

Stud offers insights for the future design of highly efficient multi-element electrocatalysts

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The (FeCoNiCu)Ox electrocatalyst was first prepared using the rapid Jouleheating method within a short duration at the temperature of 1100 K. XRD results suggest that the (FeCoNiCu)Ox electrocatalyst exhibits a characteristic pattern consistent with NiFe₂O₄, confirming its typical spinel structure. TEM image displays that the (FeCoNiCu)Ox electrocatalyst consists of many nanoparticles in tens of nanometers in diameter. The STEM and EDS elemental mapping images verify the homogeneous distribution of Fe, Co, Ni, Cu and O elements in the nanoparticles. The HR-TEM image of the (FeCoNiCu)Ox electrocatalyst mainly indexed to NiFe₂O₄ (311) and (400), which is in good



agreement with the XRD results. Credit: Science China Press

Professor Zhe Weng and Chunpeng Yang from Tianjin University <u>published</u> a paper titled "Unveiling multi-element synergy in polymetallic oxides for efficient nitrate reduction to ammonia" in the journal *Science China Materials*.

In this study, the (FeCoNiCu)Ox/CeO₂ electrocatalyst for the reduction of NO_3^- to NH_3 was prepared using the rapid Joule-heating method within a short duration. Electrochemical measurements revealed that the (FeCoNiCu)Ox/CeO₂ electrocatalyst exhibited a high Faradaic efficiency for NH_3 exceeding 90% in the potential range of 0 to -0.4 V vs.

RHE, along with a high NH₃ yield rate of 30.3 mg h⁻¹ cm⁻². Moreover, the (FeCoNiCu)Ox/CeO₂ electrocatalyst demonstrated excellent long-term stability for more than 10 h at 200 mA cm⁻².

Through a series of comprehensive experiments, the individual contributions of each element and their synergistic effect have been clearly elucidated.





Several single/binary/ternary MOx electrocatalysts (M = Fe, Co, Ni, Cu, FeCo, FeNi, FeCu, CoCu, FeCoNi and FeNiCu) as control samples were also prepared using the same Joule-heating synthesis method and electrochemical tested. Credit: Science China Press

The NO₃⁻RR performance of the (FeCoNiCu)Ox electrocatalyst was studied under different reduction potentials. The (FeCoNiCu)Ox electrocatalyst demonstrates a high FE_{NH3} of over 90% in the wide potential range of 0 to -0.4 V vs. RHE. However, the r_{NH3} of the (FeCoNiCu)Ox is not as competitive compared to other oxide electrocatalysts reported in previous literatures. TEM and SEM images reveal particle agglomeration, which could affect the dispersion of active components. Previous studies have reported that incorporating CeO₂ as a support in catalysts improves the uniform dispersion of active components and significantly increases the active surface area of the catalysts. Moreover, Ce has a much larger atomic radius than the other elements (Fe, Co, Ni and Cu) and thus Ce cannot incorporate the crystal structure of (FeCoNiCu)Ox even by the rapid Joule-heating method. Accordingly, a (FeCoNiCu)Ox/CeO₂ composite catalyst was further successfully synthesized. Credit: Science China Press

During the 10 h of electrocatalysis at 200 mA cm⁻², the FE_{NH3} and r_{NH3} demonstrate remarkable stability. The XRD pattern reveals that the position and shape of the peaks remain largely unaltered before and after electrocatalysis. The EDS elemental mapping of the (FeCoNiCu)Ox/CeO₂ electrocatalyst after electrocatalysis clearly displays identical element distribution as before electrocatalysis. These results suggests that excellent stability of the (FeCoNiCu)Ox/CeO₂ electrocatalyst. Credit: Science China Press

Specifically, the Cu active sites efficiently reduce NO_3^- to <u>nitrite</u> (NO_2^-) at low overpotential, while the adjacent Co sites facilitate the deep reduction of intermediate NO_2^- .

The Fe and Ni sites play a crucial role in promoting water dissociation to ensure sufficient proton supply. Simultaneously, the CeO₂ component increases the active surface area of the (FeCoNiCu)Ox electrocatalyst and improves the NH₃ yield rate, making it suitable for industrial applications. This work offers significant insights into the design of highly efficient multi-element electrocatalysts.

More information: Yaning Qie et al, Unveiling multi-element synergy in polymetallic oxides for efficient nitrate reduction to ammonia, *Science China Materials* (2024). DOI: 10.1007/s40843-024-3017-4

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