Electrocatalytic Synergy: Designing Functional Materials for Sustainable Ammonia Production

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The electrocatalytic conversion of nitrate to ammonia presents a sustainable alternative to the conventional, highly energy intensive Haber-Bosch-process, while addressing nitrate pollution at the same time. The challenge lies in finding suitable catalyst materials that show good catalytic activity, selectivity, and stability. Several classes of functional materials have evolved as promising candidates, such as MOFs and MOF-derived materials, high-entropy materials and phosphonate materials. In this work, we present several approaches to improve the electrocatalytic activity towards nitrate reduction by creating synergistic effects through the incorporation of different metals, metal alloying or heteroatom doping. Figure 1 shows one example system of a Co-Fe- based material that is derived from a multi-metal alloy of two isostructural single-metal phosphonate-based MOFs containing Co and Fe, respectively. The synthesis of the single-metal MOFs, as well as the alloying process, were carried out with mechanochemistry, a sustainable synthesis approach. Compared to the single-metal materials, the Co-Fe-material gives rise to a lower overpotential towards the electrochemical nitrate reduction reaction to ammonia. During controlled potential electrolysis, higher current densities were achieved at -1.2 V vs. Ag/AgCl, with an ammonia yield rate of 5398 µg/h*mg_{cat} for the Co-based material while the ammonia yield rate was 3430 µg/h*mg_{cat} for the Fe-based and 2788 µg/h*mg_{cat} for the Co-based material, respectively. These results likely arise from synergistic effects of Co and Fe, underlining the potential of alloy systems for the electrochemical reduction of nitrate to ammonia.

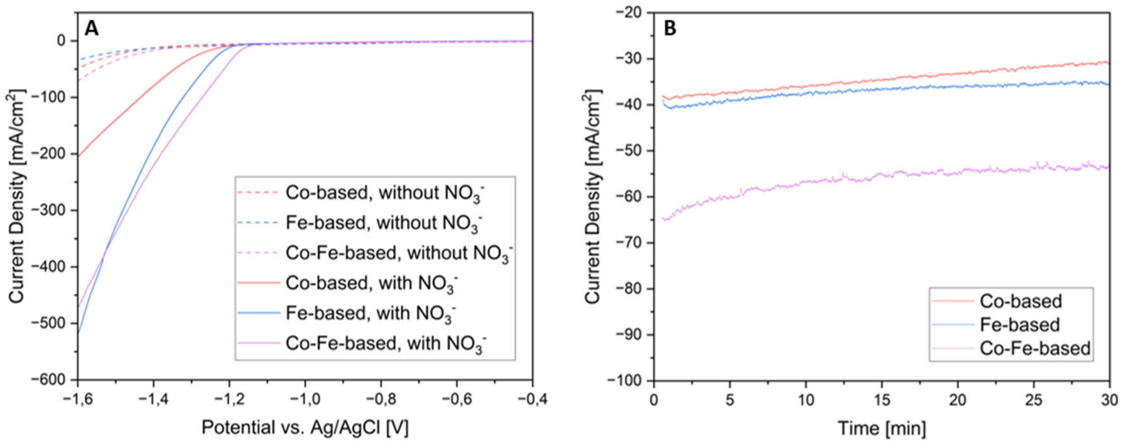

Figure 1: (A) LSV curves and (B) CA curves at -1.2 V vs. Ag/AgCl (B) of Fe-, Co-, and Co-Fe-based MOF-derived materials in 0.1 M K_2SO_4 electrolyte, containing 0.5 M KNO_3. WEs consist of the catalyst material drop casted onto a carbon paper support.