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O atom kinetics in CO_2 pulsed glow discharges

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O atoms play a key role in the efficiency of CO_2 plasma recycling processes because their reactions can either promote or reduce the conversion of CO_2. We have measured the O atom density and recombination probability by means of actinometry and TALIF and compared the results with the time evolution of CO measured by FTIR spectroscopy in pulsed glow discharge.

1. Introduction

Different strategies have emerged to deal with the excess of CO_2 emissions, whose increasing proportion in the atmosphere is the major cause of the global warming. One of these approaches is focused on CO_2 recycling, which is an initial step in building more complex organic molecules, such as energy-dense hydrocarbon fuels. In this regard, the dissociation of CO_2 to CO, as a first step, through the so called "vibration up-pumping mechanism" is believed to be the most efficient method, especially through the excitation of the asymmetric-stretch vibrational mode of CO_2 [1].

The dissociation of CO_2 results in the production of O atoms that can recombine into O_2, oxidize CO back into CO_2, or on the contrary dissociate further CO_2. O atoms therefore play a key role, beneficial or not for the efficiency of CO_2 conversion. To investigate the fundamentals of CO_2 plasma kinetics, a simple glow discharge powered with pulsed or modulated voltage is studied. This simple discharge allows to measure fundamental parameters such as the vibrational excitation of CO and CO_2 [2], the gas temperature and the electric field. This paper focuses on the measurement of O atom densities and recombination probabilities on the reactor wall and their comparison with CO production.

2. Experimental setup

The temporal evolution of the different species is studied by means of time-resolved *in situ* FTIR spectroscopy, actinometry and TALIF. The CO_2 plasma is ignited in a cylindrical Pyrex tube (2 cm inner diameter and 22 cm or 64 cm length) plasma reactor. Water circulating around the reactor allows control of the wall temperature between 5-50°C. Different gas flows (up to 50 sccm), pressures (millibar range), currents (10 to 50 mA) and pulse durations (millisecond range) were investigated.

3. Results

In actinometry experiments, the ratio of the intensities of O atom lines (at 777 nm and 845 nm) over Argon line at 750 nm were recorded and fitted obtaining the O atom recombination coefficient**.** The O atom characteristic decay time was found to be in the order of tens of milliseconds. Noticeable differences were observed in CO_2 compared to pure O_2, especially as a function of the wall temperature. The absolute O atom density was also obtained and compared with TALIF measurements. The time evolution of the dissociation fraction (ratio of CO over ($CO+CO_2$)) was determined under similar conditions by absorption spectroscopy (FTIR) and compared with the O atom density evolution. These results are analysed in light of a kinetic model [3].

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5. References

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