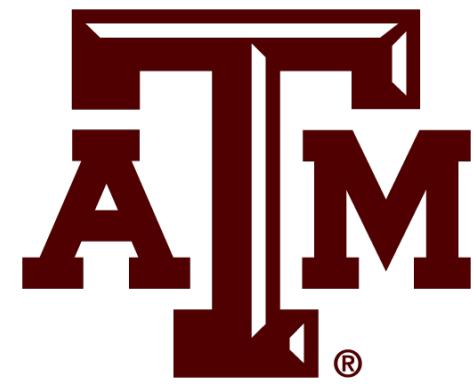


Accelerating Design of Redox-Active Polymers for Organic Radical Batteries by Machine-Learning-Assisted Virtual Screening and Insights from Molecular Simulations



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OVERVIEW

Nonconjugated redox-active polymers offer environment-friendly alternatives as opposed to metal-based inorganic batteries.

- Screening potential candidates of lead low-potential redox-active molecules with fast charge transfer kinetics.
- Atomistic MD simulations provide insights of how the choice of electrolytes limits the electron hopping between redox-active groups.
- ChiRo as a robust reorganization energy predictor.

METHODS

Virtual Molecular Screening

- Two stages
- Screening the library to get top-10% lowest reduction potential molecules using Gaussian Process Regression model
- Perform more expensive electron coupling calculations on selected top candidates.
- Identify low-potential redox-active molecules with high Boltzmann-averaged electronic coupling.

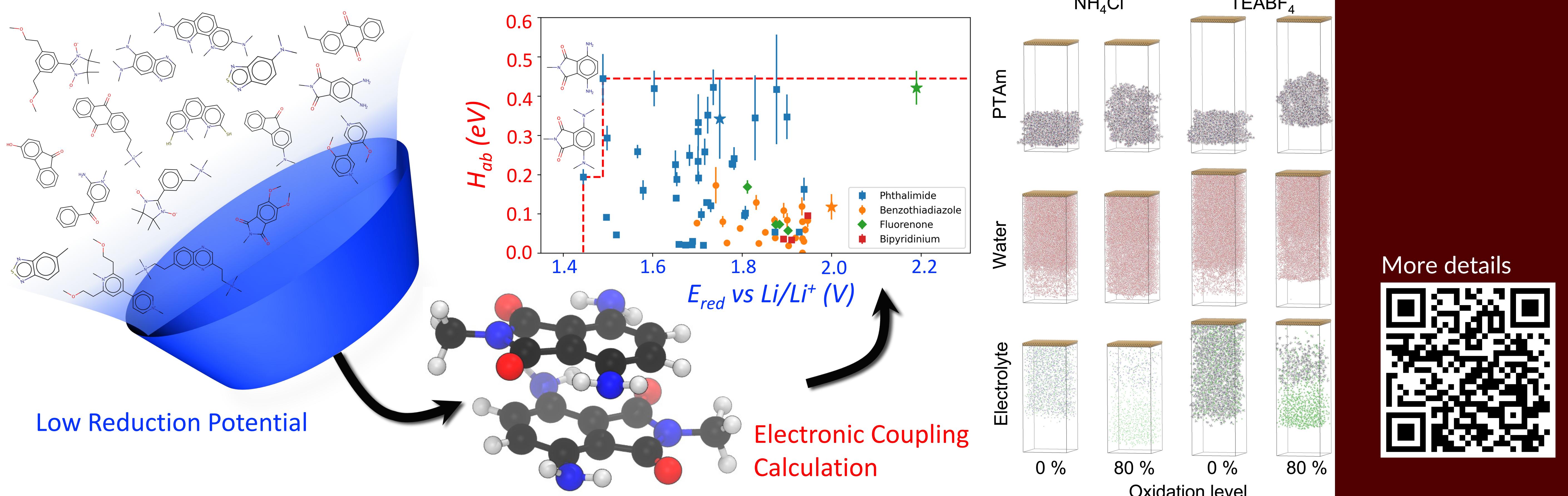
MD Simulations

- Anneal a PTAm film on top of gold layers starting from a box of 50 PTAm 20-mers using Langevin dynamics.
- Add TIP4P/Ew water and electrolytes (NH_4Cl or TEABF_4) on top of the PTAm film.
- Oxidize TEMPO groups from bottom to top and analyze the swelling behavior.

