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Ammonia Recovery from Wastewater using Integrated Tunable Electrochemical Systems

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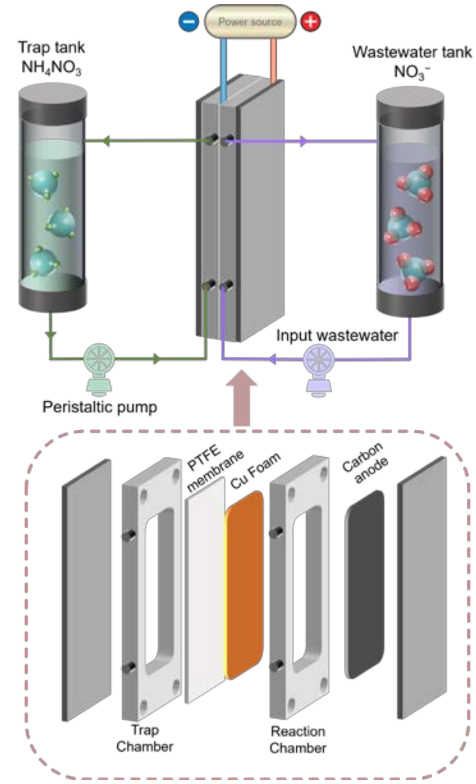
The Haber-Bosch process has long been employed to produce industrial ammonium for fertilization, which consumes fossil fuels to drive the thermodynamically unfavorable reaction between nitrogen (N_2) and hydrogen (H_2) at high pressures and temperatures. Meanwhile, the extensive use of fertilizers and industrial waste streams causes eutrophic water pollution (e.g., with high nitrate/nitrite content). Conventional biological nitrogen removal involves energy intensive nitrification and denitrification processes (~11.7−12.5 $\mathrm{kWh \cdot kg\text{-}N^{-1}}$) that eventually converts all nitrogen species into nitrogen gas. Clearly, shortening the nitrogen removal processes by converting wastewater nitrate into NH_3 can potentially reduce the energy and carbon footprints and enable nutrient recovery and reuse/recycle. This study demonstrates for the first prototype electrified membrane system for synchronizing electrochemical NO_3^- reduction and upcycling to NH_3 without any external chemical addition. Unlike those widely reported sacrificial half-reactions in the electrochemical nitrogen recovery, a paired electrolysis was employed to enable proton and hydrogen transfer between cathodic and anodic chambers to minimize energy consumption and avoid acid/base use for NH_3 capture and conversion. This study will demonstrate an electrified membrane made of a CuO@Cu foam and a polytetrafluoroethylene (PTFE) membrane for reducing NO_3^- to NH_3 and upcycling NH_3 into $(NH_4)_2SO_4$, a liquid fertilizer readily for use. A paired electrolysis process was achieved under a partial current density of 63.8±4.4 $\mathrm{mA \cdot cm^{-2}}$ on the cathodic membrane, which removed 99.9% NO_3^- in the feed (150 mM NO_3^-) after 5 h operation with a NH_3 recovery rate of 99.5%. A recovery rate and energy consumption of 3100±91 $\mathrm{g\text{-}(NH_4)_2SO_4 \cdot m^{-2} \cdot d^{-1}}$ and 21.8±3.8 $\mathrm{kWh \cdot kg^{-1}\text{-}(NH_4)_2SO_4}$ almost outcompetes the industrial ammonia production cost in the Haber-Bosch process. Density functional theory (DFT) calculations unraveled that the in situ electrochemical conversion of Cu^{2+} into Cu^{1+} provides high dynamic active species for NO_3^- reduction to NH_3. Additionally, a pilot-scale electrocatalytic reactor with an anode contact area of 0.16 m^2 has been fabricated and tested using real wastewater (e.g., RO retentate, ion exchange backwash brine, and ammonia-laden landfill leachate) to recover ammonia. Key results indicate that when 1 gallon of RO retentate (obtained from Yuma Desalination Plant, Arizona) containing 10 mM NO_3^- and 10 mM NH_4^+ is fed into the system, approximately 7.87 mM of NH_3 is converted and successfully recovered after 100 minutes of operation. Additionally, testing with wastewater of higher NO_3^- concentration (~211 mM) demonstrated the system's effectiveness, achieving the removal of ~47 mM NO_3^- during a 120-minute operation, highlighting its robust performance. Overall, this research highlights a promising sustainable approach to ammonia recovery that not only reduces energy consumption and carbon emissions but also enables nutrient recycling, offering a viable alternative to conventional industrial processes like the Haber-Bosch.

Keywords:


Power source
Trap tank
NH_4NO_3
Wastewater tank
NO_3^-
Input wastewater
Peristaltic pump
PTFE membrane
Cu Foam
Carbon anode
Trap Chamber
Reaction Chamber