

Kinetic studies of ammonia oxidation over noble metals by means of end-of-pipe and spatially resolved measurements

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Ammonia is one of the top five air pollutants regulated by the EU. However, progress in reducing ammonia emissions is lagging behind that of the other air pollutants. While most ammonia emissions originate from the agricultural sector, ammonia is gaining importance as a carbon-free energy source, either for direct use as a fuel or as a hydrogen storage medium. Here, ammonia slip during direct combustion or decomposition is one of the biggest challenges for future emission control systems. The emissions associated with the use of ammonia are particularly important as the products formed during the non-selective oxidation of ammonia, especially N₂O, are very potent greenhouse gases [1,2].

In this work, we present kinetic studies aimed at advancing the development of microkinetic mechanisms of ammonia oxidation over layered noble metal supported catalysts. We combine the systematic variation of operating conditions over a wide temperature range as well as inlet ammonia and oxygen concentrations in a 2D channel reactor with spatially resolved measurements using capillary-based techniques within monoliths and in a stagnant flow configuration. The data confirm the known very high catalytic activity of noble metals for ammonia oxidation, but also cover the aforementioned formation of secondary emissions (N₂O and NO) and the strong shifts in product selectivity between N₂, N₂O and NO as a function of temperature, stoichiometry and to some extent spatial position, covering all major pathways suggested in recent mechanistic considerations [3], which can be used to refine microkinetic mechanisms. Detailed reaction mechanisms will allow us to model the reactor behavior to optimize catalyst performance and selectivity and propose different/novel strategies for the selective oxidation of ammonia.

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