Tandem catalysis improves selective oxidation of methane to oxygenates

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Selective oxidation of methane (CH₄) to value-added chemicals with both high catalytic activity and selectivity under mild conditions remains challenging. Due to the low activity of oxygen and the overoxidation of the oxygenates, selective oxidation of CH₄ to oxygenates with O₂ or O₂/H₂ suffers from low catalytic activity and low oxygenates selectivity. Moreover, the high loading of noble metals for supported catalysts leads to high cost.

Recently, a research team led by Prof. Sun Yuhan and Prof. Zhong Liangshu from the Shanghai Advanced Research Institute (SARI) of the Chinese Academy of Sciences reported a ZSM-5 (Z-5)-supported PdCu catalyst for selective oxidation of CH₄ to oxygenates using O₂ in the presence of H₂. The catalyst exhibited a high oxygenates yield of 1178 mmol/gₚd/h with oxygenates selectivity of 95% at 120 °C.

The results were published in Angewandte Chemie International Edition.

Based on a combination of control experiments and electron paramagnetic resonance as well as in situ spectroscopic techniques, the researchers found that PdO nanoparticles facilitated in situ generation of H₂O₂. Cu single atoms not only accelerated the generation of abundant ·OH from H₂O₂ decomposition, but also enabled the homolytic cleavage of CH₄ by ·OH to ·CH₃. Subsequently, the ·OH reacted quickly with the ·CH₃ to form CH₃OH with high selectivity.

These findings may provide valuable insights into selective oxidation of methane to oxygenates and shed light on other highly efficient and low-price catalysts for the selective activation of C-H bonds in light alkanes.


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