The latest advance in imaging technology helps optimize catalysts for use in onboard fuel processing

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The presence of carbon monoxide (CO) impurities in hydrogen gas (H_2) can have a detrimental impact on the performance of fuel cells. Recent studies have shown that gold nanoparticles—particles less than five nanometers wide—can catalytically remove CO impurities from H_2 under mild temperature and pressure conditions. This breakthrough understanding has helped facilitate the development of fuel-cell vehicles that use 'onboard' fuel processing technology. Unfortunately, gold nanoparticles tend to lose their catalytic activity after a few hours of use—and scientists need to overcome this problem if gold nanoparticles are to be used.

Ziyi Zhong at the A*STAR Institute of Chemical and Engineering Sciences, Ming Lin at the A*STAR Institute of Materials Research and Engineering and co-workers have identified the subtle, atomic-scale structural transformations that can activate and de-activate gold nanoparticle catalysts, a finding that may lead to longer-lasting hydrogen fuel cells.

The researchers set out to design an improved catalyst for so-called preferential oxidation (PROX) reactions. This approach transforms CO impurities into carbon dioxide (CO_2) on a ceramic support containing metal catalysts. Previously, the team found that silica-based supports, called SBA-15, could boost CO removal by selectively absorbing the CO_2 by-product. The researchers took advantage of another SBA-15 characteristic—a mesoporous framework decorated by terminal amine groups—to engineer a novel PROX catalyst.

First, the team used amine modification to disperse a mixture of gold and copper(II) oxide (CuO) precursors evenly over the SBA-15 support. They then used heating treatment to generate gold and CuO nanoparticles on the SBA-15 support. The numerous pores in SBA-15 and the CuO particles work together to hinder agglomeration of gold nanoparticles—a major cause of catalyst de-activation.

The team then achieved a near-unprecedented chemical feat: localized structural characterization of their catalyst at atomic scale, using high-resolution transmission electron microscopy (HR-TEM) and three-dimensional electron tomography (see movie below). These imaging techniques revealed that the active catalyst sites—gold or gold–copper alloy nanoparticles in the immediate vicinity of amorphous and crystalline CuO—remained stable for up to 13 hours. However, the reducing atmosphere eventually transforms CuO into copper(I) oxide and free copper; the latter of which then alloys with the gold nanoparticles and deactivates them. Fortunately, heating to >300°C reversed the alloying process and restored the catalyst's activity.

"People working in catalysis are always curious about the 'local structures' of their materials," says Zhong. "Because the Au-CuO/SBA-15 catalyst is

active at room temperature, advanced characterization in our state-of-the-art facilities is possible—though it takes great patience and requires multidisciplinary collaboration."

More information: Li, X., Fang, S. S. S., Teo, J., Foo, Y. L., Borgna, A. et al. Activation and deactivation of Au–Cu/SBA-15 catalyst for preferential oxidation of CO in H2-rich gas. *ACS Catalysis* 2, 360–369 (2012). Article

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