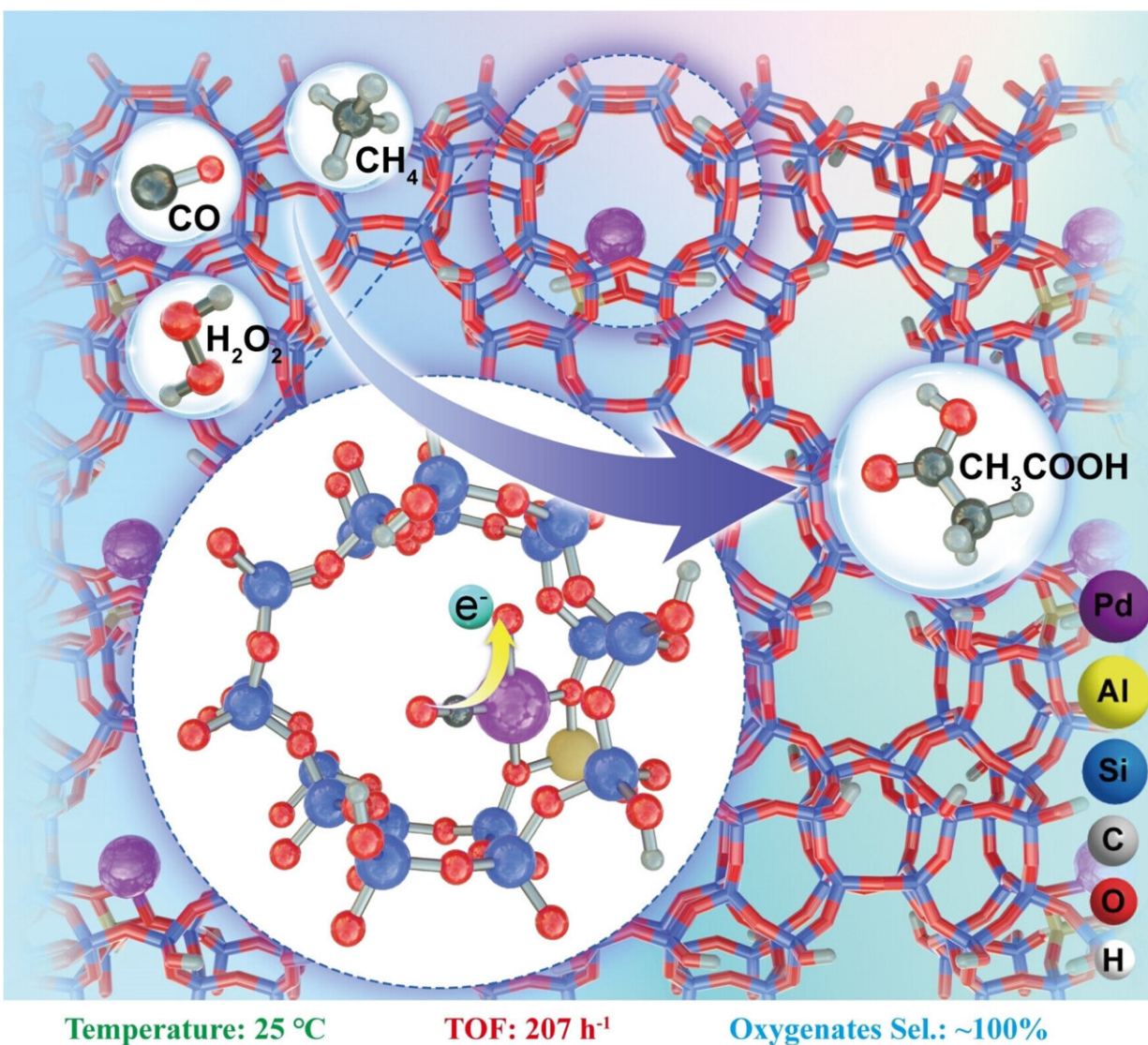


# Team modulates electronic state of single-atom catalysts by CO molecular decoration for efficient methane conversion

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In situ CO decoration can significantly tune the electronic state of metal-oxo center in single-atom catalysts for promoting oxygenates production. CO activated Pd<sub>1</sub>-ZSM-5 displays the highest turnover frequency of 207 h<sup>-1</sup> and ~100 % oxygenates selectivity with H<sub>2</sub>O<sub>2</sub> as oxidant at 25° C. Credit: *Angewandte Chemie International Edition* (2024). DOI: 10.1002/anie.202315343

Direct methane conversion has advantages such as low energy consumption, fewer processes, and better economics. However, it is difficult to activate methane at room temperature due to the high dissociation energy of C-H bonds of methane. Additionally, the target products, such as methanol, acetic acid, and other oxygenates, are prone to over-oxidation, resulting in the generation of CO<sub>2</sub>. Therefore, the design of catalysts with high activity and selectivity is important.

In a study [published](#) in *Angewandte Chemie International Edition*, a group led by Prof. Zhang Tao, Prof. Wang Xiaodong, and Assoc. Prof. Huang Chuande from the Dalian Institute of Chemical Physics (DICP) of the Chinese Academy of Sciences (CAS), collaborating with Prof. Chang Chunran's group from Xi'an Jiaotong University, has realized the efficient direct [conversion](#) of methane through single-atom catalysis at room temperature.

The researchers proposed a strategy involving CO molecule modification to regulate the electronic structure of single-atom catalyst M<sub>1</sub>-ZSM-5 (M = Rh, Ru, Fe), which enhances the efficiency of direct methane conversion. They realized catalyzing methane conversion with H<sub>2</sub>O<sub>2</sub> as an oxidant at room temperature (25 °C). The turnover frequency (TOF) of Pd<sub>1</sub>-ZSM-5 reached 207 h<sup>-1</sup> with nearly 100% selectivity towards oxygenates.

Combining experimental characterization with density functional theory

(DFT) calculations, the researchers discovered that the C atom in the CO molecule tends to coordinate with the Pd<sub>1</sub> [single atom](#), transferring electrons from CO to the active oxygen center L-Pd<sub>1</sub>-O (L = CO), resulting in the reduce of the dissociation barrier of C-H bonds of methane, from 1.27 eV to 0.48 eV.

Moreover, this strategy exhibited good universality as TOF of the M<sub>1</sub>-ZSM-5 (M = Rh, Ru, Fe) series catalysts could be increased by 3.2 to 11.3 times through CO molecule modification.

"We have developed the electronically tunable molecular sieve-supported M<sub>1</sub>-O isolated active centers, providing a new method to achieve selective [methane](#) conversion to chemicals under [mild conditions](#)," said Prof. Wang.

**More information:** Weibin Xu et al, Metal-Oxo Electronic Tuning via In Situ CO Decoration for Promoting Methane Conversion to Oxygenates over Single-Atom Catalysts, *Angewandte Chemie International Edition* (2024). [DOI: 10.1002/anie.202315343](https://doi.org/10.1002/anie.202315343)

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