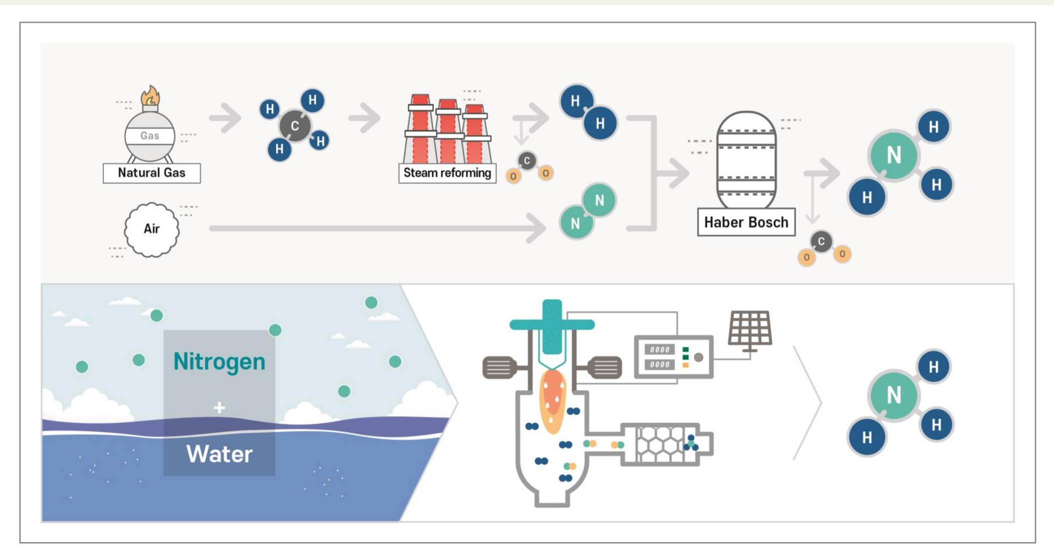
Investigation of Plasma-Based Nitrogen Fixation Using Water in Nitrogen Arc Discharge for Ammonia Production

Muzammil Iqbal[1], Majeed Muhammad[2], Dae Hoon Lee[2], Tom Butterworth[1], Gerard van Rooij[1]
[1]Circular Chemical Engineering, Faculty of Science and Engineering, Maastricht University, Netherlands
[2]Department of Plasma Engineering, Korea Institute of Machinery and Materials, South Korea

Introduction

- The global demand for clean and sustainable energy has led to extensive research into new energy production methods.
- One promising approach is the production of green fuels such as ammonia from base molecules like water and nitrogen, using renewable electricity.
- Non-thermal atmospheric pressure plasma-based NH_3 synthesis has shown significant potential for the production of carbon-free fertilizer using renewable electricity.
- Our research focused on developing an atmospheric pressure rotating gliding arc discharge reactor in conjunction with a catalyst to achieve a higher ammonia production rate.
- We used a two-step ammonia production process, including the facile production of NOx and hydrogen from nitrogen discharge in water, and the highly selective catalytic reduction of NOx to ammonia in the presence of hydrogen.
- The plasma technique we studied yielded an ammonia concentration of approximately 0.84%, with a selectivity of 95% and a production rate of 120 µmol/s by integrating a catalytic reduction system with plasma.
- Our results represent a significant advancement in the transition towards sustainable and environmentally friendly ammonia production, with a 300-400-fold increase in ammonia yield compared to previous studies on ammonia synthesis from plasma discharge.



