

Gold-palladium nanoparticles achieve greener, smarter production of hydrogen peroxide

February 19 2009

Hydrogen peroxide is one of the world's most versatile and widely used chemicals. A powerful oxidizing agent, H_2O_2 is commonly used as a bleach, an antiseptic and a disinfectant.

Despite its importance, however, says Christopher J. Kiely, hydrogen peroxide has eluded the best efforts of the chemists seeking a more direct, efficient and environmentally friendly means of producing it.

"Hydrogen peroxide has for decades been made by an indirect energy-intensive process," says Kiely, a professor of materials science and engineering at Lehigh University.

There are other disadvantages, Kiely adds. The economics of the current production method requires H_2O_2 to be produced in large quantities and in solutions with concentrations much higher, and less stable, than those used in most practical applications. This necessitates storage and transporting, which can be hazardous.

Chemists have searched nearly a century for a catalyst that can directly combine hydrogen and oxygen to produce H_2O_2 . They have had some luck with palladium, says Kiely, but their efforts have been foiled by a second problem - as fast as H_2O_2 is produced, it can decompose to water in the presence of the catalyst.

Now, a group of chemists and materials scientists from the UK and the U.S. is reporting that a carefully tailored alloy of palladium and gold nanoparticles catalyzes the direct production of H_2O_2 while "switching off" the decomposition of the compound. The breakthrough, which culminates more than five years of research on the topic, promises to enable the on-site production of H_2O_2 in smaller quantities and more desirable concentrations.

In an article in the Feb. 20 issue of *Science*, one of the world's foremost science journals, the group says the decomposition of H_2O_2 can be greatly reduced by depositing gold-palladium nanoparticles on a high-surface-area carbon support that has first been washed with nitric acid. The pretreatment decreases the average size of the particles from a range of 2 to 70 nanometers (1 nm equals one-billionth of a meter) to a range of 2 to 25 nm. The washing also results in a more effective spatial distribution of the nanoparticles, enabling them to block the active sites on the carbon support that are responsible for the decomposition of H_2O_2 .

"We learned that neither the concentration of the nitric acid nor the length of time of the washing was important," says Kiely. "What was important was to wash the support in nitric acid before putting the gold-palladium nanoparticles on it. The resulting change in particle size and distribution enables us to retain a lot more of the hydrogen peroxide and to make the direct process more economically viable."

The *Science* article, titled "Switching Off Hydrogen Peroxide Hydrogenation in the Direct Synthesis Process," was coauthored by Kiely, Graham J. Hutchings, Jennifer K. Edwards, Benjamin Solsona, Edwin Ntainjua N, Albert F. Carley and Andrew A. Herzing.

Hutchings, the lead author, is the director of the Cardiff Catalysis Institute (CCI) in the UK. Edwards, Solsona, Ntainjua N and Carley are

members of the CCI. Herzing, who earned a Ph.D. from Lehigh in 2006, operates the aberration-corrected electron microscopy facilities in the Surface and Microanalysis Science Division of the U.S. National Institute of Standards and Technology (NIST). Kiely directs the Nanocharacterization Laboratory in Lehigh's Center for Advanced Materials and Nanotechnology.

The group owes its current success to Hutchings' expertise in catalysis and to his longstanding collaboration with Kiely, who has the ability to obtain data using electron microscopes with unmatched imaging and chemical analysis capabilities.

Hutchings and Kiely have been studying the catalytic potential of gold nanoparticles for 15 years, coauthoring four papers in the past four years on the topic for Science and Nature. In 2006, they reported the potential of gold-palladium nanoparticles to oxidize primary alcohols to aldehydes in a more environmentally friendly manner. That reaction is important to the production of spices and perfumes. In 2008, they reported that bilayer clusters of gold nanoparticles measuring about 0.5 nm in diameter were responsible for enabling the oxidation of CO to CO₂.

Their research has benefited from the aberration-corrected scanning transmission electron microscopes (STEMs) at Lehigh as well as NIST. Lehigh was the first university in the world to acquire two of the instruments, whose aberration correctors greatly improve imaging resolution and chemical analysis capability by overcoming distortions in the lenses that tend to blur the electron beam.

In the current project, aberration-corrected STEMs at Lehigh and NIST were used to measure the composition and particle size distribution of the gold-palladium alloy, and to understand how they change with various acid-washing pretreatments.

The researchers employed several characterization techniques, including High-Angle Annular Dark Field (HAADF) imaging to measure the change in nanoparticle size and energy-dispersive x-ray (XEDS) analysis to determine the composition of individual alloy particles.

"Without the aberration-corrected STEMs, we would not have been able to unravel what was going on in this instance," says Kiely.

In addition to performing experiments on the gold-palladium catalyst, the researchers ran parallel control experiments on pure gold and pure palladium separately.

"We found it was important for the palladium to incorporate just a small amount of gold," says Kiely. "The gold appears to modify the electronic structure and thus the catalytic activity of the palladium."

Source: Lehigh University

Citation: Gold-palladium nanoparticles achieve greener, smarter production of hydrogen peroxide (2009, February 19) retrieved 30 April 2024 from <https://phys.org/news/2009-02-gold-palladium-nanoparticles-greener-smarter-production.html>

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