THE JOURNAL OF PHYSICAL CHEMISTRY C
A JOURNAL OF THE AMERICAN CHEMICAL SOCIETY

pubs.acs.org/JPCC

Article

Structural Evolution of Atomically Dispersed Fe Species in Fe–N/C Catalysts Probed by X-ray Absorption and ^{57}Fe Mössbauer Spectroscopies

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Cite This: *J. Phys. Chem. C* 2021, 125, 11928–11938

ABSTRACT: Iron and nitrogen codoped carbon (Fe–N/C) catalysts are considered the most promising nonprecious metal catalysts for the oxygen reduction reaction (ORR), with their activities approaching those of Pt-based catalysts. Recently, silica-based protective-layer or intermediate layer-assisted synthesis strategies have been developed to preferentially generate catalytically active Fe–N_x sites while suppressing inactive Fe clusters. However, the role of the silica layer in the formation of Fe–N_x sites remains elusive. In this study, we used X-ray absorption and ^{57}Fe Mössbauer spectroscopies to determine the evolution of the structure of Fe-based species during the silica-coating-mediated synthesis. Through X-ray absorption near-edge structure and ^{57}Fe Mössbauer spectroscopy analyses, the formation of iron silicide (Fe–Si) species after silica coating was identified. Peak parameter analyses of ^{57}Fe Mössbauer spectroscopy data suggested that the density of active Fe–N_x species with the Fe–N/C catalyst prepared with silica coating was twice as high as that of the Fe–N/C without silica coating. Consequently, the Fe–N/C catalyst with silica coating exhibited a kinetic current density for the ORR (0.9 V vs reversible hydrogen electrode, RHE) twice as high as that without silica coating.