Experimental Details

- Rotating gliding arc (RGA) plasma reactor used in study.
- SUS304 stainless-steel body and oxygen-free high thermal conductivity Cu electrode utilized.
- Pure nitrogen gas (N_2) at various flow rates (10–100 L/min) provided swirling flow to rotate the arc.
- Water (H_2O) with various flow rates (5–100 mL/min) introduced to reactor through two holes.
- Water film interacted with arc without destabilizing plasma discharge.
- Presence of water film lowered temperature, reducing reactor erosion.
- Nitrogen discharge around water produced NOx, H_2, and heat for catalytic reduction of NOx to ammonia.
- Arc discharge driven by AC power source with high-voltage transformer (up to 6 kV and 20 A at 20 kHz).
- Cogenerated NO and H_2 injected into catalyst, directly connected to plasma reactor.
- Catalyst directly heated by injecting hot gas; reaction temperature controlled at ~110 °C by changing flow rate (3–8 L/min) in catalyst system.
- Exhaust gas (including all NOx species and NH_3) analyzed using FTIR (Midac I400-E)
- AutoQuant Pro software used for quantitative analysis of species concentrations in parts per million (ppm)

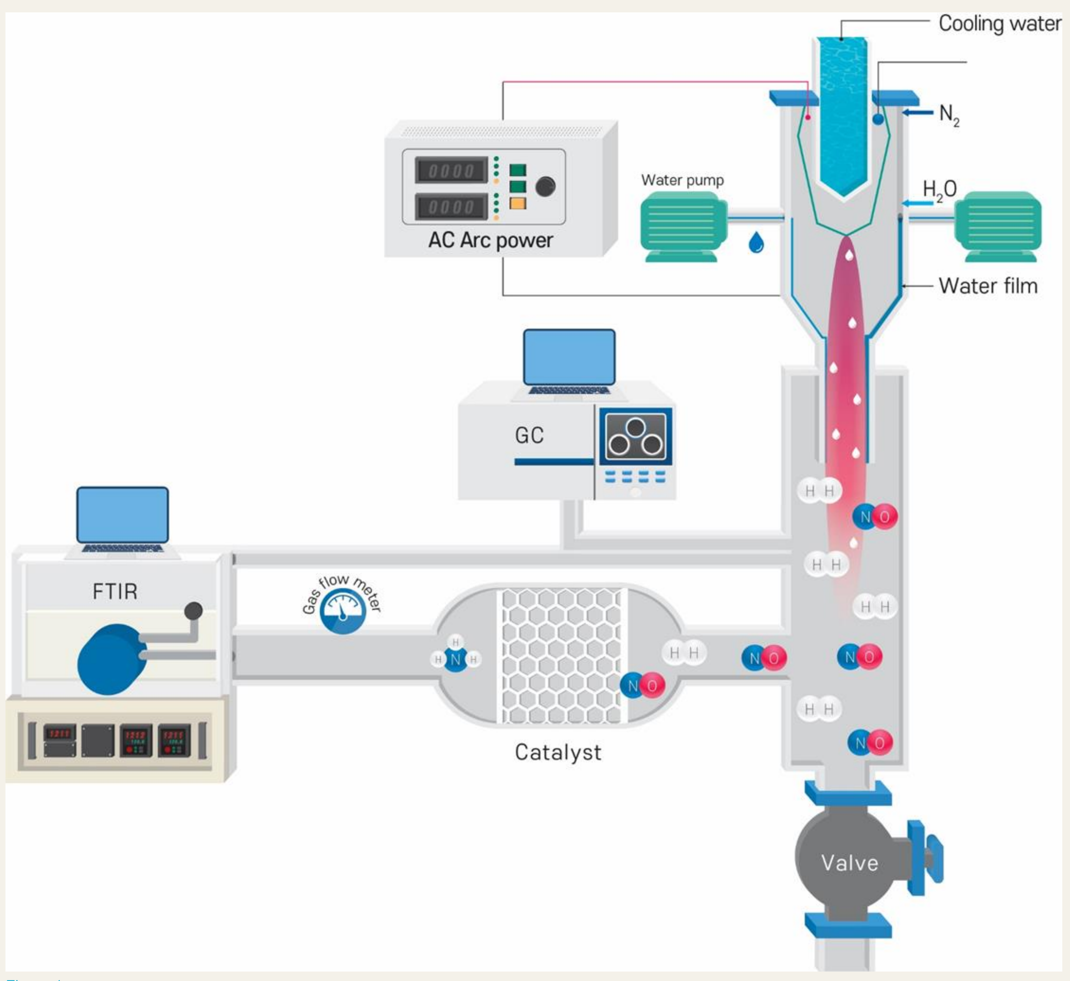


Figure 1

Results

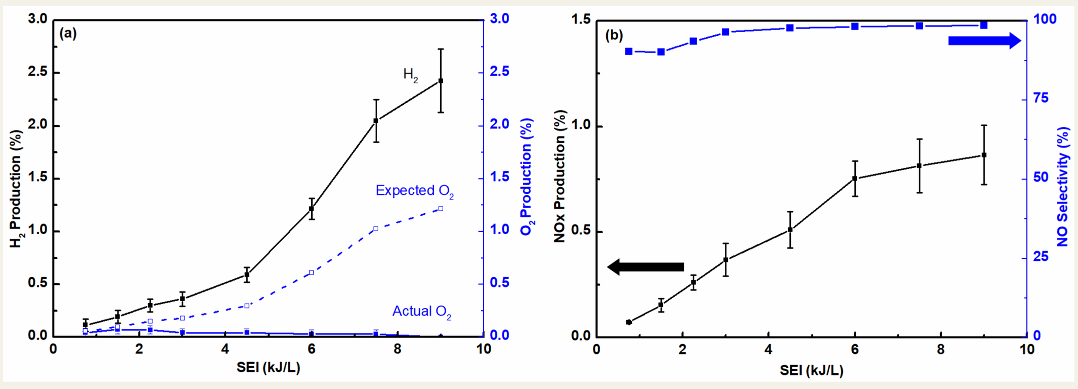


Figure 2

- Figure 2 displays H_2, O_2, and NOx production rates (%) during water splitting in the nitrogen discharge as a function of specific energy input (SEI).
- High SEI results in the disappearance of O_2, indicating the occurrence of secondary reactions involving O and N atoms.
- FTIR and NOx-emissions monitoring system confirmed the presence of these secondary reactions.

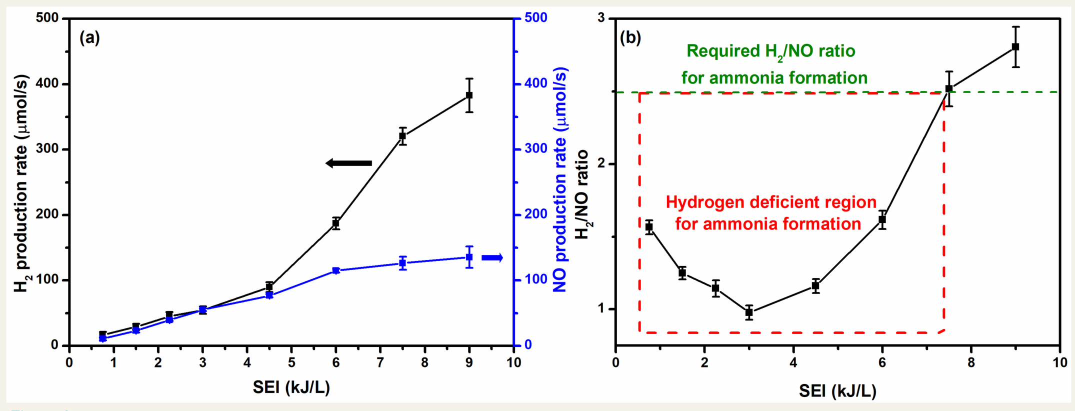


Figure 3

- Figure 3(a) displays the H_2 and NO production rates according to the SEI.
- H_2 and NO production rates increased until an SEI of 3 kJ/L was reached.
- Further increase in the SEI resulted in a substantial increase in the H_2 formation rate and a slight increase in the NO formation rate.
- This change in the H_2/NO ratio was beneficial for NH_3 production.
- Figure 3(b) shows the H_2/NO ratio on function of SEI..

