Enhanced oxygen reduction catalysis in electron beam derived ^{57}Fe–N–C: impact of precursor coordination and iron speciation

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Abstract

^{57}Fe–N–C electrocatalysts were synthesized via ultrasonic treatment and electron beam (e-beam) irradiation, enabling rapid nanoparticle formation under mild conditions. Mössbauer spectroscopy was used to analyze the oxidation and spin states of Fe–N_4 species. The ^{57}Fe-labeled samples prepared using e-beam irradiation exhibited three Fe(II)–N_4 configurations: low-spin (D1), medium-spin (D2), and high-spin (D3), observed at 4.2 K. A higher proportion of low-spin Fe(II)–N_4 sites strongly correlated with improved oxygen reduction reaction (ORR) activity. Notably, e-beam irradiation promoted the selective formation of these catalytically active low-spin sites, offering a scalable and efficient synthesis strategy.