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Graphical Abstract: A novel ammonia synthesis approach via nitrates intermediaries. Here we use rotating gliding arc discharge (RGA) and dielectric-barrier discharge (DBD) to produce nitrates in water, the nitrate solution used as an intermediary for ammonium's electrochemical synthesis.

Abstract

Ammonia is one of the largest produced chemicals due to its application in agriculture mainly as fertilizer and a crucial building block in the pharmaceutical and chemical industry [1]. Haber-Bosch process with natural gas is used to produce ca. 150 million tons of ammonia annually and the production of 1 kg ammonia with natural gas results in 1.5 kg of CO₂ production [2]. Therefore sustainable routes towards green ammonia production are actively investigated [3]. The use of renewable energy to facilitate the Haber-Bosch process so-called green process has the potential to reduce CO₂ emission however, it is costly and requires a large plant infrastructure [4]. So, electrification of ammonia synthesis with plasma or with electrocatalysis technology are other options to produce small-scale, decentralized, on-demand green ammonia [5]–[7].

In our recent work, we have introduced an energyefficient path for NO_x production by using air discharge in RGA. Which is much lower than the previously reported value of plasma-assisted atmospheric nitrogen fixation and is close to that of the Haber–Bosch process [8]. We have utilized this energy-efficient NO_x for nitrite and nitrates production by introducing ozone gas with water. The ozone gas was produced with DBD discharge and mixed with NOx and water to produce water-soluble nitrite and nitrates [9]. That can be utilized for ammonia production with an electrochemical reduction [10].

In this work, we propose a novel process to combine rotating gliding arc discharge (RGA) and dielectricbarrier discharge (DBD) with a water shower to produce highly water-soluble nitrite and nitrates. This nitrite and nitrates containing solution will be used for electrochemical conversion into ammonium (a watersoluble form of ammonia). This novel process will provide a breakthrough in the transition toward sustainable and environmentally friendly NH₃ production.

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