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Title: Splicing the active phases of copper/cobalt-based catalysts achieves high-rate tandem electroreduction of nitrate to ammonia

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Abstract: Electrocatalytic recycling of waste nitrate (NO3-) to valuable ammonia (NH3) at ambient conditions is a green and appealing alternative to the Haber-Bosch process. However, the reaction requires multi-step electron and proton transfer, making it a grand challenge to drive high-rate NH3 synthesis in an energy-efficient way. Herein, we present a design concept of tandem catalysts, which involves coupling intermediate phases of different transition metals, existing at low applied overpotentials, as cooperative active sites that enable cascade NO3--to-NH3 conversion, in turn avoiding the generally encountered scaling relations. We implement the concept by electrochemical transformation of Cu-Co binary sulfides into potential-dependent core-shell Cu/CuOx and Co/CoO phases. Electrochemical evaluation, kinetic studies, and in-situ Raman spectra reveal that the inner Cu/CuOx phases preferentially catalyze NO3- reduction to NO2-, which is rapidly reduced to NH3 at the nearby Co/CoO shell. This unique tandem catalyst system leads to a NO3--to-NH3 Faradaic efficiency of 93.3 +/- 2.1% in a wide range of NO3- concentrations at pH 13, a high NH3 yield rate of 1.17 mmol cm(-2) h(-1) in 0.1 M NO3- at -0.175 V vs. RHE, and a half-cell energy efficiency of similar to 36%, surpassing most previous reports.

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