Reorganization Energy Prediction

- Compare ChiRo and SchNet architectures

- Found lead low-potential candidates for organic radical polymer batteries with ML-assisted virtual screening.
- MD simulation reveals that chlorides make electron transfer between redox-active groups harder.
- ChiRo is more robust for reorganization energy predictions



RESULTS and DISCUSSION

Virtual Molecular Screening

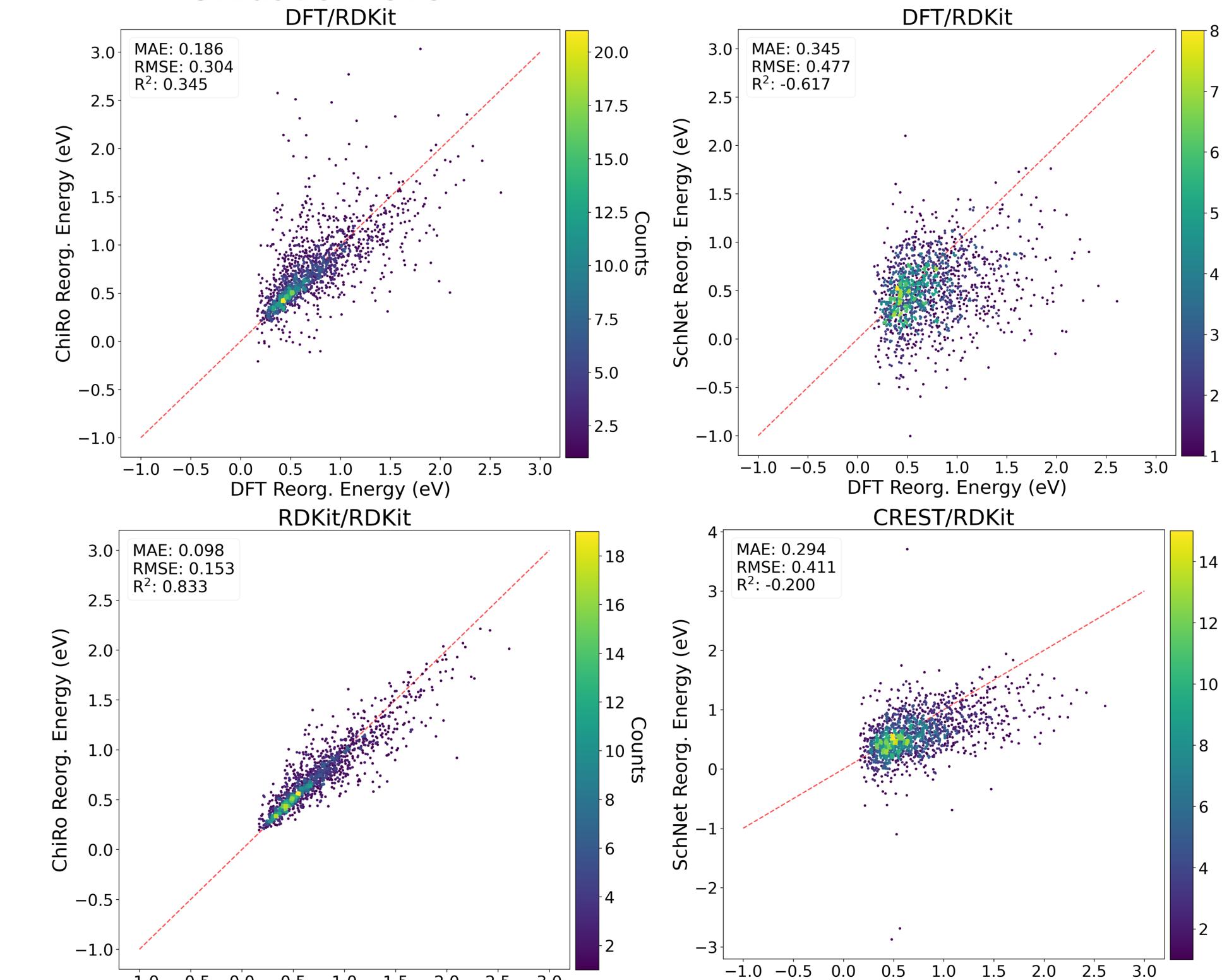
- Using only molecular descriptors from the core of each molecule prevents GP model from overfitting due to different environmental effects.
- Two Pareto-optimal molecules were identified, and both have phthalimide as their cores.
- Hydrogen bonding and favored stacking between dimers dominate the Boltzmann-averaged electronic couplings.

MD Simulations

- Interchain distances between TEMPO groups become larger after PTAm films were oxidized, while the intrachain distances stay similar.
- PTAm film swells more with NH_4Cl , which makes the electron transfer between TEMPO groups more difficult.

Reorganization energy prediction part:

- ChiRo generalizes well on low-cost RDKit geometries testing after trained with multiple RDKit-embedded conformers.



Reference:

Li, C.-H.; Tabor, D. P. *J. Mater. Chem. A*, **2022**, *10*, 8273-8282

Ma, T.; Li, C.-H.; Tabor, D. P.; Lutkenhaus, J. L., *in preparation*