$$2NO + 5H_2 \rightarrow 2NH_3 + 2H_2O$$

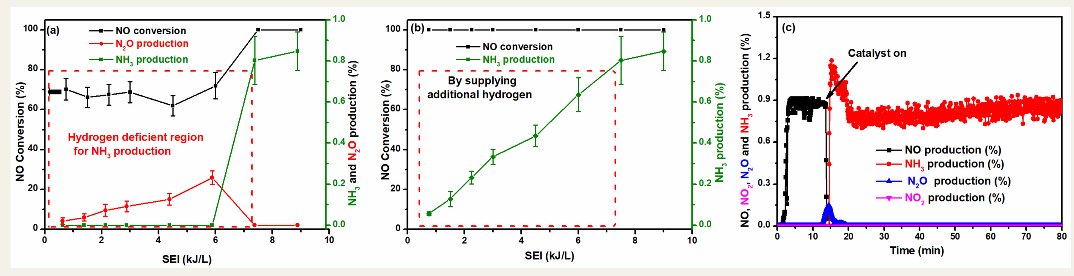


Figure 4

- Experiments were conducted to confirm the effect of the H_2/NO ratio on NH_3 selectivity in the reactor system as shown in (Figure 4a).
- Additional H_2 was injected to compensate the H_2/NO ratio at 2.5 under an SEI of < 7.5 kJ/L (Figure 4b).
- The supply of sufficient H_2, compared to the supply of sufficient NO, resulted in a reduction of N_2O production to 0.
- Figure 4c shows NOx and NH_3 production at ~9 kJ/L as functions of time based on continuous measurements with FTIR.

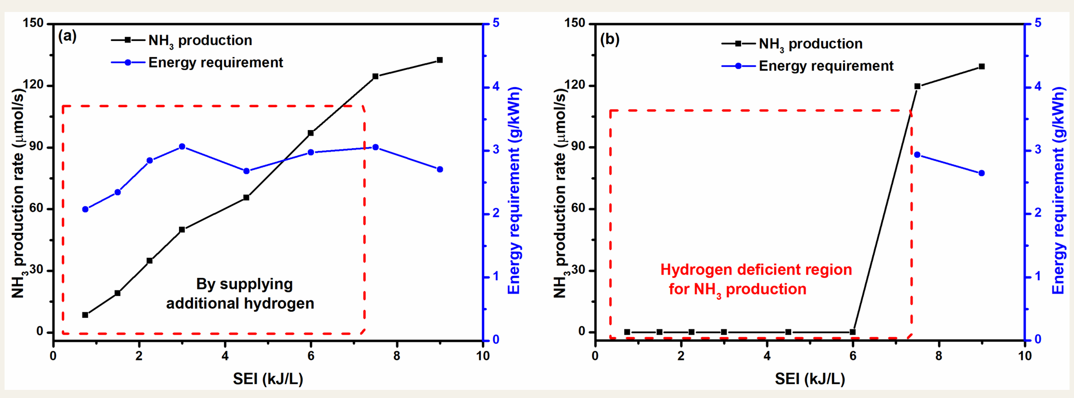


Figure 5

- Figure 5 shows NH_3 production rate and energy requirements (a) upon supplying additional H_2 in the H_2 deficient region and (b) without supplying additional H2 as functions of SEI.
- The rate of NH_3 production was 120 µmol/s (7,348 mg/h) without the injection of additional H_2 from outside the reactor system.

