Short nanosecond microwave pulses for sustaining CO_2 plasma at atmospheric pressure: advances and challenges

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Abstract: An atmospheric CO_2 plasma sustained in a coaxial torch with short, nanosecond microwave pulses is compared with a plasma sustained with continuous microwave power. The power absorbed by the plasma and the temperature of plasma species are estimated for different pulse lengths, duty cycles and gas flows. Among other challenges posed by pulsed microwave plasma is the calculation of the microwave power absorbed in the plasma which is demanding for the correct estimation of process efficiency.

Keywords: CO_2 conversion, pulsed microwave plasma, nanosecond pulses, OES.

1. Introduction

The plasma assisted conversion of CO_2 into synthetic fuels based on renewable energies is considered as promising approach for mitigation of CO_2 emission and energy storage [1]. Among different plasma discharges, namely the microwave sustained plasmas have shown to be most efficient for CO_2 splitting reaction $CO_2 \rightarrow CO + {}^1/_2\, O_2$. Due to its high electrical energy density and preferential activation of vibrational excitation states of CO_2 molecules, an energy efficiency of up to 80 % was demonstrated in microwave plasma reactors at low pressure [2]. In spite the systems operating at atmospheric pressure are more attractive for industrial applications, the energy transfer from vibrational (T_{vib}) to rotational (T_{rot}) states at higher gas pressures increases and results in plasma thermalization and deterioration of reaction energy efficiency [3]. The sustaining CO_2 plasma with short, nanosecond microwave pulses rather than with the continuous microwave may increase the energy efficiency by shifting the thermal equilibrium in the plasma towards T_{vib}.

In the present work, the energy efficiency of CO_2 splitting in atmospheric microwave plasma is studied versus an amplitude modulated microwave power at nanosecond time scale.

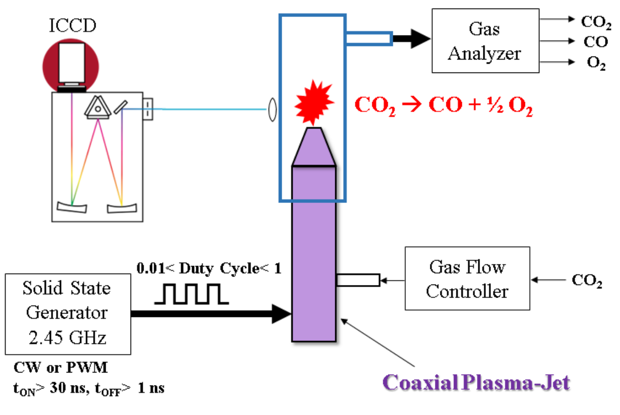

Fig. 1. Scheme of the experiment.

Fig. 2. Lab setup of the plasma experiment.

2. Experimental Setup

Figure 1 shows the scheme of the experiment whereas Fig. 2 shows a photo of the corresponding lab setup. A solid-state microwave source from HBH Microwaves GmbH with a maximum output power of 250 W and an operating frequency between 2.4 and 2.5 GHz is used. The microwave is fed via a coaxial line into the plasma reactor. A bi-directional coupler and a calibrated fast oscilloscope (Wave Runner 640Zi) allow the control of the microwave power absorbed in the plasma. A plasma torch, type *PS-Cle* from company Heuermann HF-Technik GmbH, is utilized as the reactor. It is a coaxial torch with the inner conductor made from copper and a cylindrical body, which serves as the outer conductor. The CO_2 gas flows between the outer and the inner conductor towards the field concentrator, where the plasma ignites. An Emerson X-STREAM gas analyser measures the concentrations of the reaction products: CO_2, CO and O_2. An advanced optical emission spectroscopy (OES) system for registration of emitted plasma photons features a high-resolution spectrograph (Acton SP-2756) with 750 mm focal length and 512x2048 pixel CCD camera (A-DH340-18U-03) with an intensifier and fast (≥2ns) gate from company LOT-QD GmbH. This enables the acquisition time of plasma emission spectrum to be much shorter than a single microwave pulse. In the experiment, the microwave operated in a pulsed regime with a constant power level of 200 W. The pulse duration time (t_{on}) and the interval between the pulses (t_{off}) were varied to allow duty cycle $DC = t_{on}/(t_{on}+t_{off})$ between 0.01 and 1. Three different CO_2 gas flows were used: 12 slm, 15 slm and 18 slm, measured at the input of the plasma jet. Finally, the CO_2 conversion is estimated from the measured concentration of the reaction products.

