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ABSTRACT:

Mg-doped LaNiO3 perovskites for ammonia decomposition: Kinetic Analysis at low temperatures

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Ammonia, as a sustainable and carbon-free hydrogen carrier, plays a vital role in the hydrogen economy. Developing new catalytic formulations with less critical materials is fundamental for improving both sustainability and energy efficiency [1]. Nickel-based perovskite catalysts are a promising and cost-effective alternative to Ru-based systems. The exsolution of Ni during reduction creates nanosized active metal sites with strong metal-support interactions. Additionally, the incorporation of electron-donating alkali and alkaline earth metals into perovskites enhances the electron-rich environment around the Ni sites, boosting catalytic performance [2]. In this study, Mg-doped $LaNiO_3$ perovskites have been synthesized via sol-gel combustion and characterized by N_2-physisorption, XRD, H_2-TPR, CO_2-TPD CO-chemisorption, TEM, and XPS. In-situ DRIFT spectroscopy was used for NH_3-temperature programmed desorption (TPD) experiments to gain insights into the reaction mechanism. The results show that the $MgNiO_2$ phase enhances the stability of the $La_{0.1}Mg_{0.9}NiO_3$ catalyst, as confirmed by TPR. Strong metal- metal interactions lead to the formation of small Ni particles during reduction without sintering, as demonstrated by XRD and CO-chemisorption. The electron-donor properties of Mg promote the dehydrogenation and N_2 desorption stages, as revealed by CO_2-TPD experiments. XPS analysis confirmed the partial reduction of Ni^{3+} to Ni^{2+}, consistent with XRD findings, and the presence of hydroxyl groups in the perovskite structure. Kinetic studies reveal that the reaction proceeds according to the Temkin-Pyzhev mechanism, where the recombinative desorption of N* is the rate-determining step.

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[2]. M. Pinzón, A. Sánchez-Sánchez, A. Romero, A.R. de la Osa, P. Sánchez, Fuel, 323, 124384 (2022).