Text S1. Electrochemical measurements.

To prepare the catalyst ink, 5 mg of catalyst was dispersed in a mixture of 375 μL of ethanol, 125 μL of H_2O, and 20 μL of Nafion solution (5 wt%), followed by sonication for 1 h until a homogeneous ink was formed. Subsequently, 100 μL of the well-dispersed catalyst ink was drop-cast onto a 1 cm^2 carbon paper and dried at room temperature to serve as the working electrode. Electrochemical tests were carried out in a catholyte of 0.5 M KNO_3 + 1 M KOH (Ar-purged for 30 min). LSV was measured at a scan rate of 10 mV/s with stirring at 400 rpm. EIS was conducted with an AC voltage amplitude of 5 mV over a frequency range from 100,000 Hz to 0.01 Hz. The ECSA was evaluated from CV curves measured in the non-Faradaic region at different scan rates and calculated by dividing the electrochemical double-layer capacitance (C_{dl}) by the specific capacitance (Cs) of the sample. Chronoamperometry measurements were performed at various potentials (−0.2, −0.3, −0.4, −0.5, and −0.6 V vs. RHE) for 2 h under continuous stirring (400 rpm). The electrolyte was collected after electrolysis for product analysis. Calibration curves for nitrite and ammonia were generated by correlating UV-visible absorbance (540/420 nm) with standard solution concentrations. The measured potentials were converted to the RHE scale using the following equation:

$$E_{RHE} = E_{(Hg/HgO)} + 0.0591 \times pH + 0.098$$

The Faradaic efficiency for NH_3 production is defined as the ratio of the charge used for NH_3 formation to the total charge passed during electrolysis. It is calculated using the following equation:

$$FE_{NH_3}(\%) = \frac{n \times F \times C_{NH_3} \times V}{M \times Q} \times 100\%$$

Where n is number of electrons transferred for NH_3 production from NO_3^- (n = 8, based on the reaction: $NO_3^- + 6H_2O + 8e^- \rightarrow NH_3 + 9OH^-$). F is the Faraday constant (F = 96485 C mol^{-1}). C_{NH3} is the measured concentration of NH_3 in the electrolyte after electrolysis (mol L^{-1}). V is the volume of the catholyte (V = 0.05 L). Q is the total charge passed during electrolysis (C). M is the molar mass of NH_3 (M = 17 g mol^{-1}).

For the quantification of by-products, the Faradaic efficiency for NO_2^- formation is calculated using the same formula with n = 2 ($NO_3^- + H_2O + 2e^- \rightarrow NO_2^- + 2OH^-$) and M = 46 g mol^{-1}.

$$FE_{NO_2^-}(\%) = \frac{n \times F \times C_{NO_2^-} \times V}{M \times Q} \times 100\%$$

The NH_3 yield rate (Y_{NH3}) is defined as the amount of NH_3 produced per unit time per unit mass of catalyst. It is calculated using the following equation:

$$Y_{NH_3} = \frac{C_{NH_3} \times V}{t \times m_{cat}}$$

Where C_{NH3} is the measured concentration of NH_3 in the electrolyte after electrolysis (mol L^{-1}). V is the volume of the catholyte (V = 0.05 L). t is the electrolysis time (h). m_{cat} is the mass of catalyst loaded on the working electrode (mg cm^{-2}).