Public summary of PhD-thesis of Giulia Spezzati
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Increasing the contact between palladium and cerium oxide for a cleaner environment

In the current situation of high energy demands, it is imperative to develop catalytic systems that include smaller quantities of expensive noble metals without compromising the performance. This result can be achieved by engineering the contact and interaction between the noble metal and the support at the atomic scale. I developed and studied catalysts composed of different nanostructured cerium oxide (CeO_2) supports onto which only 1 wt.% palladium was deposited. It was determined that the enhanced metal-support interaction leads to improved performance at temperatures lower than 100 °C in environmental reactions such as exhaust treatment reactions.

Since the 18th century, mankind has used catalysis to speed up the rate of a reaction, to be able to selectively obtain a higher amount of a desired product, or to operate at milder industrial conditions. In the recent decades, exceptional progress has been made in catalyst design and applications. A current trend in catalysis research focuses on decreasing the quantity of expensive and rare noble metals (platinum, rhodium, palladium, etc.) without compromising on catalytic performance.

In my PhD research, I focused on the synthesis of nanostructured cerium oxide supports, either pure or doped with inexpensive and abundant transition metals such as copper or iron. The supports were prepared either in an autoclave or with a flame-spray procedure. Cerium oxide is an excellent support because it can store and release oxygen during reaction cycles. This is important, because it helps with maintaining the optimal oxygen quantity to achieve maximum conversion of all pollutants. The interaction of these supports with palladium nanoparticles and single atoms was evaluated via *in situ* experimental techniques under realistic reaction conditions. The influence of how the cerium oxide surface atoms are arranged and how they interact with palladium was thoroughly assessed. My work revolved around the evaluation of these samples for applications known for their strong impact on the environment, such as the oxidation of carbon monoxide and three-way catalysts model reactions. An engine has the potential of releasing copious amounts of carbon monoxide, unburnt hydrocarbons and nitrogen oxides into the atmosphere, especially at low temperatures, such as when the vehicle has just been started. These compounds are dangerous both for human health and for the environment, for instance because of their global warming potential.

The palladium/cerium oxide samples showed an excellent activity at temperatures lower than 100 °C, both in terms of converted reactants and in selectivity towards non-harmful products. An especially high performance was obtained when palladium was supported on iron- or copper-doped cerium oxide. The presence of one of these transition metals in the cerium oxide crystal enhances oxygen availability, which is able to boost the reactivity in the aforementioned reactions. Moreover, we proved the existence of a significant palladium-copper synergy, which is a key player in the conversion of nitrogen oxides to inert nitrogen.

Title of PhD-thesis: Metal-support interactions in Pd/CeO_2 for environmental catalysis. Promotor: Emiel J. M. Hensen, TU/e.