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Tracking the evolution of Ni-based single atom catalysts for the CO2 electroreduction reaction: an operando XAS study assisted by machine learning techniques

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The CO₂ electroreduction process (CO₂RR) is a promising pathway for the abatement of the CO₂ emissions and in the production of valuable chemical feedstock, but suitable catalysts are needed [1]. In the last decade, transition metal-nitrogen-doped carbons have attracted attention as promising electrocatalysts due to their high activity and selectivity for the CO₂RR, which differ significantly from those belonging to bulk or nanostructured materials [2]. In these systems, a certain fraction of N atoms are incorporated in the carbon support forming a binding site for the metal species. These singly-dispersed metal sites are considered to be the active species for the CO₂RR reaction [3,4]. In particular, Ni-based catalysts have exhibited promising CO₂-to-CO conversion activities, even comparable with noble metal catalysts [5]. However, it is worth noting that a large number of different structural motifs can coexist in these systems and evolve during CO₂RR process, making the identification and interpretation of the CO₂RR reaction steps difficult to realize.
With this contribution, we aim to show how to address the over-mentioned issue combining operando XAS measurements with advanced data analysis approaches. We first identify the number of different pure Ni species, their corresponding concentration profiles and XAS spectra using unsupervised machine learning methods, such as the principal component analysis, combined with multivariate curve resolution techniques [6]. Afterwards we deduce the atomistic structures of each identified species by using a XANES fitting procedure enabled by supervised machine learning [7]. The obtained structures are further validated using the available EXAFS data and Reverse Monte Carlo (RMC) simulations [8] which also allows to account for the structural disorder effects in the environment of the identified Ni species.

References

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