# Interfacial engineering core@shell nanoparticles for active and selective direct H2O2 generation

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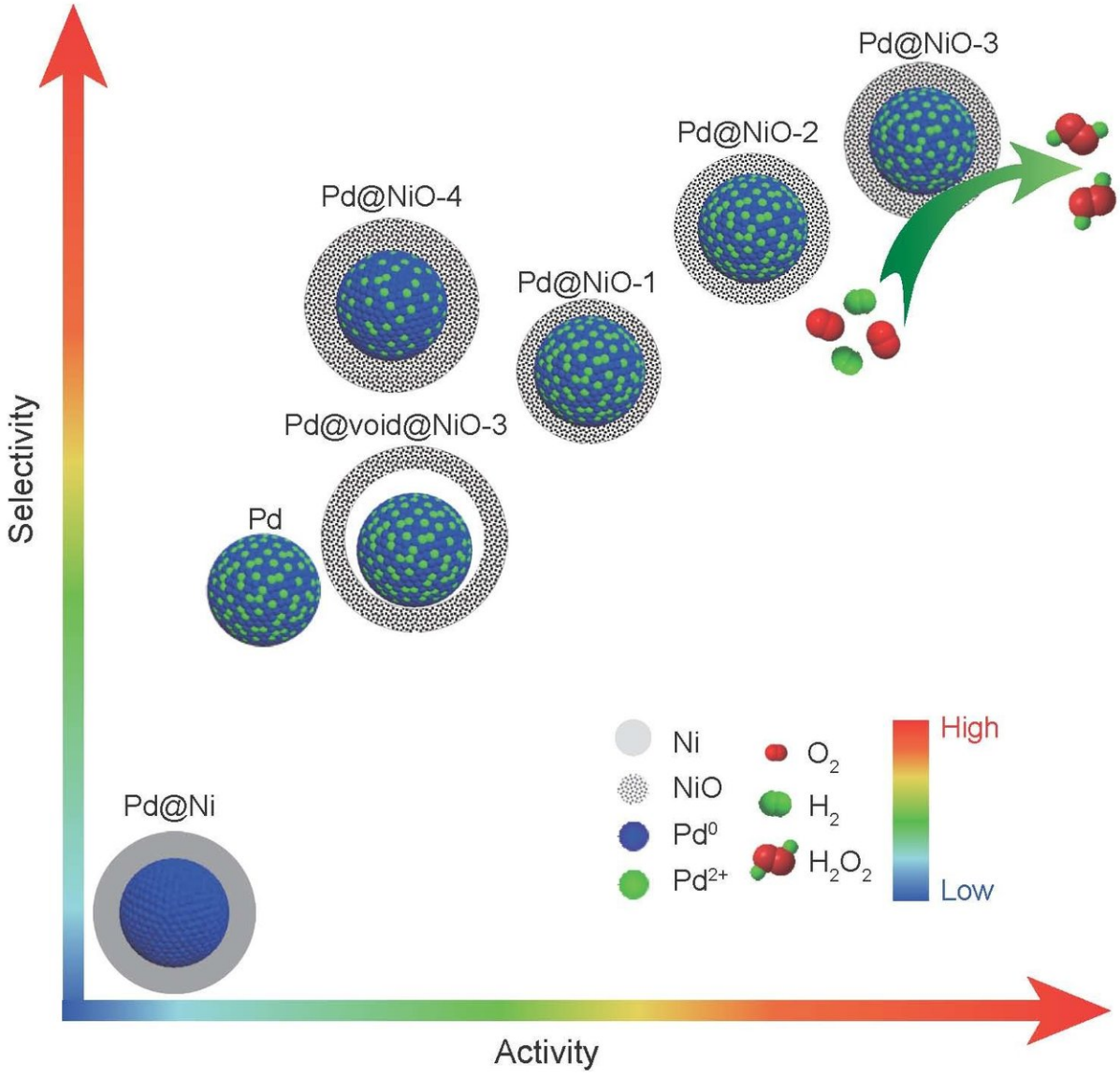

