Role of Density Functionals for Adsorption Energy Prediction for Fe SACs

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The use of accurate computational method for modeling is essential to obtain a reliable energy and electronic property. The develop of density functionals is the most affordable and relatively accurate way to reveal the electronic state of the system comparing to post-HF method. The validation of density functionals for adsorption energy and electronic properties has been investigated for periodic system by many research group respect to experimental results. However, for finite system, especially the single atom catalyst in a finite system with a diverse coordination environment, the role of density functionals is not clear. In this work, we systematically study the performance of 14 density functionals from GGA to range-separated hybrid meta GGA to reveal their capability for predicting accurate adsorption energy of H* and OH* respect to DLPNO-CCSD(T) result. Since Fe is a ferromagnetic element, we carefully studied the capability of density functionals for group spin reproducing. We found, comparing with DLPNO-CCSD(T), most of density functionals could reproduce the group spin well for close shell system but give big uncertainty for open shell system. The best correlation between functionals with DLPNO-CCSD(T) is PBE1PBE. By varying the coordination atom with C, N, O, S and P, we found the configure ration contains more S and P will give stronger binding.