Activating the Basal Plane of 2D Transition Metal Dichalcogenides via High-Entropy Alloying

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Abstract

Two-dimensional (2D) materials, such as transition metal dichalcogenides (TMDCs) in the 2H or 1T crystal phases, are promising (electro)catalyst candidates due to their high surface to volume ratio and composition of low-cost, abundant elements. While the edges of elemental TMDC nanoparticles, such as MoS_2, can show significant catalytic activity, the basal plane of the pristine materials are notoriously inert, which limits their normalized activity. Here, we show that high densities of catalytically active sites can be formed on the TMDC basal plane by alloying elements that prefer the 2H (1T) phase into a 1T (2H) structure. The global stability of the alloy, in particular whether it crystallizes in the 2H or 1T phase, can be controlled by ensuring a majority of elements preferring the target phase. We further show that the mixing entropy plays a decisive role for stabilizing the alloy implying that high-entropy alloying becomes essential. Our calculations point to a number of interesting non-precious hydrogen evolution catalysts, including $(CrTaVHfZr)S_2$ and $(CrNbVTiZr)S_2$ in the 1T-phase and $(MoNbTaVTi)S_2$ in the 2H-phase.[1] This work lays the ground for further exploration and development of high-entropy alloy (HEA) TMDCs as catalysts for hydrogen evolution. While we have proposed an effective design principle for HEAs that combines stability and catalytic activity, numerous challenges and opportunities remain for future research. The vast compositional space of HEA TMDCs, coupled with their potential for entropic stabilization, opens several intriguing avenues for investigation. Traditional computational approaches could be combined with recent advances in big data analytics and machine learning (ML) to accelerate progress in this area. Additionally, the use of ML-based atomic force fields and other surrogate models trained on density functional theory (DFT) data could enable the efficient exploration of non-equimolar HEA compositions, which were not addressed in this study.

References

[1] Akhound, Mohammad Amin, Karsten Wedel Jacobsen, and Kristian Sommer Thygesen. "Activating the Basal Plane of 2D Transition Metal Dichalcogenides via High-Entropy Alloying." Journal of the American Chemical Society, ASAP (2025).

Figures

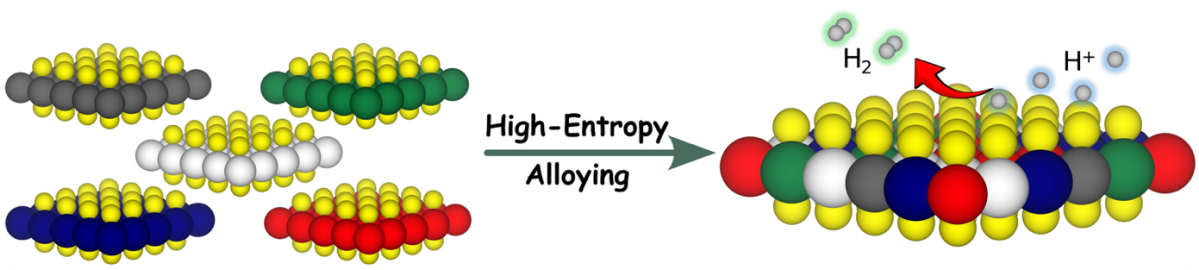

Figure 1: Graphical abstract of activating the basal plane of 2D TMDCs via high-entropy alloying.