A rigorous electrochemical ammonia synthesis protocol with quantitative isotope measurements

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The electrochemical synthesis of ammonia from nitrogen under mild conditions and using renewable electricity is in principle an attractive alternative to the demanding, energy-intensive Haber-Bosch process, which dominates industrial ammonia production. However, the electrochemical alternative faces considerable scientific and technical challenges and most experimental studies reported thus far achieve only low selectivities and conversions. In fact, the amount of ammonia produced is usually so small that it is difficult to firmly attribute it to electrochemical nitrogen fixation and exclude contamination due to ammonia that is either present in air, human breath or ion-conducting membranes, or generated from labile nitrogen-containing compounds (for example, nitrates, amines, nitrites and nitrogen oxides) that are typically present in the nitrogen gas stream, in the atmosphere or even the catalyst itself. Although these many and varied sources of potential experimental artefacts are beginning to be recognized and dealt with, concerted efforts to develop effective electrochemical nitrogen reduction processes would benefit from benchmarking protocols for the reaction and from a standardized set of control experiments to identify and then eliminate or quantify contamination sources. Here we put forward such a rigorous procedure that, by making essential use of $^{15}\text{N}_2$, allows us to reliably detect and quantify the electroreduction of $\text{N}_2$ to $\text{NH}_3$. We demonstrate experimentally the significance of various sources of contamination and show how to remove labile nitrogen-containing compounds present in the $\text{N}_2$ gas and how to perform quantitative isotope measurements with cycling of $^{15}\text{N}_2$ gas to reduce both contamination and the cost of isotope measurements. Following this protocol, we obtain negative results when using the most promising pure metal catalysts in aqueous media, and successfully confirm and quantify ammonia synthesis using lithium electrodeposition in tetrahydrofuran.

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