Direct electroconversion of air to nitric acid realized under mild conditions

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A research group led by Prof. Deng Dehui from the Dalian Institute of Chemical Physics (DICP) of the Chinese Academy of Sciences (CAS) has realized direct conversion of air to (nitric acid) HNO₃ under ambient conditions through a hydroxyl radical (·OH)-mediated hetero-homogeneous electro-chemical route.

Schematic illustration of the proposed reaction path for direct electroconversion of air to HNO₃. Credit: Nature Synthesis (2023). DOI: 10.1038/s44160-023-00399-z
In the route, the N\textsubscript{2} was oxidized to HNO\textsubscript{3} by employing heterogeneous electroreduction of O\textsubscript{2} to H\textsubscript{2}O\textsubscript{2} on a graphite rod catalyst combined with homogeneous Fe\textsuperscript{2+}-induced dissociation of the in situ generated H\textsubscript{2}O\textsubscript{2} to OH\textsuperscript{-} as reactive oxidation species.

The study was published in *Nature Synthesis* on Sept. 25.

HNO\textsubscript{3}, as an important chemical building block, is produced industrially from the oxidation of ammonia via the Ostwald process, which requires high temperatures above 800°C. In this process, the feed ammonia gas is industrially synthesized via the energy-intensive Haber-Bosch method typically operating at high temperatures (400 ~ 500°C) and pressures (20 ~ 50 MPa).

Direct conversion of dinitrogen (N\textsubscript{2}) and oxygen (O\textsubscript{2}) in the air to HNO\textsubscript{3} under mild conditions is a promising and sustainable route for the production of HNO\textsubscript{3}. However, the thermodynamic and kinetic restrictions of this reaction, as well as the high stability of N\textsubscript{2} and the low activity of O\textsubscript{2}, make it challenging to achieve co-activation and efficient conversion of the two molecules in the air toward HNO\textsubscript{3} synthesis.

In this study, the researchers developed a hetero-homogeneous electrochemical route for the direct conversion of air to HNO\textsubscript{3} at the cathode compartment. At the cathodic potential of 0 V vs. RHE, the faradaic efficiency of nitric acid was as high as 25.37\%, and the selectivity was over 99\%, which was superior than the previously reported electrocatalytic oxidation of N\textsubscript{2} at the anode with high potentials above 1.23 V vs. RHE.

Multiple in-situ spectroscopic characterizations and theoretical calculations revealed that ·OH produced from Fe\textsuperscript{2+}-induced dissociation of in-situ generated H\textsubscript{2}O\textsubscript{2} could efficiently activate N\textsubscript{2}. This delivers a high HNO\textsubscript{3} productivity of 141.83 μmol·h\textsuperscript{-1}·g\textsubscript{Fe}\textsuperscript{-1}, which is 225 times that
of directly using H₂O₂ as the oxidant.

"This process for HNO₃ synthesis from N₂ and O₂ effectively avoids the traditional high-temperature and high-pressure reaction conditions, and provides a new route for the co-activation and efficient conversion of N₂ and O₂ under mild conditions," said Prof. Deng.


Provided by Chinese Academy of Sciences

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