

Mixed Non Noble/Noble Catalytic Interfaces for Ammonia Decomposition at Lower Temperature

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Introduction. Hydrogen is considered as one future energy vector due to its high mass energy density. However, its low energy density by volume (8 MJ.L^{-1}) makes it difficult to store and distribute. Hydrogen carriers, able of absorbing and releasing it through different chemical reactions, can be used to densify it. Ammonia is a promising decarbonated carrier of hydrogen, due to its high hydrogen mass fraction (17.8 wt%) and its easy liquefaction.[1] However, the required high pressure and high temperature, for synthesis and decomposition, affects the overall energetic efficiency of the storage process. In this work, we are interested in the catalyzed decomposition reaction of ammonia aiming to regenerate hydrogen. Ruthenium, one of the reference metal catalysts for working in milder conditions, cannot be considered on an industrial scale because of its high cost and limited abundance. Other catalytic systems, such as alkali or alkaline-earth metal amides, present competitive catalytic performance with minimal cost. In particular, lithium amide (LiNH_2) when coupled with other transition metals (TM). [2] From a joint experimental and theoretical investigation of LiNH_2 – TM based catalytic systems, we propose to address this difficult question of understanding and improving the efficiency of these catalytic systems.

Results and discussion. From the experimental standpoint, synthesis, characterization and catalytic performance are currently examined. In parallel, density functional theory calculations are performed to model $\text{Ru}(0001)/\text{LiNH}_2(001)$ interfaces. The solid-solid contact helps to stabilize both materials. $\text{Ru}(0001)$ thermodynamic termination interacts preferably with NH_2 molecular planes in LiNH_2 materials. A thinner LiNH_2 slab will exhibit a larger ability to adapt to the Ru surface, thus minimizing the cost of the crystalline mismatch. The most stable system is obtained when the terminal plane of $\text{LiNH}_2(001)$, opposite to the contact with Ru, is reconstructed with a 180° rotation of NH_2 fragments (pointing now towards vacuum) around the axis perpendicular to the Ru/ LiNH_2 interface. [3] Based on this theoretical study, the catalytic mechanism is currently being examined, in particular to determine the main surface intermediates of this complex reaction mechanism.

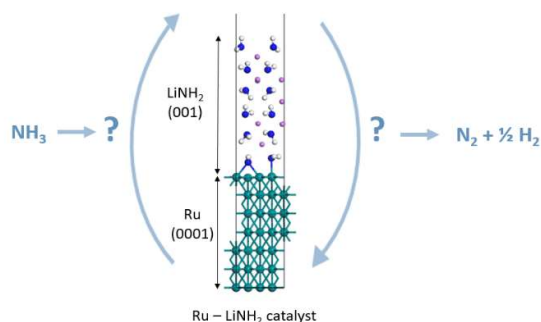


Fig. 1. Lateral views of the most stable $\text{Ru}(0001)/\text{LiNH}_2(001)$ optimized, base of the model for the study of the catalytic activity

[1] C. H. Christensen et al., Catal Today, **111**, 140-144 (2006)

[2] J. Guo et al., Angew. Chem. Int. Ed Engl, **54**, 2950-2954 (2015)

[3] J. Dorival, J. Delmas and D. Loffreda, submitted J. Phys. Chem. C (2023)