Advanced Reactor Design for Ammonia Production from Electrochemical Nitrogen and Nitrate Reduction

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Abstract:

Motivated by the increasing demand for flexible and sustainable routes of ammonia (NH_3) production, the electrochemical nitrogen (N_2) and nitrate reduction reaction (NRR and NO3RR) have attracted intense research interest in the past few years[1,2]. Compared to the centralized Haber-Bosch process that operates at elevated temperature and pressure, the electrochemical pathway features mild operating conditions but high input energy density, allowing for distributed and on-site generation of NH_3 with water as the proton source, thereby reducing the transportation and storage costs of NH_3 and H_2[3]. Besides N_2 which is highly abundant in the atmosphere, nitrate-N exists widely in agricultural and industrial wastewaters, and its presence has raised severe concerns due to its known impacts on the environment and human health[4,5]. In this regard, NO3RR provides a promising strategy of simultaneously removing the harmful nitrate-N and generating NH_3 as a useful product from those wastewater streams.

While research activities on both NRR and NO3RR are blooming with substantial progress in the field of electrocatalysis, some major challenges remain unnoticed or unresolved so far. Due to the wide existence of reactive N-containing species in laboratory environments, the source of NH_3 in NRR measurements is sometimes elusive and requires rigorous examination by control experiments with costly $^{15}N_2$[6,7]. On the other hand, while the electro-reduction of nitrate is much more facile, additional costs arising from the enrichment and purification of nitrate in contaminated waste resources have challenged the practical feasibility of NO3RR both technically and economically[2].

In this talk, we will present our latest research progress as part of the solutions to these challenges in state-of-the-art NRR and NO3RR studies, from the perspective of reactor design. By taking advantage of the prior developments in $^{15}N_2$ control experiments, here we suggest an improved $^{15}N_2$ circulation system that is effective and affordable for NRR research, allowing for more accurate and economized quantitative assessment of NH_3 origins, so that false positives and subtle catalytic activities can be identified more reliably. For NO3RR, we developed a compact reactor system for rapid and efficient electrochemical conversion of nitrate to NH_3 from real nitrate-containing waste sources, accompanied by the concurrent separation and enrichment of the produced NH_3 in a trapping solution to yield pure ammonium compounds. Our work highlights the importance of advanced reactor design in N-related electrochemistry research, which will facilitate the transformation of the current N-centric chemical industries towards a sustainable future.

References

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