Electrical Characterization of CO_2-Ar Atmospheric Pressure Discharges Sustained by High-Voltage Nanosecond Pulse-Radiofrequency Excitation

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Abstract: A combined high voltage ns pulse-radiofrequency (RF) excitation system was developed to perform non-thermal plasma chemistry at atmospheric pressure and with high reactive to inert gas ratios. Plasma power dissipation via sub-breakdown RF excitation was determined for gas mixtures ranging from 5 to 50% CO_2 in Ar. An alternative method based on measurements taken on the RF generator side was used to determine the plasma power dissipation.

Keywords: Non-thermal plasma, ns pulsed plasma, radiofrequency plasma

1. Introduction

Chemical process electrification through plasma processing offers novel and promising avenues for the synthesis of various chemicals and alternative fuels with the ultimate goal of drastically reducing the emissions and circularizing resource uses. Developing an effective plasma chemical synthesis or conversion process requires the integration of optimized reactor design, catalyst design and efficient power delivery. By attempting to 'power' chemical processes with electricity, the choice of method of delivering power is critical to create ideal plasma chemical conditions for a given synthesis reaction.

It has been demonstrated that sufficient chemical kinetics for certain synthesis reactions can be obtained by implementing low to moderate temperature non-thermal plasma [1]. Amongst the many excitation sources that can create these plasma conditions, high voltage (HV) nanosecond (ns) pulsed plasma sources and radiofrequency (RF) sources are common. High voltage ns pulsed plasma sources are effective at igniting and sustaining plasmas in atmospheric pressure gases and gas mixtures. These pulses produce highly transient non-equilibrium conditions and, in-turn, create large quantities of excited species and highly reactive radicals participating in the desired chemical reaction pathways. When sufficiently separated in time, the power delivery of each pulse is relatively discrete resulting in minimal memory effect. The rapid quenching of the electron and excited species densities causes the discharge to essentially face re-ignition conditions every pulse. This highly dynamic load impedance leads to low efficiency of power delivery from the electrical mains to the plasma.

RF discharges have been studied and applied extensively to functionalization and synthesis reactions under vacuum conditions. Applications extending into the atmospheric pressure regime require the plasma gas to mainly consist of easily ignitable and maintainable noble gases and can withstand reactive gas additions up to a few percent. At higher concentrations of reactive gases, the RF discharge can experience significant constriction followed by extinction. Commercially available RF power supplies cannot reach the breakdown voltage thresholds required to ignite and sustain a useful discharge volume at atmospheric pressure and in high percentage reactive gas mixtures. An advantage of RF excitation is that the electrical energy conversion into plasma chemical energy efficiency can be quite high. It has been theorized that low reduced electric field (E/n) discharges can provide a highly efficient chemical reaction pathway, and the E/n for RF excitation waveforms are situated directly in this region as shown in Fig. 1 [2]. Experimentally it has been demonstrated that large fractions of the discharge energy are coupled into these vibrational excitation modes confirming the plausibility for utilizing this pathway [3].

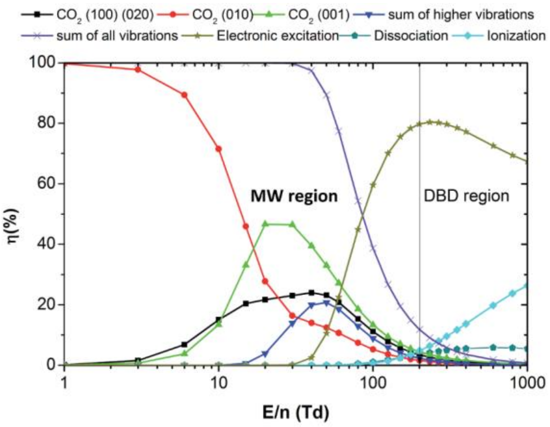

Fig. 1. Energy loss channels as a function of reduced electric field (E/n) of CO_2. The E/n region of typical microwave (MW) plasma is indicated, which is a similar region to RF [2].

2. Methodology

A proposed concept of combining a ns HV pulse system with a RF system should provide a way to utilize the advantages of each while mitigating the drawbacks. The high voltage ns pulse can easily breakdown the gas mixture by supplying charged species into the interelectrode gap. A sub-breakdown RF waveform utilizes the charged species left over from the ignition pulse and can proceed to dissipate energy into the plasma. This allows a significant portion of the electrical energy to be delivered into the plasma via RF excitation. Since the applied power from the RF generator is not representative of the dissipated plasma power, it is necessary to electrically characterise the system to distinguish between the two.

The plasma power deposition in a capacitively coupled RF discharge can be calculated from experimental measurements using:

$$P = V_{rms} I_{rms} \cos\left(\left(\Delta\theta - \theta_{ref}\right) + \frac{\pi}{2}\right) \quad (1)$$

where V_{rms} and I_{rms} denote the voltage and current measurements taken at the RF electrodes, respectively. $\Delta\theta$ is the phase angle during sustained RF discharge, and θ_{ref} is the plasma off state phase angle. Highly reactive plasma loads, although functionally important to achieve high-Q and high RF peak voltages, can lead to error prone calculations as the θ_{ref} is always close to 90°.

