NH_3 synthesis in a DBD: a study from low to atmospheric pressure

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Abstract: In this contribution the formation of NH_3 in a Dielectric Barrier Discharge is investigated at various operating conditions. Special attention is devoted to the understanding of the discharge by deriving electric parameters from Lissajous curves and plasma chemistry quantities from emission spectra.

1. Introduction

The formation of NH_3 from H_2-N_2 plasmas using Dielectric Barrier Discharges (DBDs) at atmospheric pressure has been widely studied in literature. In most cases, studies focus on performance with various packing materials [1], while some works couple modelling and experimental efforts to derive insights into the underlying mechanistic pathways [2]. There is an apparent lack of a systematic study of the discharge, either from insights that can be gained from Lissajous curves [3] or from emission spectra [4].

In this work we study the formation of ammonia as a function of macroscopic parameters and complement it with further understanding of the discharge using Lissajous curves, from which quantities such as the applied reduced electric field or partial discharging can be derived, and high-resolution optical emission spectra, with which rotational and vibrational temperatures as well as N_2^+/N_2 intensity ratios are obtained.

2. Methods

A coaxial DBD with a gap of 4.65 mm is operated at 23.6 kHz in a wide range of operating conditions, including discharge lengths from 27 to 82 mm, packings of dielectric and metal-coated dielectric beads, and pressures from 14 mbar up to atmospheric (Fig.1). The concentration of NH_3 in the outlet stream is quantified using mass spectrometry (MS), while electric characteristics are derived from Lissajous curves. Time-averaged and axially obtained optical emission spectra (OES) of the discharge are recorded.

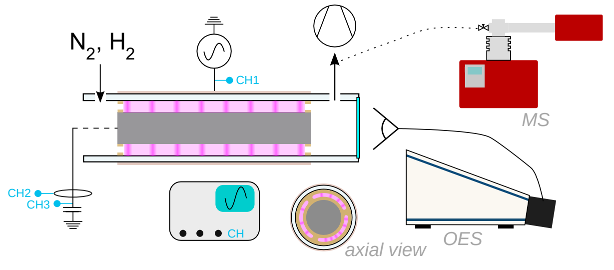

Fig. 1. Experimental setup.

3. Results and Discussion

The NH_3 energy yield, defined by the outflow of ammonia divided by the plasma power, alongside the rotational and vibrational temperatures of N_2 (C-B) as a function of pressure are displayed in Fig. 2a, for H_2-N_2 75-25, a total inlet flow rate of 100 sccm and a 27 mm long empty reactor. An increase in yield with a decrease in pressure is observed, which is both due to a moderate increase in [NH_3] from 1100 to 1300 ppm and a decrease in plasma power from 60 to 46 W. The slight increase of [NH_3] towards lower pressures is accompanied by a decrease in rotational temperature of N_2 (C-B) from around 600 to 375 K and a significant increase in applied E/N from around 40 to 1000 Td (Fig. 2b).

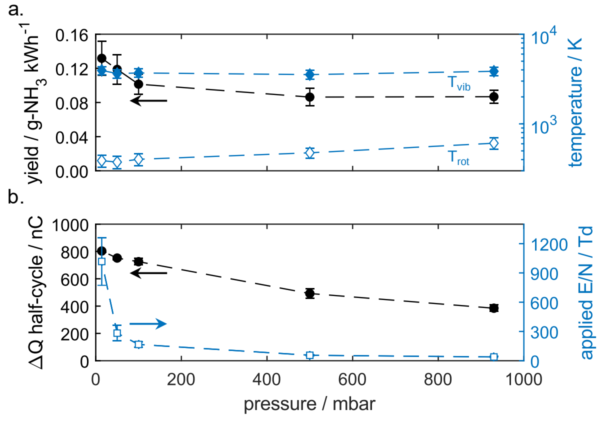

Fig. 2. a. energy yield, rotational and vibrational temperatures of N_2 (C-B) as a function of pressure. b. charge transferred per negative half-cycle and applied reduced electric field as a function of pressure.

The plasma visibly transitions from a filamentary to a more diffuse discharge for pressures below 500 mbar. This behavior is reflected in the current waveforms: while high-intensity current peaks are observed at high pressures, the waveforms become markedly sinusoidal at low pressures. The larger plasma volume at lower pressures is consistent with an increase in partial surface discharging (not shown) and charge transferred per half-cycle ΔQ (Fig. 2b), both of which determined from Lissajous curves.

References

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