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Transition Metal Carbodiimide vs. Spinel Matrices for Photocatalytic Water Oxidation

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Abstract : The increasing demand for renewable energy sources and storable fuels underscores the high potential of artificial photosynthesis. The four electron transfer process of water oxidation remains the bottleneck of water splitting, so that special emphasis is placed on the development of economic, stable and efficient water oxidation catalysts (WOCs). Our investigations introduced cobalt carbodiimide CoNCN and its transition metal analogues as WOC types, and further studies are focused on the interaction of different transition metals in the convenient all-nitrogen/carbon matrix. This provides further insights into the nature of the 'true catalyst' for cobalt centers in this non-oxide environment. Water oxidation activity is evaluated with complementary methods, namely photocatalytically using a Ru-dye sensitized standard setup as well as electrocatalytically, via immobilization of the WOCs on glassy carbon electrodes. To further explore the tuning potential of transition metal combinations, complementary investigations were carried out in oxidic spinel WOC matrices with more versatile host options than the carbodiimide framework. The influence of the preparative history on the WOC performance was evaluated with different synthetic methods (e.g. hydrothermally or microwave assisted). Moreover, the growth mechanism of nanoscale Co3O4-spinel as a benchmark WOC was investigated with in-situ PXRD techniques.

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