[Physical Sciences] Learning Catalysts, One Piece at a Time

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A CS229 Project at Stanford University

Summary

We extend previous research on screening for new catalyst materials for the CO_2 reduction reaction. We introduce a new set of features, describing the valence electron environment of surface layers neighboring an adsorbate. We have found that this strategy allows for fast prediction of the CO binding energy of various adsorbate configurations on a variety of materials with good accuracy using kernel-based models and moderate accuracy with other methods.

Background

- The CO_2 reduction reaction has garnered widespread attention in recent decades due to its potential to both produce renewable fuels and mitigate increasing CO_2 concentrations in the atmosphere.
- Polycrystalline copper (Cu) is the sole transition metal known to produce hydrocarbons but at a large energy cost[2].
- In order to produce useful fuels from CO_2 reduction, the CO binding energy has to lie in a narrow sweetspot near -0.3eV[3]. Small deviations from the optimal binding energy significantly decrease the predicted activity of the catalyst.
- ML can be used to quickly estimate CO binding energy in order to screen various materials and create a list of materials with the optimal CO binding energy.

Data

- Data was collected from DFT calculations with the Quantum Espresso code in the Atomic Simulation Environment (ASE).
- Our current dataset has almost 1000 crystal motif examples spanning more than 80 materials systems.
- Each example is labeled by ground truth; the output comes directly from DFT calculations containing all atomic structure information.

Features

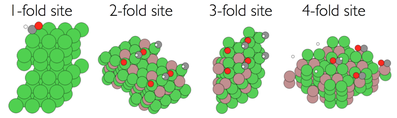

Figure 1: Adsorption Sites on Nickel Systems

- Predicting CO binding energy on a new motif by searching for 'similar' motifs found in the training set.
- Features describe geometric and electronic nature of nearest neighbor atomic network.
- As quantum mechanical features were reported to perform better than basic chemistry descriptors[4], we added valence electron environment as a feature from DFT calculations on bulk material structures.
- Using theoretical priors, we have manually reduced our feature space down to a total of 56 features, from 180 in our milestone.

4 NN Atoms to Adsorbate	4 2nd-NN Atoms
Element Group	Element Group
Bond Angle and Distance	–
Δ Coordination	Δ Coordination
Val. Electron Environment	Val. Electron Environment

Models

- Gaussian process (GP) regression with an RBF kernel with a regularization factor of 0.1.
- A fully connected Neural Network with two hidden layers of 8 and 2 nodes[1], optimized Limited Memory-BFGS on a regularized squared error loss. The hidden layers use a tanh activation function while the output layer is linear.
- Support Vector Regression (SVR) with a Laplacian kernel[5] and Kernel Ridge Regression (KRR) was used to compare with GP as both are also models that regress based on motif similarity.
- Hyperparameters for models were optimized via cross-validated grid searches: kernel-based models with shuffle-split, neural networks with K-fold.

Results

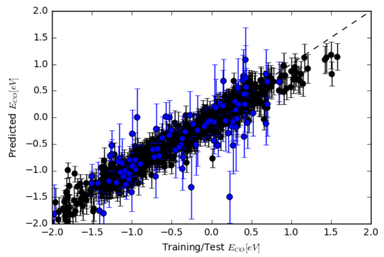

Figure 2: GP Parity Plot (859 Training:100 Test)

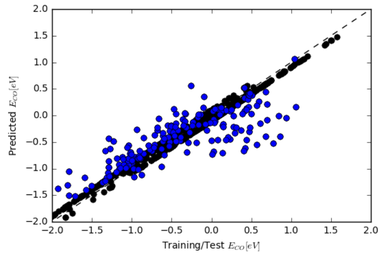

Figure 3: SVR Parity Plot (859 Training:100 Test)

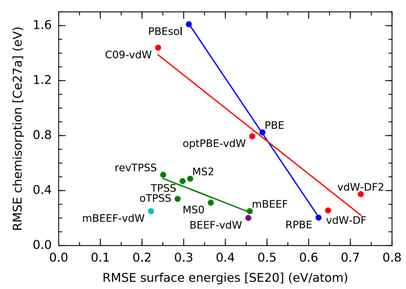

Figure 4: DFT error by functional

Model	Train RMSE [eV]	Test RMSE [eV]
Gaussian Proc.	0.178	0.432
Supp. Vec. Regr	0.094	0.495
Kernel Ridge Regr.	0.084	0.496
Nearest N. Regr	0.394	0.690
Linear Regr.	0.607	0.667
Neural Networks	0.791*	0.822*

Table 1: Errors by Model. *Rudimentary NN Model

Discussion

- For kernel-based models, the predicted CO binding energy is generally in agreement with the DFT-calculated binding energy.
- Automatic feature selection with Recursive Feature Elimination ranked the valence electron environment of the 2nd layer as the most helpful feature.
- The neural networks were difficult to train, primarily because of limited dataset size and shortcomings of fully connected architecture.

Future Directions

- Refine the feature set to succinctly describe the bonding nature of atoms, or to help other models gain better accuracy.
- Generate larger datasets and develop novel featurizations to apply deep learning architectures such as conv. nets.
- Scale this work to encompass trimetallic alloys, highly uncoordinated surfaces, and strained motifs.

References

[1] Ma et al. *J. Phys Chem. Lett.* 2015.

[2] Hori et al.*Modern Aspects of Electrochem.* 2008.

[3] Peterson et al. *Energy Environ. Sci.* 2010.

[4] Ward et al. *Computational Materials.* 2016.

[5] Rupp et al. *Int. J Quantum Chem.* 2015.

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