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## **Electronic Supplementary Information**

## Electrocatalytic Selectivity for Nitrogen Reduction vs. Hydrogen Evolution: A Comparison of Vanadium and Cobalt Oxynitrides at Different pH Values

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**Figure S1:** CoN films before (red) and after (green) ambient exposure. After ambient exposure, in-situ atomic ratio is CoO<sub>0.7</sub>O<sub>0.3</sub>.



**Figure S2:** Grazing (red) and normal (green) of (a) Co 2p (b) O 1s (c) N 1s spectra of  $CoO_{0.8}N_{0.2}$  after deposition and subsequent ambient exposure.



**Figure S3:** XPS of  $CoO_xN_y$  film before/after EC in Ar-saturated solution. This shows that lattice N is lost during cathodic polarization.



**Figure S4:** VON films before (red) and after (green) ambient exposure. After ambient exposure, the insitu atomic ratio is  $VO_{0.4}N_{0.6}$ .



**Figure S5:**  $VO_{0.4}N_{0.6}$  film showing that there is no evolution of hydrogen during electrolysis in basic solution. The same was observed in the neutral solution.