Carbon-Encapsulated Iron Particles for Less Expensive, More Efficient Fuel Cells

Fuel cells are an important power source for an array of vehicles and other applications. However, the lack of cost-effective catalysts has prevented the use of fuel cells from spreading more widely. Iron-based non-precious metal catalysts have long been seen as an abundant, low-cost alternative to state-of-the-art platinum-based catalysts. Yet, such iron-based catalysts have remained out of reach due to challenges in identifying exactly which compound in the iron-containing, non-precious metal catalyst facilitates the oxygen reduction reaction. Researchers working at the APS used a sequential chlorine/hydrogen gas treatment to selectively remove all of the components of an iron-based catalyst that were not involved in oxygen reduction, leaving a single type of particle as the catalyst's active form: a carbon-encapsulated iron nanoparticle. This discovery may improve catalyst function and lower production costs, leading to more efficient, less expensive fuel cells that can be useful in vehicles and other power-hungry applications such as backup power generators and satellites.

Researchers from the University of Illinois at Urbana-Champaign, Knox College, Argonne, Yeshiva University, and Kyushu University (Japan) demonstrated that a high-temperature (900° C) gas-phase chlorine treatment deactivated non-precious metal catalysts, while a high-temperature (900° C) hydrogen treatment completely reactivated the catalysts (Fig. 1). They observed that iterative rounds of the treatments decreased the heterogeneity of the catalyst, which then allowed for the direct characterization of the species present in the deactivated and reactivated catalysts.

The team of researchers then used Mössbauer and x-ray absorption spectroscopy (XAS), carried out at the 9-BM-B,C beamline and MR-CAT

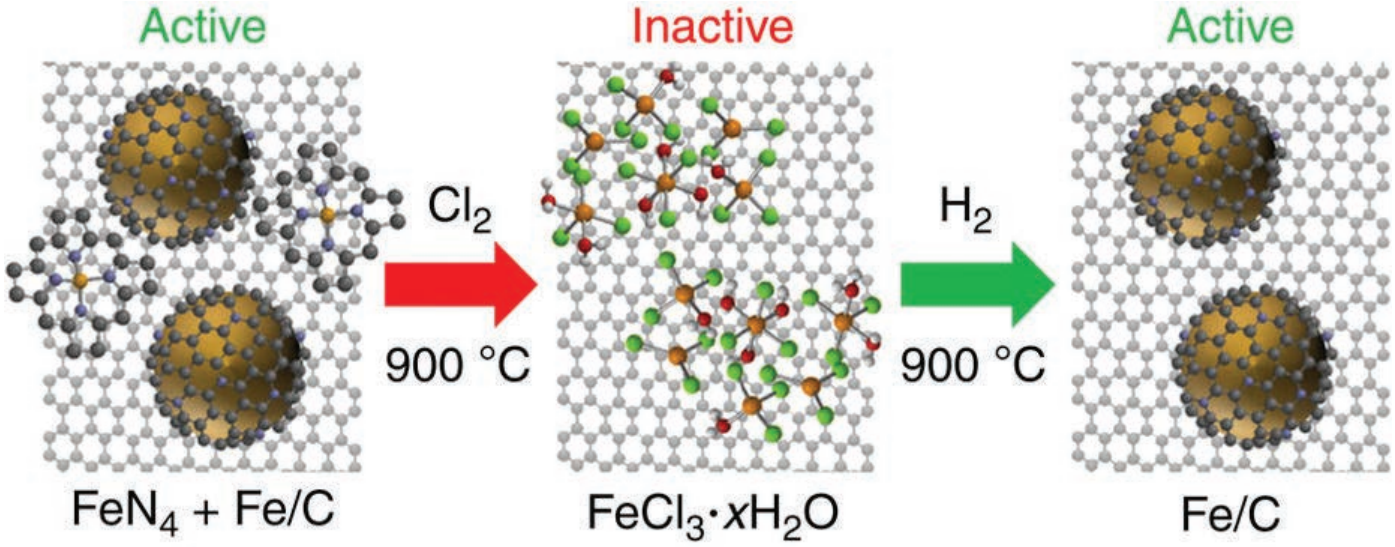

Fig. 1. Effect of a sequential high-temperature chlorine/hydrogen treatment on the activity of an iron-containing non-precious metal catalyst. Metallic Fe and FeN_4 species are converted into dispersed $FeCl_3 \cdot xH_2O$ that is then reformed into reduced Fe species by the H_2 treatment. The absence of FeN_4 sites in the H_2-treated catalyst indicates that these sites are not required for the observed oxygen reduction reaction activity.

beamline 10-BM-A,B at the APS, to identify the active species in the catalyst. The former technique was used to investigate nuclear structure using the absorption and re-emission of gamma rays, while the latter technique was used to deduce the local atomic environment of a compound by analyzing the way the compound absorbs a spectrum of x-rays.

The researchers observed that the untreated iron-based catalyst contained

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a combination of FeN_4 and reduced iron (Fe) species, making the direct characterization of the active species impossible, as expected. However, following the chlorine treatment, the Mössbauer and XAS analyses indicated that the metallic Fe and FeN_4 species were converted into dispersed $FeCl_3 \cdot xH_2O$, which is then reformed by the H_2 treatment into iron nanoparticles encapsulated by a carbon shell that helps stabilize the reactivated catalyst (Fig.1).

The researchers also used XAS, along with transmission electron microscopy, to measure the size of the nanoparticles. Their size indicates that the nanoscale features of the iron particles in the catalyst might contribute to their activity. Further experiments using x-ray photoelectron spectroscopy indicated that the surface of the carbon-encapsulated iron nanoparticles were doped with nitrogen, which the researchers speculated could also increase the oxygen reducing activity of the catalyst.

These findings may allow for the design and synthesis of purer forms of non-precious metal oxygen reduction catalysts with a higher loading of the active species. The research team will explore optimizing the catalyst nanoparticles by changing their size and increasing their abundance, using multicomponent nanoparticles, and doping the surface of the catalyst with a variety of elements.

Catalyst enhancements derived from these experiments could possibly bring down the price of fuel cells, making them cost-effective for more types of applications, including powering automobiles. *— Chris Palmer*

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