

Revealing the Dynamic Behavior of Heterogeneous Catalysts via Machine Learning-Driven Molecular Dynamics

Umberto Raucci [1], Axel Tosello Gardini [1,2], Michele Parrinello[1]

1. Italian Institute of Technology, Via E. Melen 83, Genoa, 16152

2. Università di Milano-Bicocca, Via R. Cozzi 55, Milan, 20125

The traditional view of heterogeneous catalysts as static entities is increasingly challenged by the evidence that their activity often stems from their ability to undergo significant transformations at the *operando* conditions. This dynamic behavior is essential for understanding how catalysts maintain stability and activity under the harsh conditions of industrial reactors, which involve high temperatures, pressures, and intense flows of reactants and products. Despite extensive efforts, many fundamental questions about the stability and behavior of catalysts under *operando* conditions remain unanswered. Investigating these dynamic transformations is critical to develop more efficient catalytic processes that can meet the demands of a sustainable and green economy. Our study leverages recent advancements in machine learning-driven molecular dynamics simulations to explore the behavior of catalysts in these challenging conditions [1]. We focus on the thermally activated synthesis of ammonia catalyzed by barium hydride (BaH_2). This system has been experimentally characterized [2], showing high catalytic activity if and only if it is alternately exposed first to N_2 and later to H_2 in a chemical looping process [3]. Our simulations reveal that BaH_2 undergoes a profound transformation when exposed to N_2 , forming a superionic mixed compound, $\text{BaH}_{2-2x}(\text{NH})_x$, characterized by high mobility of hydrides and imides. This transformation is not limited to the surface but involves the entire catalyst and facilitates the subsequent exposure to H_2 , leading to the production of ammonia and regeneration of the catalyst. Contrary to the traditional view of catalysts as static providers of active sites, our findings demonstrate that the effectiveness of a catalyst can be deeply rooted in its dynamic transformations and behavior under reaction conditions.

References

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[2]. Nature Materials 2015, 14, 95

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