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Towards first-principles based prediction of highly accurate electrochemical Pourbaix diagrams ZHENHUA ZENG, Purdue University, MARIA CHAN, Argonne National Laboratory, JEFF GREELEY, Purdue University — Electrochemical Pourbaix diagrams lie at the heart of aqueous electrochemical processes and are central to the identification of stable phases of metals for processes ranging from electrocatalysis to corrosion. Even though standard DFT calculations are potentially powerful tools for the prediction of such Pourbaix diagrams, inherent errors in the description of strongly-correlated transition metal (hydr)oxides, together with neglect of weak van der Waals (vdW) interactions, has limited the reliability of the predictions for even the simplest bulk systems; corresponding predictions for more complex alloy or surface structures are even more challenging . Through introduction of a Hubbard U correction, employment of a state-of-the-art van der Waals functional, and use of pure water as a reference state for the calculations, these errors are systematically corrected. The strong performance is illustrated on a series of bulk transition metal (Mn, Fe, Co and Ni) hydroxide, oxyhydroxide, binary and ternary oxides where the corresponding thermodynamics of oxidation and reduction can be accurately described with standard errors of less than 0.04 eV in comparison with experiment.

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