
**Reveal and Correlate Working Geometry and Surface Chemistry of Ni
Nanocatalysts in CO₂ Reforming of Methane**

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ABSTRACT

The commercialization of Ni-based catalysts in CO₂ dry reforming of methane (DRM) suffers from their quick deactivation. Here, we reveal each reaction pathway for DRM based on the Ni catalyst composition and geometry under working conditions, through one working platform combining *in situ* high resolution Cs corrected environmental transmission electron microscopy and electron energy-loss spectroscopy coupled with mass spectroscopy. The formation of Ni₃C has been found to inhibit the decomposition of CO₂ and CH₄, and to promote the formation of onion-like carbon to encapsulate the Ni catalysts, leading to the deactivation of the Ni-based catalysts. Designing the suitable supports or promoters to keep the Ni surface structure under Ni-NiO cycle can drive the simultaneously amorphous carbon deposition-consumption cycle and minimise the coke formation. This research is not only for developing coke resistance Ni catalysts in the DRM, but also significant for investigating many catalysis challenges both in research and engineering.