INORGANIC CHEMISTRY FRONTIERS

EDITORIAL

View Article Online
View Journal | View Issue

Cite this: *Inorg. Chem. Front.*, 2024, **11**, 2202

DOI: 10.1039/d4qi90016b

rsc.li/frontiers-inorganic

Electrochemical frontiers in the nitrogen cycle

Xuping Sun,*[a] Jieshan Qiu[b] and Chenghua Sun[c]

[a]*Institute of Fundamental and Frontier Sciences, University of Electronic Science and Technology of China, Chengdu 610054, Sichuan, China. E-mail: xpsun@uestc.edu.cn*
[b]*College of Chemical Engineering, State Key Lab of Chemical Resource Engineering, Beijing University of Chemical Technology, Beijing 100029, China*
[c]*Department of Chemistry and Biotechnology, Swinburne University of Technology, Hawthorn, VIC 3122, Australia*

The essence of nitrogen-cycle electrocatalysis lies not only in the practical applications for ammonia (NH_3) synthesis or environmental remediation but also in its profound scientific underpinnings. This realm offers a unique lens through which to scrutinize the intricate interplay between chemical transformations and the natural world. At its core, it holds the promise of unlocking secrets about Earth's past, present, and future.

Understanding the nitrogen cycle has implications far beyond immediate technological advancements. It is a gateway to comprehending the historical evolution of our planet. Exploring the role of

Xuping Sun

Xuping Sun received his Ph.D. degree from the Changchun Institute of Applied Chemistry (CIAC), Chinese Academy of Sciences in 2006. During 2006–2009, he carried out postdoctoral research studies at Konstanz University, the University of Toronto, and Purdue University. In 2010, he started his independent research career as a full professor at CIAC and then moved to Sichuan University in 2015. In 2018, he joined the University of Electronic Science and Technology of China, where he founded the Research Center of Nanocatalysis & Sensing. Now, he is also a professor at Shandong Normal University. His research mainly focuses on the rational design of functional nanostructures for applications in electrocatalysis, sensing, and biomedicine.

Jieshan Qiu

Jieshan Qiu obtained his Ph.D. degree from the School of Chemical Engineering at Dalian University of Technology (DUT) in 1990. He is now the Cheung-Kong Distinguished Professor of Carbon Science and Chemical Engineering at Beijing University of Chemical Technology, China. His research encompasses both fundamental and applied aspects of carbon materials and science, with a focus on the methodologies for producing carbon materials for energy storage and conversion, catalysis, and environment protection.

Chenghua Sun

Chenghua Sun obtained his PhD from the Institute of Metal Research, Chinese Academy of Science in 2007. Later, he joined the University of Queensland to start postdoctoral research, working in the Royal Institute of Technology, Princeton University and Harvard University. In 2013, he joined Monash University as a faculty lecturer and obtained the prestigious Future Fellow funded by the Australia Research Council. In 2017, he moved to Swinburne University of Technology as an associate professor. His research focuses on computer-aided design of novel catalysts for clean energy and environmental applications.

iron sulfides and their catalytic abilities provides a fascinating perspective on the chemical processes that might have spurred life's origins. The link between iron, sulfur, and nitrogen speciation over geological timescales offers insights into how Earth's conditions might have shaped and influenced these vital elements.

Moreover, the pursuit of sustainable NH_3 synthesis serves as a testament to humanity's quest for energy efficiency and environmental responsibility. The traditional Haber–Bosch process, while revolutionizing agriculture and sustaining human life, has left a considerable carbon footprint. Electrocatalysis, by contrast, presents an avenue towards greener and more energy-efficient alternatives. Its potential to harness renewable energy sources and convert them into valuable chemicals like NH_3 speaks volumes about our aspirations for a sustainable future.

Within this realm, challenges persist. The delicate balance between achieving high yields of NH_3 and suppressing competing reactions such as the hydrogen evolution reaction (HER) requires meticulous engineering of catalysts. The ongoing efforts to design catalysts with specific structural features, be it oxygen vacancies, tailored coordination environments, or precise doping strategies, underscore the intricate dance between materials science and catalysis.

To delve deeper into this transformative narrative, let's explore the intricate fabric woven by various pioneering studies:

