

Maximizing nitrogen fixation using bipolar pulsed spark discharge

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Abstract: In this contribution, we report holistic analysis of NO_x formation using bipolar pulsed spark discharge operated at high frequency. The key factors determining the performance of plasma NO_x formation were systematically evaluated. The well known trade-off relation and strategy will be discussed based on both kinetics and experiment incorporating new perspective such as critical oxygen line, and dynamic equilibrium maximum.

1. Introduction

Artificial nitrogen fixation using plasma is one of the promising options for electricity-driven valuable compounds which is also referred to as power-to-X (X = H₂, NH₃, NO_x, alcohols, etc) processes. In this work, we present one-step conversion of air into NO_x by warm plasma, which is characterized by the higher vibrational and rotational temperatures than those of the nonthermal plasmas. High-frequency spark discharge showed much higher performance for NO_x synthesis than conventional spark discharge operated at low frequencies. To get insight into the main factors for enhancing energy efficiency and understanding the underlying mechanism, the performance of the reactor was evaluated at different operating conditions. The results revealed that higher vibrational and rotational excitations at a higher frequency and a long electrode gap are critical factors in improving the efficiency of NO_x production.

2. Methods

A high-voltage pulsed power supply (PEKURIS, KJ19-5655-04) with variable voltage, pulse widths (Pw) and frequencies (*f*) is used for NO_x formation experiments. The spark discharge was monitored with a high-speed camera (Photron FASTCAM SA1.1) with a frame rate of 20,000 frames per second (fps) and an intensified charge-coupled device (ICCD, Andor iStar DH334T) camera coupled with various band-pass filters.

3. Results and Discussion

Figure 1 summarizes the energy cost data from recent publications that reporting NO_x yield higher than 1% at atmospheric pressure [1]. Most important advancements have been made during the last five years achieving both high yield and energy cost.

Application of plasma for PtX requires both high NO_x yield and good energy efficiency. However, most plasma chemistry exhibits a trade-off between yield and energy efficiency, which becomes especially apparent when NO_x yield exceeds 2%. Interestingly, most publications reporting higher NO_x yield with improved energy cost have emerged within the last 5 years.

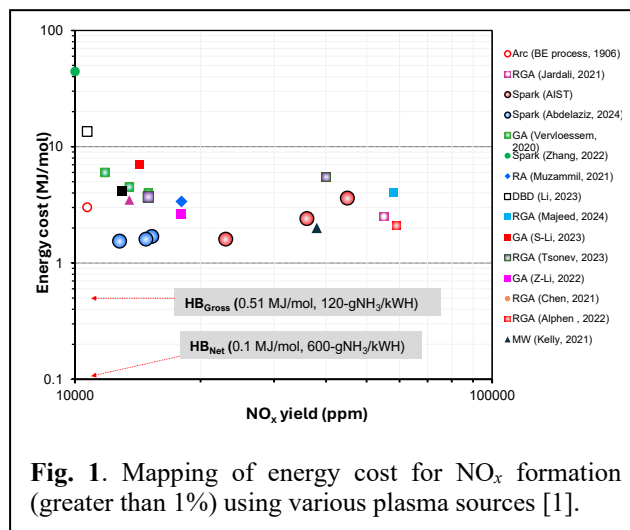


Fig. 1. Mapping of energy cost for NO_x formation (greater than 1%) using various plasma sources [1].

4. Conclusion

With the increasing global demand for a low-carbon society, plasma technology is gaining significant attention for its role in power-to-X solutions, especially when integrated with renewable energy. Gap distance and the oxygen contents were found to be important factors determining the balance between forward and backward reaction. Fine tuning of the various operating conditions enabled the energy costs below 2 MJ/mol in achieving NO_x yield greater than 2%.

Acknowledgement

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