

ABSTRACT

Tailoring the Catalytic Properties of Pd Nanoparticles via Plasma-assisted Synthesis

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In the transition towards decarbonized energy solutions, ammonia (NH₃) is emerging as a promising carbon-free fuel and hydrogen carrier. It offers the advantage of large-scale production using well-established infrastructure and can be efficiently stored and transported. Beyond its traditional role in fertilizer production, NH₃ is being explored for direct combustion, fuel cells, and hydrogen storage/release applications. In this context, controlling its oxidation to maximize N₂ selectivity is crucial, as this minimizes NO_x emissions. To this end, we have investigated oxide-supported arrays of Pd NPs, prepared via plasma-assisted micelle nanolithography, as catalysts for NH₃ oxidation, focusing on the role of subsurface species in governing activity and selectivity. Simultaneous operando grazing incidence X-ray diffraction (GIXRD) and grazing incidence small-angle X-ray scattering (GISAXS) were performed at the SixS beamline of Synchrotron SOLEIL (France). We demonstrate that plasma-assisted synthesis allows for tuning the reactivity of Pd NPs and their activity in oxidation reactions. Our experimental results highlight the crucial role of plasma-assisted synthesis in the architectural design of nanomaterials: by precisely altering the synthesis procedure, one can effectively tailor metal-support interactions, ultimately impacting the structure, morphology, and catalytic performance of the nanoparticles.