Predicting catalytic activity and phase transitions in liquid metal alloys by molecular dynamics with machine-learned force fields

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Supported catalytically active metal solutions (SCALMS) constitute a promising new catalyst concept. SCALMS are droplets of a low melting metal matrix metal (Ga, In, Sn, GaIn, GaSn, GaCu, GaAg) with a small amount of dissolved active metal, here Pt, on a support. At reaction temperatures, the droplets are liquid and the dissolved active metal atoms frequently appear at the surface of the droplet. In this way, dynamically single atoms are provided for catalysis. SCALMS turned out to be highly active catalysts for direct alkane dehydrogenation, which show high coking resistance. Coking has been a major obstacle for large-scale industrial applications of direct alkane dehydrogenation.

Predicting the catalytic activity of SCALMS by theory requires the determination of the distribution of elements at the surface of SCALMS. Molecular dynamics (MD) simulation of slab models of SCALMS can yield this information. However, classical MD is not suited for the description of liquid metals that do not exhibit stable bonds between individual atoms. Ab-initio MD based on density-functional theory is able to describe with high accuracy liquid metals but is computationally too expensive to be applied to more than the simplest systems. Here we present MD with force fields generated by machine learning from ab-initio MD runs. The resulting approach has almost the quality of ab-initio MD but requires only a fraction of the computational costs and thus enabled a screening of SCALMS with various compositions. In combination with the calculation of the position of the d-band of the active metal, the catalytic activity of SCALMS for propane dehydrogenation could be predicted.

Furthermore, MD with machine-learned force fields turned out to be capable to describe the formation of intermetallic phases, which are frequently observed in liquid metal mixtures and therefore are highly relevant for SCALMS.

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[2] A. Shahzad, et al., J. Phys.: Condens. Matter 2024, **36**, 175403, 10.1088/1361-648X/ad1e9f