Optical Emission Spectroscopy of an Electron Beam Sustained Hybrid Discharge of Nitrogen at 1 mbar

D. J. Schreuder[1,2,3], G. Mattausch[2], B. Zimmermann[2], E. von Hauff[(*)2,3]

[1] *Boysen-TU Dresden-Research Training Group, TUD Dresden University of Technology, Dresden, Germany.*

[2] *Fraunhofer Institute for Organic Electronics, Electron Beam and Plasma Technology (FEP), Dresden, Germany.*

[3] *TUD University of Technology, Faculty of Electrical and Computer Engineering, Dresden, Germany.* [(*)] *david.schreuder@tu-dresden.de*

Power-to-X (PtX) processes can be exploited for conversion and storage of electrical energy from fluctuating renewable sources in chemical energy, and have great potential to contribute to a sustainable energy system and economy. Compared to other approaches, these processes can advantageously be realized in non-equilibrium gas discharges, serving as a reactive medium for the conversion of different gas molecules. In this context, the energy efficiency of CO_2 dissociation in cold discharges has been studied extensively over the past years [1]. Computational studies suggest that vibrational excitation provides an energy-efficient pathway for the dissociation of CO_2 (via ‚ladder climbing') [2]. In practice however, the conventional plasma sources such as microwave discharges (MW) and dielectric barrier discharges (DBD) suffer from a trade-off in conversion degree and energy efficiency [1]. This trade-off behavior originates from the facts that conventional discharges, first, operate at high electric field strength and feed energy in chemically less-effective dissociation, ionization and excitation processes (figure 1), and second, provide inhomogeneous power density across the educts' gas flow.

Electron Beam (EB) sustained hybrid discharges appear as promising tools for improving the current state of the art. The essential feature of such plasmas is that the ionization required for a voluminous atmospheric glow discharge is externally provided (‚non-self-sustained discharge') via EB irradiation at comparably low power. Because of that, the reduced electric field of the EB sustained hybrid discharge can be significantly lower than the minimum attainable field in self-sustained discharges. This allows flexibility in the choice of the electric field, which drives the high-power glow discharge, thus enabling mode-selective energy transfer to chemically most efficient molecular vibrations (figure 1).

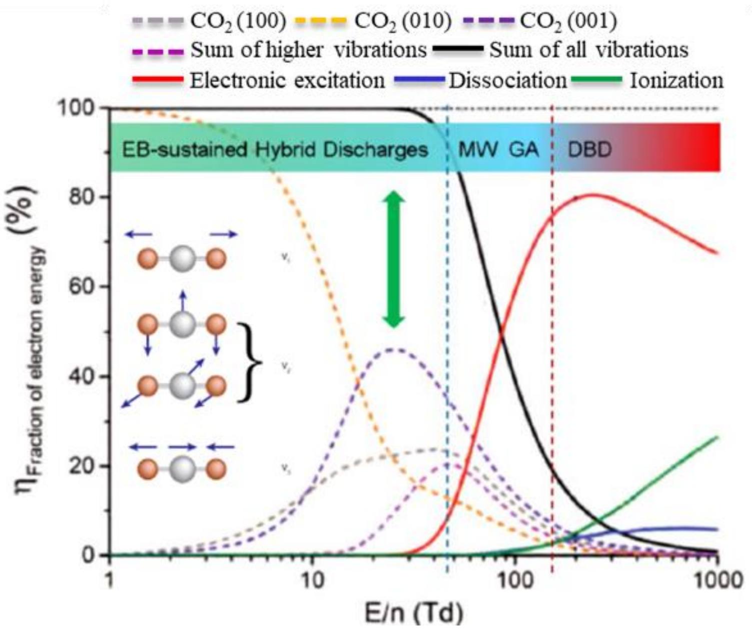

Figure 1: Fraction of electron energy transfer to different processes in CO_2 as a function of the reduced electric field E/n (adapted from [1]).

High-energy beam electrons transfer their energy to the gas via inelastic collisions resulting in ionization processes and secondary electrons [3]. The secondary electrons produced can be heated by electric and magnetic fields to couple supplementary power to the discharge.

