

Structural Evolution of Fe Center in Fe-N/C Oxygen Reduction Reaction Electrocatalysts during Silica Coating-Mediated Synthesis

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Iron and nitrogen-doped carbon (Fe-N/C) catalysts are considered the most promising alternative to Pt-based catalysts due to their excellent oxygen reduction reaction (ORR) activity. Recently, we have developed a silica coating-mediated synthesis strategy that can preferentially generate ORR-active Fe-N_x sites while suppressing inactive Fe clusters. However, in previous studies, the origin of preferential formation of Fe-N_x active sites was not clearly revealed. In this work, we investigated how the active Fe center evolves during the steps of silica coating-mediated synthesis by X-ray absorption spectroscopy and ⁵⁷Fe Mössbauer spectroscopy. We confirmed that the bond between the Fe center and Si of the silica is formed when the Fe-N-C composite is coated with silica. As a result, the catalyst prepared with silica coating comprised twice higher density of Fe-N_x sites than the catalyst without the silica coating, leading to twice higher current density.