

OXYGEN REDUCTION REACTION

Bleached carbon

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Materials based on composites of iron, nitrogen and carbon (Fe–N–C) are promising alternatives to the expensive platinum catalysts used for the oxygen reduction reaction (ORR) that occurs at the cathode of proton-exchange membrane fuel cells. While gains have been made in the activity and even the stability of these materials under certain conditions, in working fuel cells their long-term performance is not sufficient. In particular, the presence of hydrogen peroxide (H_2O_2), a byproduct that forms when O_2 is only partially reduced, is known to have a negative impact on the activity of Fe–N–C catalysts, but the details of how this occurs are poorly understood. Now, Chang Hyuck Choi, Hyungjun Kim, Frédéric Jaouen and colleagues in the Republic of Korea, France, Germany and Slovenia show that it is the oxidation of the carbon surface, induced by H_2O_2 , that is the main cause for deactivation in Fe–N–C catalysts.

The researchers prepare a typical Fe–N–C catalyst in which the Fe is mainly present as an atomically dispersed species, coordinated by nitrogen atoms. Spectroscopic characterization suggests that the Fe ion and its immediate environment — together, typically considered to be the active site for ORR — is hardly modified after treatment with H_2O_2 in acidic media. However, the oxygen content of the catalyst, measured by X-ray photoelectron spectroscopy, increases significantly and C=O carbonyl bonds are observed, indicative of surface carbon oxidation. The researchers propose that this oxidation causes changes in the electronic properties of the carbon surrounding the active site, and by extension, the active site itself. This leads to suboptimal O_2 binding at the active sites, lowering their intrinsic activity.

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