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Observation of Atomically Dispersed Fe, Co, and Ni Sites Supported on a Single MOF-Derived Carbon

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1. Background

Single-atom catalysts (SACs), with their catalytic sites dispersed at the atomic level, have very recently emerged as a new research frontier in catalysis. SACs based on two or more different metals have been demonstrated to present better catalytic behaviors compared with their corresponding single metal-based SACs, possibly owing to the cooperative effects among different single metal sites. In this report, we attempted to synthesize atomically dispersed Fe, Co and Ni atoms supported on a single carbon network by pyrolysis of the metal-organic frameworks with loading of corresponding metal species precursors.

2. Experiments

X-ray absorption fine structure (XAFS) analyses of the hierarchically porous carbon networks with Fe, Co and Ni atoms (FeCoNi-NC) were performed at Aichi Synchrotron Radiation Center (BL11S2/BL5S1). Each powder sample pressed as pallets were analyzed.

3. Results and Discussion

Currently, element-selective XAFS, including X-ray absorption near-edge spectroscopy (XANES) and extended X-ray absorption fine structure (EXAFS) spectroscopy, has been considered as a necessary characterization for the MOF-derived single-atom catalysts, which can demonstrate the atomically dispersed metal atoms via verifying the absence of metal-metal bonds in the spectra.[1] So, XAFS analyses were performed to investigate the local environment of single Fe/Co/Ni atoms in FeCoNi-NC. As shown in Figure 1, the obtained Fe/Co/Ni K-edge XAFS spectra showed that our samples exhibited distinctly different profiles from those of corresponding metal foils (blue lines). The coordination configurations were then determined by the Fourier transformed k^3-weighted $\chi(k)$-function of the corresponding EXAFS spectra in R space. We have confirmed the absence of metallic Fe–Fe/Co-Co/Ni-Ni coordination in FeCoNi-NC, which certified that Fe/Co/Ni sites should be atomically dispersed in the samples. The on-going data analysis is about the structural fitting by virtue of DFT calculation.

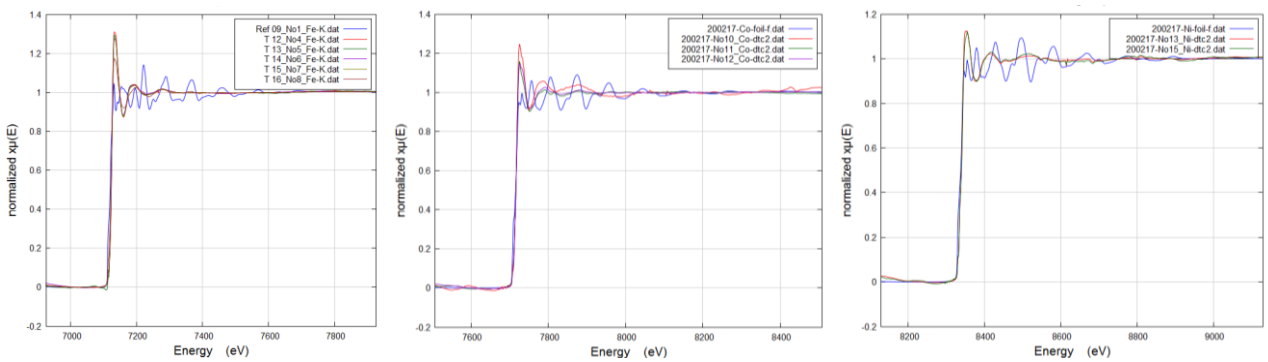

Figure 1. Fe, Co, and Ni K-edge XAFS spectra.

4. References

1. Xu, Q. et al. Puffing Up Energetic Metal-Organic Frameworks to Large Carbon Networks with Hierarchical Porosity and Atomically Dispersed Metal Sites. *Angew. Chem., Int. Ed.* (2019) 58, 1975-1979.