An alternative proposed method for calculating the plasma dissipated power is by examining the change in load impedance due to the formation of a discharge. The comparison between the circuit impedance with and without discharge can provide the power dissipation into the plasma. Determining the change in load impedance can be done by taking measurements on the source side (50 Ω output of generator), and accounting for the impedance transformation through the matching network.

3. Experimental Design

The experiments were carried out at room temperature and atmospheric pressure with various CO_2 to Ar ratios, adjustable by mass flow controllers. The reactor construction consists of a borosilicate glass tube (OD: 10mm, ID: 8mm) acting as a dielectric separating the RF electrode on the exterior and enclosing the gas flow and ns-pulsed electrode (3.18mm OD SS316 rod) positioned on the centerline. The high voltage ns pulsed power supply is a custom design utilizing magnetic pulse compression to achieve a maximum output of 10 kV with a 75 ns rise time. The RF system consist of (in order) a function generator, amplifier, bi-directional coupler, matching network, and load circuit. The bi-directional coupler is custom made and provides the source side measurements of forward and reflected power, and voltage and current measurements. At the RF operating frequency of 13.56 MHz, the maximum applied power of the amplifier is 120 W. The excitation frequency and sequence are completely controllable via a trigger generator. This sequence defines the overall excitation period which consists of the triggering and delay time of the high voltage pulse, and the duty cycle (ON time) and delay time of the RF pulse.

4. Results & Discussion

The experiments performed provide a relationship between the plasma power dissipation and gas mixture ratio (referred to as x% CO_2 in Ar). The gas mixture ratio was varied from 5 to 50%, and for each mixture ratio, several power levels were tested. For each mixture ratio, the minimum power level was determined by finding the power/gain level such that the RF system began dissipating power into the plasma. This onset of power dissipation is signified by observing a change in the bi-directional coupler signals, indicating a change in impedance. Additional trials for each gas mixture ratio were performed in increments of 10 to 20 W from the previous power level. Fig. 2 shows the plasma dissipated power at certain applied RF power for a gas mixture ratio of 25% CO_2 in Ar.

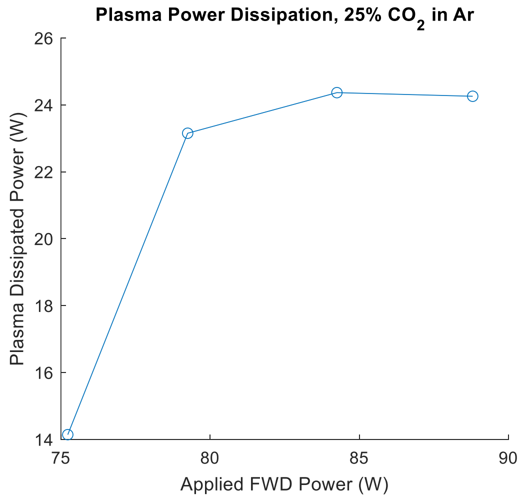

Fig. 2. Plasma dissipated power vs. applied RF power for 25% CO_2 in Ar.

Maintaining a discharge with more than a few percent of reactive gas content is energy intensive due to the increased number of modes for energy dissipation, as compared to an atomic noble gas like Ar. CO_2 is no exception to this trend and this is evident with the rapid increase in minimum power required to sustain the RF discharge power dissipation for increased CO_2 percentages. For higher CO_2 percentages, the amplifier is limited by the maximum applied power it can provide and therefore, the maximum power than can be dissipated in the plasma. Characteristic to all gas ratio trials is the steep increase in dissipated power after the minimum power level to observe RF discharge power dissipation. Following this point, plasma dissipated power still increases with increasing applied power, however, with smaller increments.

Observing RF discharge power dissipation indicates a coupling of electrical power to the plasma, which is due to the measured change in impedance. Since the matching network settings are kept constant, the changing impedance leads to an increase in reflected power, which for the sake of power transmission, is a decrease in electrical efficiency.

5. Conclusions

An investigation into determining the power coupled into an independently ignited sub-breakdown RF discharge was performed. An alternative method for measuring the plasma impedance was proposed to obtain measurements of plasma dissipated power. Discharge experiments were carried out in different gas mixture ratios ranging from 5 to 50 % CO_2 in Ar, and at several power levels for each mixture ratio. Coupled power was measured and a non-linear relationship between increments of applied power and dissipated power was observed. The consequence of RF transmission losses as a product of coupling RF power to the plasma was also observed. Further investigation into the effect of matching network settings will be carried out with the system discussed. This could determine if coupled power levels can be maintained while reducing the RF transmission losses that arise from the changing load impedance.

6. References

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