Machine-Learning Discovery of Highly oxidized IrOx Phases

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Abstract

Herein, we will fill in the abstract and it will be fairly concise but descriptive as well. Talk about ML, materials screening, OER, and Ir systems.

Introduction

Iterative Active Machine Learning and unique prototype identification to discover stable new materials and catalysts. Motivation for IrOx, low representation, longstanding controversy

over oxidation states and topology, and demonstrates promise for OER and Li ion batteries. Reported +6 oxidation state phases are achievable leading to high degree of structural variability, which is the highest for transition metals. High oxidation states (low pH high anodic voltage, harsh oxidizing conditions) unexplored, need very specific structures with precise oxygen connectivity (aka high pressure SrIrO3) that can exist. Machine learning is the efficient way to explore this "exploring antarctica for life" sparse space. What we show here...

Chris

Crystallographic Discovery and Machine Learning. Current state of databases (OQMD, MP, CatHub, alfowlib). What parts of the database are missing (e.g. IrO3). Ankit - Condensed version of Ankit paper Prototyping databases to identify knowledge gaps. Ankit Deriving features from structures to describe heats of formation. What has been done (simple models) more recently using DFT+Machine Learning.

Chris

Oxides for batteries/fuel cells, Iridium Oxide, OER, Lithiated IrO3 Highly oxidized phases of oxides for fuel cell and energy storage applications.

Michael

Results and discussion

I. IrO2

For Structure Rendering Use: Michael To Send example vesta files, font Avenir

Figures: Summary figure of found structures, P1 AB2 Structures Ankit - There are XYZ unique AB2 structures (or multiples, e.g. A2B4) - Of those we found 697 unique AB2 prototypes (unique SG/Wyckoff combination) in OQMD/MP - To generate our test set we substituted Ir for A and O for B, then isotropically expanded cell volume to constrain a minimum Ir-O distance of XYZ -Next translated each of the 697 structures to be described

by 271 features (invariant to isotropic expansion/compression), then reduced to 30 using PCA, described in methods XYZ -To generate initial training data use existing DFT. Not enough on IrO2, so used OQMD to generate initial training data from nearest structures in phase space, described in Methods XYZ. Training set of 30 structures in SI XYZ. P2 Iterative Training of Gaussian Process - Trained Gaussian Process, rational quadratic kernel, variable lengthscales. CV error of XYZ ev/atom, initial predictions in figure XYZ. - Selected 10 structures with lowest prediction-uncertainty for DFT. Structures were volume optimized, then fully relaxed, described in methods XYZ. -Model retrained with the 10 DFT computed structures ONLY, 271 features-¿110 features applicable to IrO2-¿20 principle components for 99.9 percent variance. CV error... -Repeat until XYZ, final predictions shown in Fig XYZ

P3 IrO2 Structure Analysis -Describe convex hull plot (energy vs. Ir-O distance), computed amorphous phase to define synthesizability -While only 2 IrO2 in MP/OQMD, we can compare our structures to other computed IrO2 not in open databases.

This is a citation example.[1] Without it I think stuff breaks

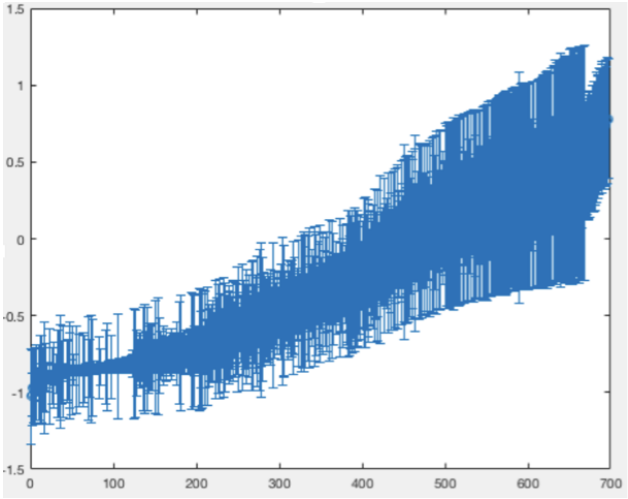

Figure 1: TMP.

II. IrO3

P1 AB3 Structures and Training -XYZ unique AB3 Structures, 259 unique prototypes.

Substitute Ir and O, expand to minimum Ir-O distance ¿ XYZ -followed same procedure as in 3.1, Training Set of 35 structures, 8 of which are IrO3 -Describe initial training and training after first 10 DFT structures

P2 Analysis of IrO3 Structures -Describe convex hull, classes of structures (alpha-AlF3 like, rutile like, and layered, should be segregated in hull plot) -briefly describe structures within each class, cite in literature where appropriate

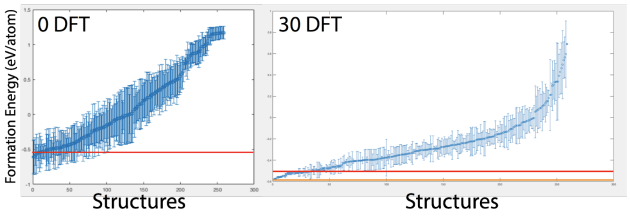

Figure 2: TMP.

