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Title: Single-entity Electrochemistry Unveils Dynamic Transformation during Tandem Catalysis of Cu₂O and Co₃O₄ for Converting NO₃⁻ to NH₃

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Abstract: Electrochemically converting nitrate to ammonia is an essential and sustainable approach to restoring the globally perturbed nitrogen cycle. The rational design of catalysts for the nitrate reduction reaction (NO3RR) based on a detailed understanding of the reaction mechanism is of high significance. We report a Cu2O+Co3O4 tandem catalyst which enhances the NH3 production rate by approximate to 2.7-fold compared to Co3O4 and approximate to 7.5-fold compared with Cu2O, respectively, however, most importantly, we precisely place single Cu2O and Co3O4 cube-shaped nanoparticles individually and together on carbon nanoelectrodes provide insight into the mechanism of the tandem catalysis. The structural and phase evolution of the individual Cu2O+Co3O4 nanocubes during NO3RR is unveiled using identical location transmission electron microscopy. Combining single-entity electrochemistry with precise nano-placement sheds light on the dynamic transformation of single catalyst particles during tandem catalysis in a direct way.

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