In this work, we investigated in the potential for idependently controlling ionization and plasma excitation processes via the electron beam and the inductively coupled plasma (ICP), respectively, using nitrogen as a test gas. We performed quantitative analysis of emission from a 1 mbar N_2 ICP discharge sustained by an electron beam (U_b = 25 kV, I_b = 1-5 mA). The optical emission from the first negative system (FNS) and the second positive system (SPS) of nitrogen is quantified as a function of the EB and ICP currents. Within the beam current range investigated here (I_b = 1 – 5 mA), the FNS (0, 0) (391 nm) has the highest emission intensity (figure 2, solid line) and is linearly correlated to the electron beam current. Furthermore, the emission intensity agrees with the calculated and measured emission in low-pressure hollow cathode electron beam sources reported by Lock et al.[4]. Supplementary ICP power enhances the emission of the SPS bands, while the emission of the FNS (0, 0) signal at 391 nm remains constant. Under assumption of the Corona model, the vibrational energy distribution of the excited state N_2C_3 is calculated via the Boltzmann method and compared to results obtained from a glow discharge experiment at a similar pressure [5]. The influence of the pressure and other parameters on the measured optical emission signals will be discussed. The results show the potential of this experimental approach for chemical conversion processes in non-equilibrium plasmas.

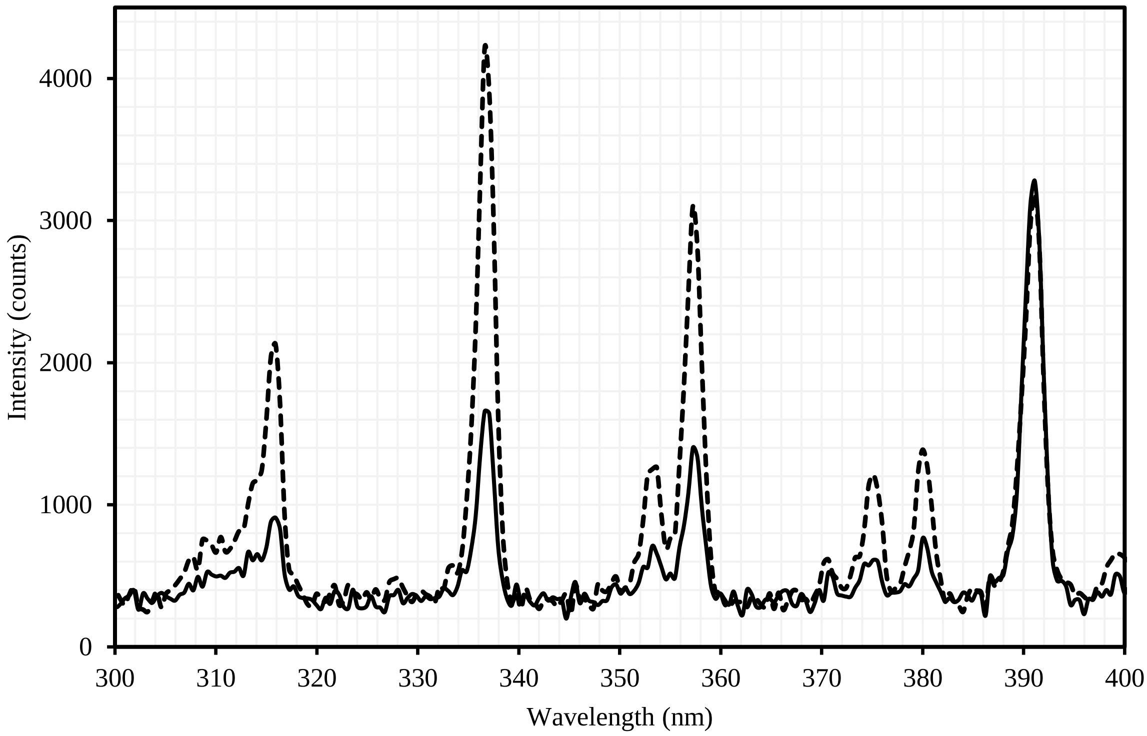

Figure 2: Optical emission spectrum of a 0.54 mbar nitrogen from an EB discharge (solid line) and an EB + ICP hybrid discharge (dashed line). For both spectra, conditions of the EB were U_b = 20 kV, I_b = 1 mA.

[1] R. Snoeckx, A. Bogaerts, *Chem. Soc. Rev.*, **46** (2017) 5805-5863.

[2] A. Bogaerts, T. Kozák, *Plasma Sources Sci. Technol.*, **24** (2015) 015024.

[3] R.F. Fernsler, S.P. Slinker, S.G. Lambrakos, *J. Appl. Phys.* **104** (2008), 063312.

[4] E.H. Lock, R.F. Fernsler, S.P. Slinker, I.L. Singer, S.G. Walton, *J. Appl. Phys. D*, **47** (2014), 425206.

[5] J. Levaton, J. Amorim, V. Monna, J. Nagai, A. Richard, *Eur. Phys. J. Appl. Phys.*, **26** (2004), 59-64.