A rigorous electrochemical ammonia synthesis protocol with quantitative isotope measurements

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.

General information
Publication status: Published
Organisations: Department of Physics, Experimental Surface and Nanomaterials Physics, Department of Chemistry, Stanford University, Imperial College London
Corresponding author: Chorkendorff, I.
Number of pages: 5
Pages: 504-508
Publication date: 2019
Peer-reviewed: Yes

Publication information
Journal: Nature
Volume: 570
ISSN (Print): 0028-0836
Ratings:
BFI (2019): BFI-level 3
Web of Science (2019): Indexed yes
Original language: English
DOIs:
10.1038/s41586-019-1260-x
Source: FindIt
Source-ID: 2447743416
Research output: Contribution to journal › Journal article – Annual report year: 2019 › Research › peer-review