Interfacial engineering core@shell nanoparticles for active and selective direct H\textsubscript{2}O\textsubscript{2} generation

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A class of supported Pd@NiO-x core@shell catalysts have been constructed for direct \( \text{H}_2\text{O}_2 \) generation. The optimized Pd@NiO-3/TiO\textsubscript{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 \( \text{H}_2\text{O} \) formation and guarantees high selectivity of \( \text{H}_2\text{O}_2 \). The present work highlights the importance of interface engineering of Pd-based catalysts for direct \( \text{H}_2\text{O}_2 \) synthesis.

Hydrogen peroxide (\( \text{H}_2\text{O}_2 \)) is a versatile chemical, widely applied in modern industry. To date, \( \text{H}_2\text{O}_2 \) 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 \( \text{H}_2\text{O}_2 \) from \( \text{H}_2 \) and \( \text{O}_2 \) is expected to be the most efficient way to produce \( \text{H}_2\text{O}_2 \) due to the remarkable advantages of atom economy, low energy consumption and \( \text{H}_2\text{O} \) 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 \( \text{H}_2\text{O}_2 \). Despite great efforts devoted to constructing Pd-based catalysts, understanding high-performance Pd-based catalysts for direct \( \text{H}_2\text{O}_2 \) 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 \( \text{H}_2\text{O}_2 \) 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 \( \text{H}_2\text{O}_2 \) 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 \( \text{H}_2\text{O}_2 \) as well as the formation of \( \text{H}_2\text{O} \)," they state in an article titled "Surface engineering in the interface of core/shell nanoparticles promotes \text{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 H₂O₂ synthesis," they add. "This arises because the barrier for O-O bond scission is sensitive to Pd surface structure, the key parameter governing H₂O₂ synthesis and decomposition activity."

The creation of porous NiO shell is beneficial for exposing Pd active sites and thus enhancing the productivity of H₂O₂. "By tuning the composition of Pd@NiO-x NPs and the reaction condition, the efficiency of H₂O₂ synthesis could be well optimized with 5 wt% Pd@NiO-3/TiO₂ exhibiting the highest productivity (89 mol/(kgcath)) and selectivity (91%) to H₂O₂ 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 O²⁻ to O²⁻ within the cavity of NiO interfacing with Pd surface. (...) The high selectivity and activity make it one of the best catalysts for the direct H₂O₂ 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 H₂O₂ synthesis with largely enhanced activity and selectivity."


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