3. Energy efficiency

Generally, the energy efficiency is defined as the ratio of the energy spent for the dissociation of the CO_2 molecules versus the total microwave energy consumed:

$$\eta = \frac{Flow\ (mol/s)\cdot\chi\cdot\Delta H_R^0(J/mol)}{P_{eff}(W)}. \quad (1)$$

Here χ, ΔH_R^0, P_{eff} are the CO_2 conversion, reaction enthalpy and effective microwave power absorbed in the plasma, averaged power over the period of power modulation.

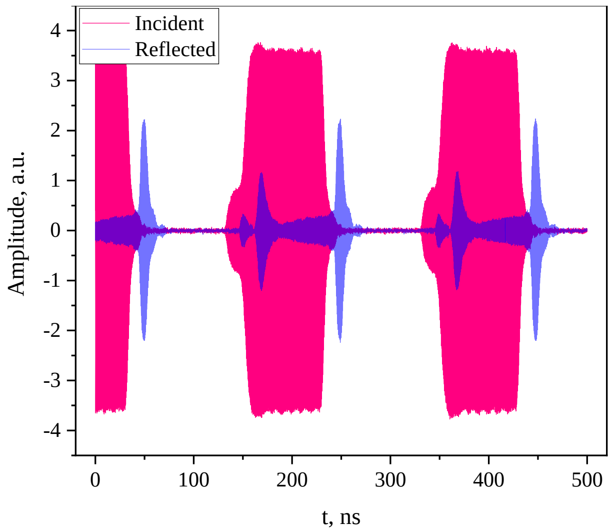

Fig. 3. Example of incident and reflected signals for t_{on}=100 ns and t_{off}=100 ns measured at oscilloscope.

The correct estimation of P_{eff} requires the integration of the power of both incident and reflected signals over the period of power modulation ($t_{on} + t_{off}$). Those signals are far from ideal rectangular form (see Fig. 3) and cannot be evaluated analytically but rather numerically for every individual t_{on} and t_{off}. This procedure includes the calibration of oscilloscope and waveguide components with the power generator (Anritsu MG3694C) and power meter (Anritsu ML2488B), and the integration of recorded oscilloscope signals and their post-processing with MATLAB. As an example, for three duty cycles, 0.20, 0.33 and 0.50, the estimated absorbed power is shown in Fig. 4 with symbols. The dashed lines correspond to 100% absorption of an ideal rectangular pulse in a plasma.

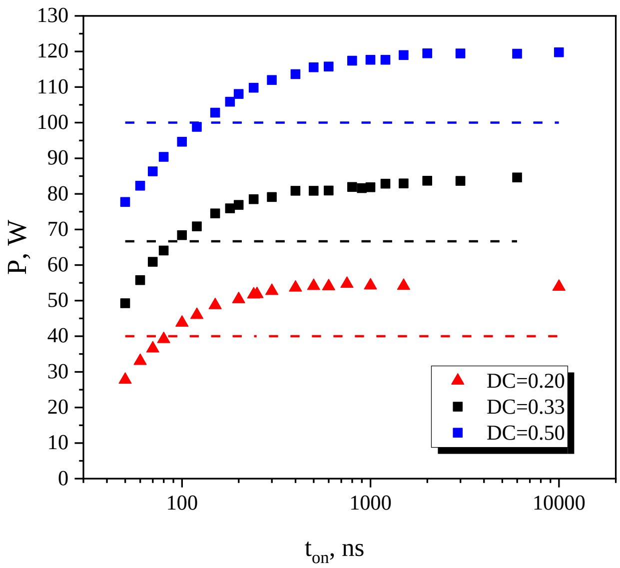

Fig. 4. Calculation of absorbed microwave power in plasma averaged over the period of power modulation (symbols). Incident power per period for an ideal rectangular form of pulse given with dashed lines as reference.

It is seen, that the effective absorbed power in the plasma increases from short pulses towards longer ones by 30% to 40% that, of course, is not negligible.

After the calculation of the CO_2 conversion rate χ from the gas analyser signals and P_{eff} from procedure described above, the energy efficiency for different scenarios was estimated with equation (1).

Within the parameter scan, a maximum energy efficiency of about 40 % is reached for a duty cycle of 0.2 and an input gas flow of 12 slm. It corresponds to a specific energy input (SEI) of 0.05 eV/mol.

It is also found that a plasma sustained with continues microwave power (t_{off} =0) is less energy efficient as compared to a plasma sustained with pulsed microwaves. This is a direct indication that the pulsed energy supply influences the thermal equilibrium in plasma towards the vibrational energy states.

The optical emission spectroscopy (OES) diagnostic for estimation of T_{vib} and T_{rot} of plasma species both within power-on and power-off phases is utilized and the results will be discussed in the paper.

References

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