Importance of systematic geometric searching for computational catalysis: Case studies in heterogenous catalysis

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Theoretical calculations have been widely applied to interpret the experimental phenomena, ranging from the structure determination to the promoting of catalytic activity for a given catalyst. Catalysts can accelerate chemical rection by reducing activation barrier or changing reaction mechanism. Therefore, catalytic activity of catalysts can be discussed by calculating transition states and reaction pathways. I was working on the investigation of structure-activity relationships in catalysis based on the global reaction route mapping (GRRM). By screening multiple possible active species and active sites of metal clusters, it was found that the most stable active species and intermediate structures do not always lead to the lowest transition state or the pathway with lowest barrier.[1, 2] The same conclusion was also proved by a collaboration work with experiment.[3] Moreover, the importance of systematic searching was shown for the solid state catalyst.[4,5] Therefore, a systematic searching/screening is essential to reveal the origin of catalysts.

The discrepancies in the stability of the molecular structures of intermediates and transition states can occur in organometallic catalysts as well. After joining WPI-ICReDD, I started to work on systematic searching for pathways of the chemical reaction catalysed by organic molecule and some initial results also will be shown in the presentation.

References

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