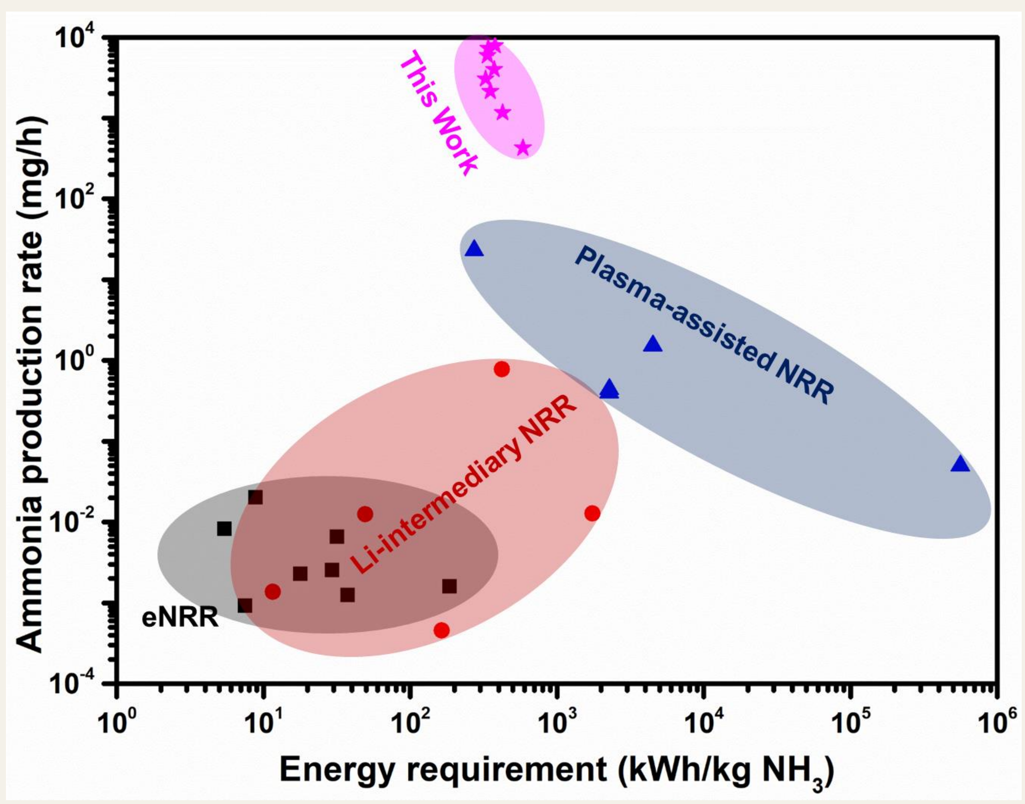


Figure 6

NH_3 production rate and energy consumption compared with recently reported state-of-the-art results.

