

Pretty exciting - Calibrated OH laser-induced fluorescence spectroscopy, opportunities and challenges in unravelling the role of H₂O in CO₂ plasma conversion

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1. Introduction

In the course of climate change, interest in CO₂ utilisation techniques like plasma conversion is steadily increasing. Striving for application, it is crucial to not limit the research to ideal laboratory conditions but also to consider the real world situation. In particular, impurities must be considered among which H₂O plays a vital role since it can potentially serve as abundant hydrogen source.

To illuminate the role of water in carbon dioxide conversion, laser-induced fluorescence spectroscopy (LIF) is used to detect the hydroxyl radical in a pulsed DC glow discharge consisting of CO₂ with H₂O admixture. OH formed by H₂O dissociation, is of high interest due to its reactivity and can be used to probe the CO₂ conversion process.

2. Results

2.1. Quantitative OH LIF in a CO₂-H₂O glow discharge

A novel technique is introduced to calibrate the OH LIF setup by means of two-photon absorption LIF spectroscopy measurements on a CO-filled gas cell. OH densities in the order of $1 \times 10^{18} \text{ m}^{-3}$ are reported at a pressure of 6.67 mbar with the admixture of 20% of water and a discharge current of 50 mA. Furthermore, the time evolution of the CO₂ conversion is determined from collision energy transfer LIF and validated against literature. The time-dependent rotational temperature of OH is obtained with LIF thermometry [1].

2.3. Spectral crosstalk with excited CO

When decreasing the water content in the used glow discharge, spectral overlap between the 3064 Å system of OH, in particular the excitation of the P₁(3) transition for number density determination and the Q₁₂(1), Q₂(1), Q₁(6), Q₁₂(3) and Q₂(3) transitions for temperature determination, and the third positive system of CO, in particular transitions of the (0,0) and (0,2) bands, becomes apparent. The overlap is dis-

torting excitation and fluorescence spectra, see fig. 1, as well as fluorescence time decays after excitation of the P₁(3) transition of OH. As a consequence, systematic errors are introduced into the determination of temperatures, gas compositions and absolute number densities. A new set of transitions is proposed to circumvent the distortion while still allowing for quantitative measurements due to the availability of non-radiative rate coefficients for these transitions [2].

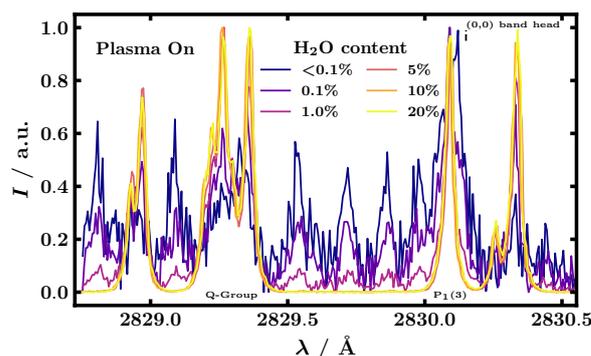


Fig. 1: Distortion of an excitation spectrum of OH for decreasing water content in a CO₂ glow discharge. The yellow line represents a spectrum dominated by OH. Important OH transitions are indicated at the bottom.

Here Q-Group abbreviates the Q₁₂(1), Q₂(1), Q₁(6), Q₁₂(3) and Q₂(3) transitions. Appearing lines belong to the (0,0) band of CO, see the band head at 2830.15 Å.

References

- [1] M. Budde, L. M. Martini, M. Ceppelli, S. Quercetti and R. Engeln 2021 *Plasma Sources Sci. Technol.* submitted for publication
- [2] M. Budde and R. Engeln 2021 *Applied Spectroscopy* submitted for publication

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