Optimizing the CO_2 conversion efficiency in a low-pressure pulsed microwave plasma source

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The CO_2 decomposition process in a pulsed 2.45 GHz microwave surfaguide discharge has been studied. The CO_2 conversion efficiency is found to be mainly affected by the plasma pulse repetition rate (at fixed applied power), along with the other discharge parameters such as the gas flow rate, residence time, etc. The electron and gas temperatures have been additionally studied using spectroscopic methods. A several time increase in the CO_2 conversion/energy efficiency points out on a crucial role of the pulsed plasma regime for better CO_2 conversion. The found effects are explained by the relevant energy relaxation mechanisms in the discharge, such as the electron-vibrational, vibrational-vibrational, and vibrational-translational ones.

1. Introduction

In spite of the numerous works devoted to plasma-based greenhouse gas conversion, related to microwave (MW) plasma [1], dielectric barrier discharge (DBD) [2], gliding arc plasma (GAP) [3], as well as those involving plasma catalysis, the effects of CO_2 conversion in the pulsed discharges are still far from being understood clearly. So far the beneficial role of plasma power modulation has been only demonstrated in the DBD case [4]. The present work studies the power modulation effect in MW surfaguide discharge, as a promising candidate for plasma-assisted CO_2 conversion.

2. Experimental

A surfaguide-type pulsed microwave plasma source has been used. The plasma was sustained by the electromagnetic waves with the filling frequency of 2.45 GHz, and modulated by nearly rectangular pulses with the repetition rate ranging from 0.01 to 2.5 kHz. The pulse duty ratio was mainly fixed at 50%. The discharge has been sustained in a quartz tube (14 mm in diameter and 31 cm long) in which the gas flow was regulated by digital mass flow controllers. The quartz tube has been cooled down by 10 °C flow of Si oil. More details are available in [1,5]. The optical emission actinometry [1] and two-photon absorption laser induced fluorescence (TALIF) techniques were used for monitoring the CO_2 conversion efficiency in our case [5].

3. Main results

In this work it was shown that the CO_2 conversion efficiency depends dramatically on the plasma pulse repetition rate. Up to a fourfold improvement in the CO production in the post-discharge (and thus in the CO_2 conversion rate) has been detected (see Fig. 1).

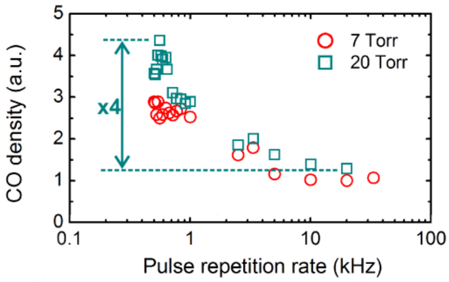

Fig. 1. Relative CO density in the post-discharge as a function of plasma pulse repetition rate.

It is also shown that the CO_2 conversion depends on the molecule residence time and the gas pressure. The electron temperature determined based on the Ar line ratio shows rather minor variations in the studied pulse frequency range, leading us to a conclusion that the vibrational energy exchange is the main reason for the observed effects. The estimates made for the electron-vibrational (e-V), vibrational-vibrational (V-V) as well as vibrational-translational (V-T) energy relaxation time point out on a primary role of vibrational excitation for CO_2 decomposition at long plasma pulse durations (low rep. rates), as well as its contribution to the gas heating via the V-T process. At the same time at high repetition rates these processes are less efficient due to shorter plasma pulse duration.

4. References

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