III. TEMP TEMP

IV. Electrochemical OER Application

a. Bulk Pourbaix (Michal and Raul)

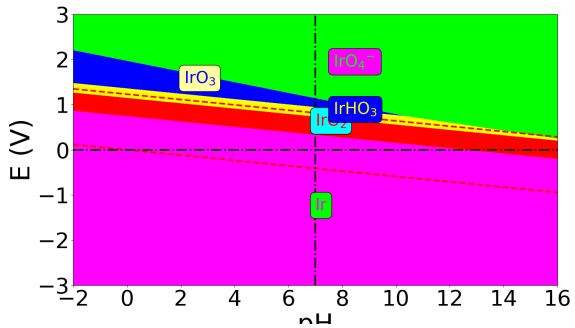

Figure 3: Electrochemical bulk phase stability diagram (Pourbaix) of the Ir-O-H chemical space.

b. OER Activities and Surfaces

Procedure: - For the top/most stable bulk structures the following procedure was carried out

* Stable stoicheometric terminations were cut from the bulk Stable termination planes were guesstimated via intution, and the x-ray diffraction pattern tool from Vesta

* Electrochemical surface coverage was elucidated via a surface Pourbiax analysis Need to know the coverage of surface under operating conditions (¿1.23 VRHE)

* Thermodynamic/limiting potential analysis of the OER mechanistic pathway Volcano plot, limiting potentials, etc.

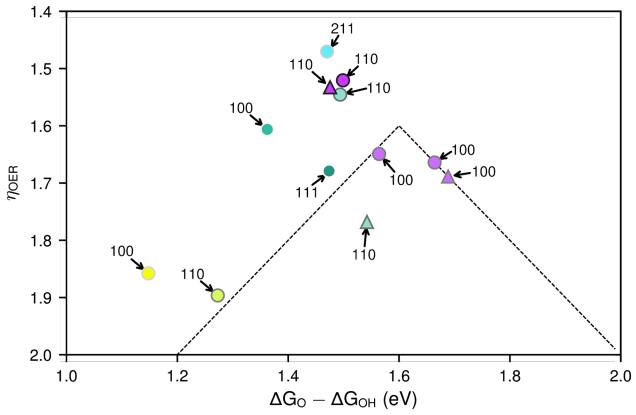

Figure 4: OER activity volcano for most relevant facets of IrO2 (yellow), IrO3 (blue), and IrO3-rutile-like (purple). Circles designate oxygen covered surfaces while triangles designate hydroxyl (*OH) terminated surfaces (relevant surface terminations were found via surface Pourbaix analyses. Surface energies at standard conditions (pH and V = 0) are reflected in the border color for each data point, where black indicates a low energy surface termination and white indicates more unstable surfaces. The color range goes form x to y. Maybe construct a color bar to include here?

To determine the most likely experimentally abundant surface facets and surface coverages, a surface energy Pourbaix diagram was constructuted.

c. OER Intermediate Scaling

Write stuff

Conclusion

And in conclusion we presented work here...

Computational Methods

CM - 1

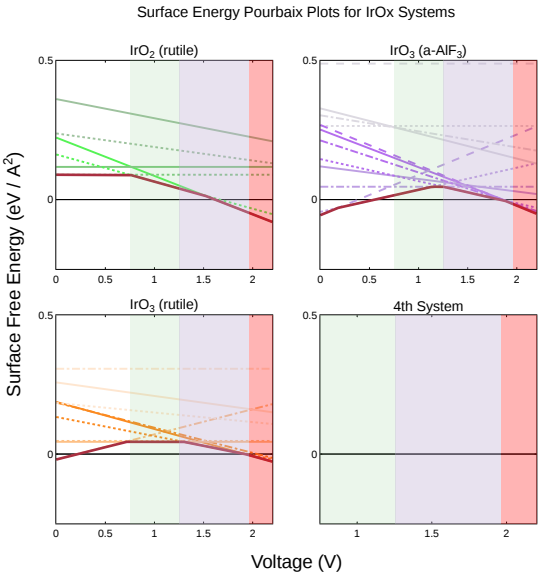

Figure 5: Surface energy of different facets of IrO2, IrO3-rutile, and aAlF3-IrO3 crystal structures.

Paragraph goes here.

Acknowledgement

Organizations to acknowledge TRI SUNCAT Stanford NERSC etc.

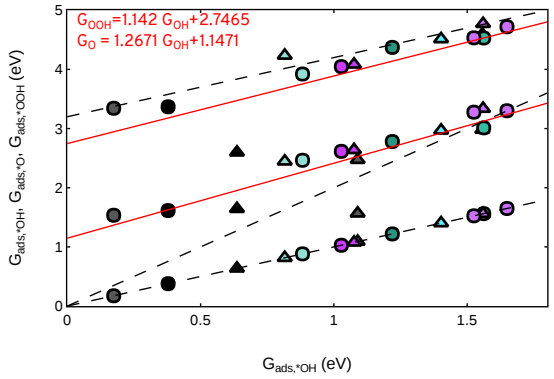

Figure 6: Electrochemical bulk phase stability diagram (Pourbaix) of the Ir-O-H chemical space.

Supporting Information Available

I. SI Section 1

First section of SI is here

II. SI section 2

Second section of SI is here

References

(1) Nørskov, J. K.; Rossmeisl, J.; Logadottir, A.; Lindqvist, L.; Kitchin, J. R.; Bligaard, T.; Jónsson, H. Origin of the overpotential for oxygen reduction at a fuel-cell cathode. *Journal of Physical Chemistry B* **2004**, *108*, 17886–17892.

Graphical TOC Entry