Plasma Chemical Characterization of a Combined ns Pulsed – RF Excitation Source for CO_2 Conversion at Atmospheric Pressure

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Abstract: We explore a novel 3-electrode system for plasma-assisted gas conversion (consisting of a ns-pulsed, RF and grounded electrode) by applying custom excitation sequences from both the ns-pulsed and RF sources. The formation of two different RF discharge modes is demonstrated and characterized using optical emission spectroscopy and electrical measurements, and effects on the CO_2 conversion via FTIR spectroscopy.

1. Introduction

Utilizing low to moderate temperature plasma provided by low reduced electric field (E/n) sources has demonstrated sufficient chemical kinetics for CO_2 conversion reactions [1]. In a recent publication, our group performed an extensive electrical characterization of our custom-built ns pulse – RF system [2]. Sub-breakdown voltage RF discharges were applied to a range of CO_2 – Ar gas mixtures demonstrating that RF plasma power deposition values of 85% in pure Ar and 76% in pure CO_2 were achievable. Additionally, CO_2 conversion values of up to 5.5% in a 100% CO_2 atmosphere were reported and supplemented by a preliminary plasma chemical analysis via optical emission spectroscopy. A further characterization of the RF plasma chemistry is necessary to provide insight into the utilization of this combined power supply for plasma processing reactions.

2. Methods

The reactor contains a 3-electrode system, two of which form a pin-to-pin geometry (RF and grounded electrode), and the third (ns-pulsed electrode) adjacent to the RF and ground but separated by a quartz dielectric (Fig. 1 A)). Power delivery is defined by a certain pulse sequencing of both the ns-pulsed and RF source, and additionally, the RF source can be modulated by means of either pulse bursting or amplitude modulation. Measurements obtained from the system include mid-resolution optical emission spectroscopy to approximate electron densities and excited species temperatures, as well as CO_2 conversion for the CO_2 dissociation reaction with in-line FTIR spectroscopy.

3. Results and Discussion

The RF waveform is applied at t = 0 without the formation of a plasma. After 250 µs, a ns-pulse burst is applied (20 kHz, 2 pulse burst) to break down the gap and initiate the RF plasma. RF is continually applied for a specified duration (variable), then turned off. The entire sequence is repeated every 10 ms (100 Hz). Triggering the RF pulse pre-ns-pulse provides validation of whether the RF waveform is sub-breakdown from pulse to pulse.

Depending on the RF pulse parameters, it was found that two different RF discharge modes could occur. Shorter duration RF pulses form between the RF electrode and the dielectric-separated ns-pulsed electrode (termed RF-DBD). By increasing the RF pulse length, ambipolar diffusion and drift enable the RF plasma to form between the pin-to-pin electrodes, from RF to ground (termed RF-PtP). At these conditions, highspeed imaging has revealed that the longer RF pulses develop from RF-DBD to RF-PtP every cycle. The plasma characteristics between the two modes differ. The RF-PtP discharge exhibits higher plasma power deposition and a closer rotational-vibrational temperature equilibrium, when compared to RF-DBD mode. Accompanying the higher species temperatures, the RF-PtP mode results in higher CO_2 conversion values (>2%, vs. ~0.5% in RF-DBD mode). The above-mentioned characteristics are summarized in Fig. 1 B).

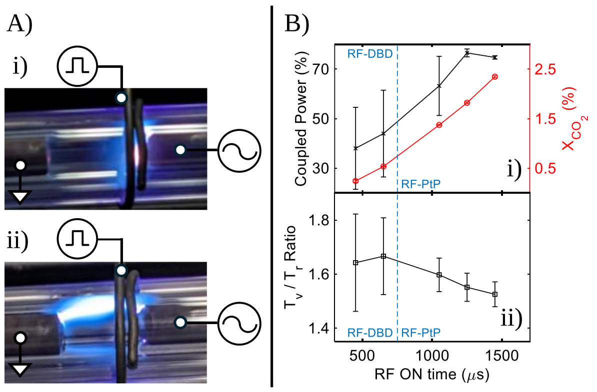

Fig. 1. A) Reactor geometry showing 3 electrode system (RF, right; ns-pulse, top; ground, left), with RF plasma in RF-DBD mode i) and RF-PtP mode ii). B) Measured characteristics for RF pulses between 300 and 1500 µs: i) RF plasma power deposition and CO_2 conversion, ii) vibrational-rotational temperature (T_v/T_r) ratio based off emission from the 309 nm OH band.

4. Conclusion

A 3-electrode geometry utilizing a combined ns-pulse and RF excitation system is investigated. Depending on which two electrodes the RF plasma forms between, plasma characteristics such as power deposition and rotational-vibration equilibrium are found to vary.

References

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