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Artificial intelligence in chemical kinetics: Potential energy surface reconstruction for rate constant calculations

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Improving computational tools in reaction kinetics is a task of paramount importance, due to the great relevance that this field has in environmental and in industrial processes, such as those regarding combustion systems. In particular, the accurate determination of pressure dependent rate constants for gas phase systems poses significant challenges to theoretical modeling, especially for reactions involving radicals, that do not present saddle points in the Potential Energy Surface (PES).

The golden standard for this reaction class is the Variable Reaction Coordinate – Transition State Theory (VRC-TST) [1], that entails a computationally intensive Monte Carlo sampling of the PES, requiring the calculation of about 10^4 single point energies employing electronic structure software and using multireference theories, needed to account for the open shells of diradical systems. Within this context, artificial intelligence holds tremendous promise.

This proposal aims at mapping the PES as a function of the six relative degrees of freedom of the two interacting fragments characterizing the reaction, developing Artificial Neural Network (ANN) models incorporating as much physical information as feasible, that are supposed to substitute the electronic structure software in the extensive sampling required by the VRC-TST protocol. The ANN efficacy is evaluated in the computation of rate constants for gas phase reactions showing an accuracy within 20% of theoretical values.

This approach exhibits remarkable perspectives for increasing the robustness of the VRC-TST protocol, mainly due to the decoupling of the training of the ANN from the VRC-TST calculations, allowing the routine performing the necessary thousands of single point energy calculations to be optimized. Once these are done, the ANN training and the VRC-TST calculations performed require a computational time that is negligible with respect to that required for the electronic structure calculations, thus showing impressive time savings while maintaining accuracy.

References

[1] Georgievskii, Y., Klippenstein, S.J., Transition State Theory for Multichannel Addition Reactions: Multifaceted Dividing Surfaces. J. Phys. Chem. A 107 (2003) 9776-9781.