

Machine Learning Generalized Geometric Descriptors for Oxygen Reduction Activity on Transition Metal Sulfides

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Sabatier-type activity volcano plots have been used to determine optimal adsorption characteristics for maximizing catalytic activity. In this process of catalyst design, an important step involves designing catalysts involves identifying the active sites for optimal adsorption strength and engineering catalyst materials with target active sites since it has been shown that the variation in binding energies across heterogeneous surface sites is strongly dependent on the local environment¹. Therefore, catalyst discovery can be accelerated through structure-energy-activity relationships, which allow us to formulate the active site design as merely finding the optimal local coordination environment that maximizes activity. Recently, structure-activity relationships have been developed for metals¹², and we extend this through a **machine learning based approach** to obtain a **structure-energy-activity relationship for transition metal sulfides**. Transition metal sulfides represent an attractive class for ORR³ and their rich phase diagram allows tunability of activity. We employ density functional theory (DFT) to study ORR activity on various transition metal sulfides including scandium, titanium, vanadium, chromium, ruthenium, nickel, cobalt, molybdenum, manganese, copper and iron sulfides.

We illustrate the robustness of the developed geometric descriptor for an example class of nickel sulfides⁴. As observed on metals and metal oxides, we show that there exists scaling between oxygen intermediates, that leads to a volcano relationship for the limiting potential in the OH* adsorption energy. We identify a simplest geometric descriptor using a least-squares approach to obtain the structure-energy descriptor: $G_{OH} = 0.29(CN_S + 0.06CN_{Ni})$, which has excellent predictability (RMSE = 0.16 eV). Therefore, to a first approximation, a simple counting of neighboring atoms around the surface site can be used to screen catalysts. Our analysis suggests that having 3 nearest-shell sulfur atoms leads to high activity for Ni-based sulfides.

We will discuss a generalization of this structure-energy descriptors for transition metal sulfides and the trends in the coefficients will be discussed to formulate a generalized structure-activity descriptor for the transition-metal sulfides class. These coefficients are expected to strongly depend on metal-metal and sulfur-metal bonding characteristics. Identification of structure-activity relationships for classes of compounds allows for rapid identification of candidates and robust activity quantification. This approach could allow for rapid screening within different material classes for a variety of reactions.

References:

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