Navigated by Rosalie K. Hocking and co-workers (**https://doi.org/10.1039/D3QI01553J**), the exploration of iron sulfides' role in nitrogen species' reactions sheds light on their catalytic and direct reductant capabilities, offering insights into Earth's historical nitrogen transformations. Sadeeq Ullah and co-workers (**https://doi.org/10.1039/D3QI01536J**) discussed the challenges in electrochemical nitrate (NO_3^-) reduction for NH_3 synthesis, highlighting strategies like oxygen vacancy incorporation in catalysts to enhance efficiency and selectivity. Zhong-Yong Yuan and co-workers (**https://doi.org/10.1039/D3QI01113E**) focused on first-row transition metal-based catalysts for NO_3^-/NO_2^- reduction, emphasizing the potential of Cu-, Fe-, Co-, Ni-, and Ti-based electrocatalysts for efficient NH_3 production. Wenda Chen and co-workers (**https://doi.org/10.1039/D3QI00732D**) detailed the suitability of Ti-based nanomaterials, especially Ti-based metallic and TiO_2 nanomaterials, for NO_3^- reduction, highlighting recent advancements and future challenges. Dongpeng Yan and co-workers (**https://doi.org/10.1039/D3QI00683B**) reviewed progress in photoelectrocatalytic NH_3 synthesis, emphasizing strategies like vacancy engineering and heterojunction construction for better NH_3 synthesis mechanisms. Hao Li, Fei Wang, and co-workers (**https://doi.org/10.1039/D3QI00148B**) explored the potential of thermal reduction in NO_3^-/NO_2^- reduction for water treatment, discussing catalyst synthesis, product selectivity, and the viability of forming NH_3. Luchao Yue, Xuping Sun, and co-workers (**https://doi.org/10.1039/D3QI00554B**) detailed the electrochemical NO_3^- reduction progress, exploring diverse catalysts like noble-metal-based materials and transition-metal-based catalysts, emphasizing strategies for high efficiency. Debabrata Chatterjee and co-workers (**https://doi.org/10.1039/D3QI00199G**) explored the role of Ru(edta) complexes in nitrogen cycle electrochemical transformations, elucidating their mechanistic impact in nitrogen fixation and denitrification. Hongda Li and co-workers (**https://doi.org/10.1039/D3QI00895A**) introduced a novel bionic FeV-cofactor system for photocatalytic N_2 reduction, demonstrating enhanced NH_3 yield through Fe-doped $BiVO_4$ decorated with 2D black phosphorus. Ji Zhang and co-workers (**https://doi.org/10.1039/D3QI00517H**) presented the theoretical design of dual metal catalysts for N_2 reduction, offering insights into their efficiency, selectivity, and suppression of the HER during N_2 reduction. Aijun Du and co-workers (**https://doi.org/10.1039/D3QI00798G**) investigated Fe-alloyed bimetallics for electrochemical N_2 reduction, unveiling their enhanced activity and low HER compared to pristine group-IVA elements. Jianping Yang and co-workers (**https://doi.org/10.1039/D3QI00793F**) explored self-supported Fe/support catalysts synthesized through hydrothermal and thermal reduction strategies, showcasing their improved performance in electrochemical NO_3^- reduction. Yi Feng and co-workers (**https://doi.org/10.1039/D3QI00795B**) demonstrated NiFe LDH nanoarrays on Co_3O_4 nanosheets for superior NO_2^- reduction and oxygen evolution, showcasing high efficiency and stability in NH_3 electrosynthesis. Jinmeng Cai, Siyu Lu, and co-workers (**https://doi.org/10.1039/D3QI00865G**) discussed spin density modulation using carbon dot materials to enhance Co_3O_4's electrocatalytic NO_3^- reduction, resulting in increased NH_4^+ faradaic efficiency and stability. Fengling Zhou and Chenghua Sun (**https://doi.org/10.1039/D3QI00568B**) investigated the high activity of electrodeposited Ni/Ru hydroxide hybrids for NO_3^- to NH_3 conversion, unveiling the role of surface-oxidized nickel layers in catalytic enhancement. Ke Chu and co-workers (**https://doi.org/10.1039/D3QI00268C**) unveiled Sb_2S_3's role as a catalyst in efficient NO-to-NH_3 conversion, highlighting the unique functionality of Sb_{AIU} sites for high selectivity and activity. Yuting Sun and coworkers (**https://doi.org/10.1039/D3QI00225J**) proposed the β-$PdBi_2$ monolayer as a highly efficient catalyst for electrochemical NO reduction to NH_3, emphasizing its low limiting potential and high efficiency. Longcheng Zhang and co-workers (**https://doi.org/10.1039/D3QI00209H**) detailed the high efficiency of a RuO_2 nanoparticle-decorated TiO_2 nanobelt array in synthesizing NO_3^- from NO, showcasing a significant yield and electrochemical insights. Guohui Wang and co-workers (**https://doi.org/10.1039/D2QI02757G**) introduced La-doped VS_{2-x} for electrochemical NO_3^- reduction to NH_3, highlighting its high selectivity and efficiency, which are promising for NH_3 synthesis. Xiaoya Fan and co-workers (**https://doi.org/10.1039/D2QI02409H**) demonstrated the efficacy of Ag@TiO_2 in the electrosynthesis of NH_3 from NO_2^-, emphasizing its high yield and efficiency, and indicating its potential in wastewater treatment.

These pioneering studies underscore a collective dedication to unravelling the intricacies of nitrogen-cycle electrocatalysis, steering the course toward sustainable NH_3 synthesis. The amalgamation of diverse catalysts, electrochemical pathways, and novel materials not only holds promise for mitigating environmental burdens, but also heralds a new era of sustainable nitrogen transformations, underscoring humanity's commitment to a greener future.