Efficient Machine Learning barrier height and reaction enthalpy corrections for low- and mid-level quantum mechanical calculations

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Automated methods for the prediction of reaction mechanisms are a key tool for the study of complex chemical reactions. Many of these methodologies involve a first step based on extensive exploration of the configurational space using a low-level electronic structure method, followed by a further refinement by high-level calculations, which results in a considerable computational effort when a complex reaction involves a very large number of elementary steps. Therefore, the availability of efficient, low- or mid-cost yet accurate approaches for thorough exploration of complex reactive potential energy surfaces is of great interest. In this work, efficient machine learning (ML) corrections were developed to improve the accuracy of a few low- and mid-level electronic structure methods for the calculation of barrier heights and reaction enthalpies. We followed the Δ-ML strategy, training the model on the energies of stationary points in the RDB7 dataset.[a] The baseline methods utilized in this work were PM6, HF/3-21G, and r^{2}SCAN-3c. The results show that the r^{2}SCAN-3c method, together with our corrections, provides very good accuracy, similar to or even surpassing that of other ML potentials published recently. The use of energies only, without including gradients or hessians, makes it easier to extend the approach to large chemical spaces.

References:

[a] Spiekermann, K.; Pattanaik, L.; Green, W.H. High accuracy barrier heights, enthalpies, and rate coefficients for chemical reactions. Sci. Data 2022, 9 (417), 1-12. https://doi.org/10.1038/s41597-022-01529-6.