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The impact of nitrogen mobility on the activity of zirconium oxynitride catalysts for ammonia decomposition

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Abstract

A zirconium oxynitride catalyst was used for the decomposition of ammonia to hydrogen and nitrogen. The onset of catalytic activity at ~550 °C coincided with the onset of nitrogen ion mobility in the material and a phase change from the initial β' phase ($\sim Zr_7O_{11}N_2$) to the nitrogen-rich β'' ZrON phase ($\sim Zr_7O_{9,5}N_3$). No hydrazine formation during an extended time on stream was detectable. Moreover, the onset of activity was also correlated to a rapid change in the electronic structure of the surface accompanying formation of the more active β'' ZrON phase. The results presented here show for the first time a direct correlation among the onset of ion conductivity as a bulk property, a modified electronic structure of the surface, and the catalytic performance of a heterogeneous catalyst.