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PROCEEDINGS

Green Ammonia-Mediated CO_2 Capture and Conversion to Valuable Chemicals

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ABSTRACT

Direct electrochemical conversion of CO_2 capture solutions (instead of gaseous CO_2) into valuable chemicals can circumvent the energy-intensive CO_2 regeneration and pressurization steps. While commonly used CO_2 capture agents include alkali and amine solutions, ammonia has been rarely investigated. In another aspect, mismanagement of reactive nitrogen (Nr) in waste has emerged as a major problem in water pollution to our ecosystems, causing severe eutrophication and health concerns. Sustainably recovering Nr [such as nitrate (NO_3^-)-N] and converting it into green ammonia (NH_3) could mitigate the environmental impacts of Nr and reduce the NH_3 demand from the carbon-intensive Haber-Bosch process, as well as a possible CO_2 capture agent due to its alkaline nature. In this talk, we will present our rencet research on integration of electrodialysis and electrocatalysis for ammonia synthesis from dilute waste Nr sources [1], and green ammonia-mediated CO_2 capture (to ammonium bicarbonate, NH_4HCO_3) and subsequent reduction to ammonium formate (NH_4HCO_2) as a new approach to CO_2 capture and utilization (CCU) [2]. We have demonstrated a record-high NO_3^--to-NH_3 performance in a scalable, versatile, and cost-effective membrane-free alkaline electrolyzer (MFAEL): an unprecedented NH_3 partial current density of 4.22 ± 0.25 A cm^{-2} with a faradaic efficiency of 84.5 ± 4.9%. We also discovered that an ammonium bicarbonate (NH_4HCO_3)-fed electrolyzer with an anion exchange membrane (AEM) outperforms the state-of-the-art $KHCO_3$ electrolyzer with a bipolar membrane (BPM) owing to its favorable thermal decomposition property, which allows for a 3-fold increase in the *in situ* CO_2 concentration, a maximum 23% increase in formate faradaic efficiency, and a 35% reduction in cell voltage by substituting BPM with the AEM. Our integrated process by combining NH_4HCO_3 electrolysis with CO_2 capturing by on-site generated green ammonia from the electro-reduction of nitrate in MFAEL has shown a remarkable 99.8% utilization of CO_2 capturing agent. Our recent progress in this direction will also be briefly presented. Such a multi-purpose process may offer a sustainable route for the simultaneous removal of N_r wastes and streamlined CO_2 capturing and upgrading to valuable chemicals.

KEYWORDS

CO_2 capture; CO_2 utilization; electrocatalysis; electrochemical engineering

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