Reaction Mechanism

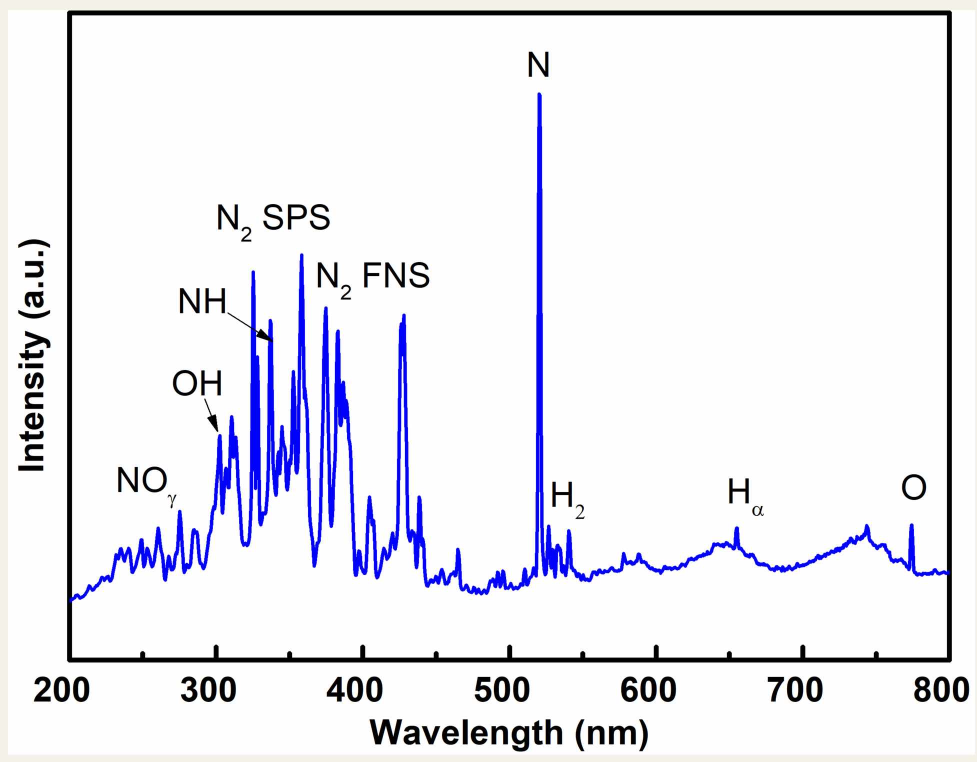


Figure 7

- Optical emission spectroscopy (OES) of the nitrogen discharge in H_2O shows the N_2 second positive band (SPS), as well as the NOγ, OH, NH, H_2, Hα, and O bands. Based on the emission spectrum analysis, N, O, H, OH, and NO were the key species in the water splitting reaction during nitrogen discharge.

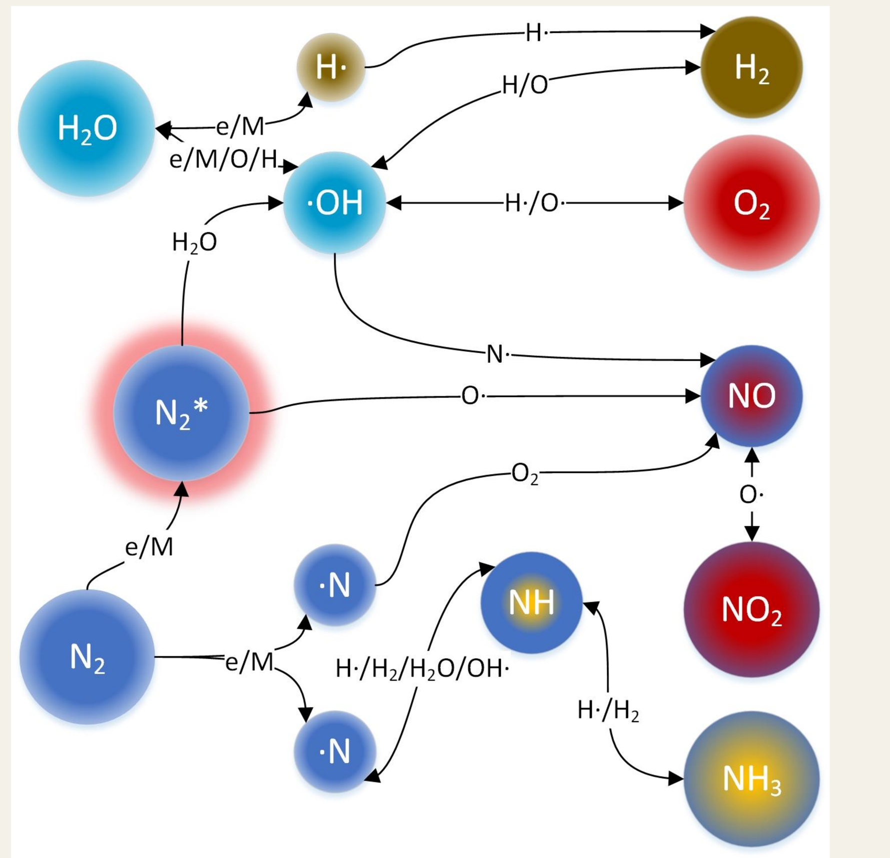


Figure 8

Conclusion

- Plasma catalyst-integrated strategy developed for NH_3 production using renewable electricity and ambient pressures.
- Exceptionally high NH_3 production yield with 95% selectivity and 120 µmol/s production rate demonstrated.
- Potential for scaling up NH_3 production rate by increasing gas flow rate and SEI, with promising production and cost analyses for energy and environmental sustainability.

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Contact Information

Muzammil Iqbal
Brightsite Plasmalab,
Maastricht University, Netherlands
Tel: +31-684433859
Email: muzammil.iqbal@maastrichtuniversity.nl
Web: Plasmascholar.com

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