Machine Learning Lateral Interactions: Beyond DFT

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Under industrial operating conditions, heterogeneous catalysts surfaces are densely populated, and lateral interactions have pronounced effects on the catalytic mechanisms.[1,2] Lateral interactions can influence the trajectory of elementary reaction steps or prevent the surface diffusion of critical intermediates between active sites. Modelling these effects at high chemical accuracy (i.e. at the level of DFT) is computationally intractable due to the inherently large set of adlayer configurations. To overcome this complexity, we have developed an artificial neural network method to map the potential energy surface of the adlayers, specifically targeting the lateral interactions. This enables us to perform high-frequency Monte Carlo (MC) sampling close to quantum chemical accuracy. Deep learning networks interpret the MC trajectories to construct temperature-dependent lateral interaction potentials. These potentials have been used to elucidate high-coverage effects of diffusion through incorporation into microkinetic models, providing new insights into equilibrium adlayer configurations. A systematic formalism for network construction lent itself to a low-cost, fully-automated process, which in turn allowed a direct connection to be made between DFT parameters, such as in the XC-functional, and microkinetic output. Feedback between microkinetics and experimental results led to an adaptive modelling strategy, with which the nature of the surface processes could be understood. As microkinetic models can readily be incorporated at the reactor level, this marks an important step towards full multiscale modelling in catalysis.

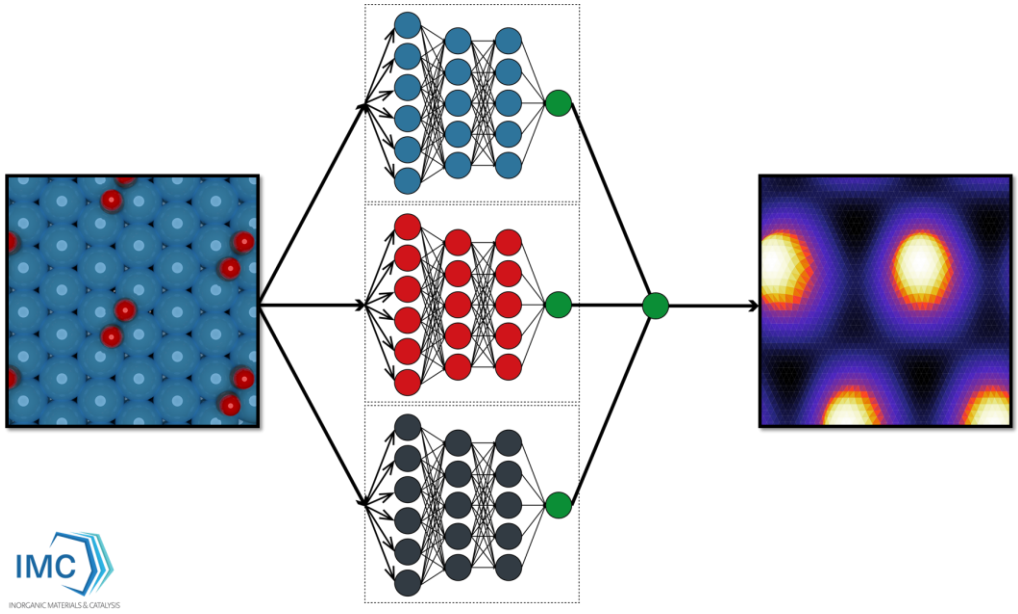

Figure 1: Mapping structures to potential energy surfaces using artificial neural networks

References
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