-atoms) form a template that assists the assembly of H lattices
- Utilizing a template can significantly improve the efficiency of searching for new complex compounds.

Methods and Workflow

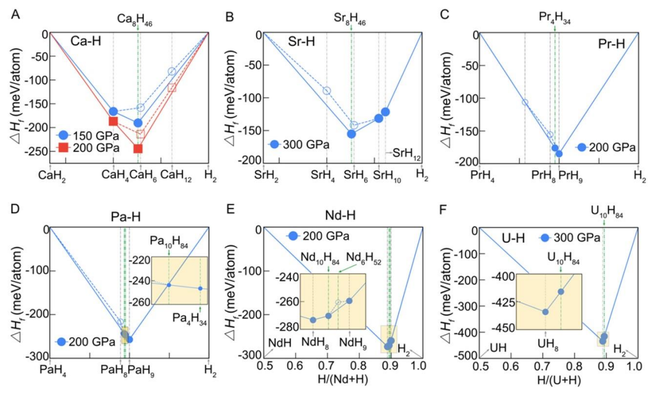


Results

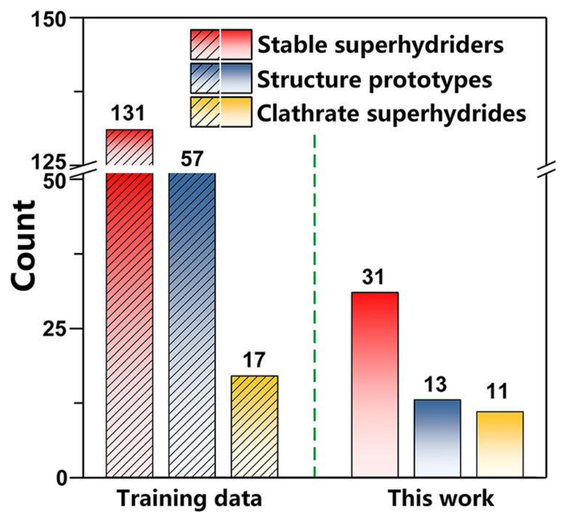
Crystal Structures of Newly Identified Metal Superhydrides



- Template guided search uncovers a family of previously unknown MH$_x$ structures, including M_8H_{46}, $M_{10}H_{84}$, $MH_{9+\delta}$, MH_{10}, and M_4H_{52} with distinct hydrogen cage topologies and multiple metal sublattices
- These structures show that the chemical template workflow can discover very complex superhydrides with non-integer ratios and complicated structures that are difficult to explore with conventional unconstrained crystal structure searches



- Convex hull calculations at 150 to 300 GPa show that the newly discovered MH$_x$ phases with non-integer ratios lie on or very near the ground state hull across several metal hydride systems
- The updated hulls are reshaped by the new phases and reveal previously missed stability windows at non-integer hydrogen ratios
- Template guided search adds 31 new stable superhydrides, 13 new structure prototypes, and 11 new clathrate superhydrides on top of the original training set, substantially enlarging the design space.
- Demonstrates that the new workflow based on chemical templates can discover complex compounds with modest training data.



Conclusion

- Template effect is a fundamental chemical mechanism that stabilizes many compounds with high non-metal content.
- Implementing a machine learning workflow based on chemical templates enables batch discovery of new compounds with complex stoichiometry and structure beyond the training set.
- Many of the predicted superhydrides lie on or very near the convex hull, and 19 have estimated superconducting transition temperatures above 100 K, so the workflow prioritizes experimentally relevant high T_c candidates.
- This concept driven ML strategy can be extended to ternary superhydrides and other materials where localized electrons act as templates, offering a scalable route to discover complex quantum materials.

Acknowledgments

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