Schematic illustration showing the activity and selectivity toward H2O2 synthesis of 5 wt% Pd@Ni-3/TiO2, 5 wt% Pd@NiO-x/TiO2 (x = 1, 2, 3, 4), 5 wt% Pd@void@Ni-3/TiO2 and 5 wt% Pd/TiO2. Credit: Science China Press

A class of supported Pd@NiO-x core@shell catalysts have been constructed for direct $H_2O_2$ generation. The optimized Pd@NiO-3/$TiO_2$ exhibited high activity, superior selectivity, low degradation activity and excellent stability. The unique, cavity-contained interface structure can suppress the overbinding between Pd-core and (O-O)*, which is effective to prevent $H_2O$ formation and guarantees high selectivity of $H_2O_2$. The present work highlights the importance of interface engineering of Pd-based catalysts for direct $H_2O_2$ synthesis.

Hydrogen peroxide ($H_2O_2$) is a versatile chemical, widely applied in modern industry. To date, $H_2O_2$ is industrially manufactured by an indirect process that involves the sequential hydrogenation and oxidation of alkyl anthraquinone, an energy-intensive, multi-step process with high cost. By contrast, the direct synthesis of $H_2O_2$ from H2 and O2 is expected to be the most efficient way to produce $H_2O_2$ due to the remarkable advantages of atom economy, low energy consumption and $H_2O$ as its only byproduct.

Currently, the direct synthetic route is mainly achieved by the supported Pd-based catalysts. The major problem associated with that is related to the low selectivity of $H_2O_2$. Despite great efforts devoted to constructing Pd-based catalysts, understanding high-performance Pd-based catalysts for direct $H_2O_2$ generation from either deep characterization or theoretical investigation are still extremely limited.

In a new overview published in the Beijing-based *National Science*

*Review*, scientists at the Soochow University present the latest advances in direct $H_2O_2$ generation. Co-authors Yonggang Feng, Qi Shao, Bolong Huang, Junbo Zhang, and Xiaoqing Huang developed a class of Pd@NiO-x nanoparticles with a unique core@shell interface structure, which achieves high activity, selectivity and stability for the direct $H_2O_2$ synthesis.

These scientists interpreted the mechanism from both electronic and energetic views. "Traditional Pd-based catalysts are very active for the side reactions, such as the decomposition and hydrogenation of $H_2O_2$ as well as the formation of H2O," they state in an article titled "Surface engineering in the interface of core/shell nanoparticles promotes hydrogen peroxide generation."

"It is considered that the intrinsic surface property of Pd-based catalysts is essential for the selectivity and activity of the direct $H_2O_2$ synthesis," they add. "This arises because the barrier for O-O bond scission is sensitive to Pd surface structure, the key parameter governing $H_2O_2$ synthesis and decomposition activity."

The creation of porous NiO shell is beneficial for exposing Pd active sites and thus enhancing the productivity of $H_2O_2$. "By tuning the composition of Pd@NiO-x NPs and the reaction condition, the efficiency of $H_2O_2$ synthesis could be well optimized with 5 wt% Pd@NiO-3/TiO2 exhibiting the highest productivity (89 mol/(kgcath)) and selectivity (91%) to $H_2O_2$ as well as excellent stability," they state.

"The first principles simulations further revealed the mechanism from both electronic and energetic views," the scientists wrote. "The superiority in selectivity is achieved by a spontaneous bond scission of H-H and charge transfer from O20 to O22- within the cavity of NiO interfacing with Pd surface. (...) The high selectivity and activity make it one of the best catalysts for the direct $H_2O_2$ synthesis reported to date,"

they add. "The present work reported here highlights the importance of surface and interface engineering of Pd-based catalysts for the direct $H_2O_2$ synthesis with largely enhanced activity and selectivity."

**More information:** Yonggang Feng et al, Surface engineering at the interface of core/shell nanoparticles promotes hydrogen peroxide generation, *National Science Review* (2018). DOI: 10.1093/nsr/nwy065

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