Temperature measurements in low-temperature plasma by optical emission spectroscopy



Introduction: With the more and more apparent consequences of climate change, we have to look for new and innovative ways to reduce the CO_2 concentration in the atmosphere. A very appealing possibility is the plasma induced conversion of CO_2 into hydrocarbon fuels. If the plasma is generated with electricity from renewable energy sources these fuels are called solar fuels. Plasma is an ideal tool to conduct the most energy intensive step in the conversion process, namely the splitting of CO_2 into carbon monoxide and oxygen. Due to its non-equilibrium nature the plasma can split the CO_2 molecule in a very energy efficient way.

To tailor the plasma process a detailed understanding of the present molecular species and their interactions is therefore crucial. Furthermore, we want to assess the effectivity of the plasma. At PMP we use optical emission spectroscopy (OES) to measure spectra of molecules that allow us to infer the different temperatures of the plasma. Those temperatures in turn are used to assess the beforementioned aspects.

Different temperatures can be distinguished among which the rotational temperature is commonly determined from the molecular bands in the emission spectra from OES. It is usually assumed that the rotational temperature is the same for all molecules in the plasma, for instance often N_2 or CO are used. However, it has been shown that the temperatures are not necessarily the same [1]. Furthermore, we have observed some unexpected temperature trends like in figure 1 where the temperature decreases even though the plasma is still on [2].

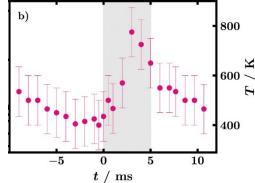


Figure 1: Rotational temperature of OH in a CO_2 - H_2O glow discharge determined from LIF thermometry. Grey background indicates the time the plasma is on [2].

Project description: Time-resolved OES will be applied to a glow discharge plasma to obtain the rotational bands of different molecules. Starting from existing tools those spectra will be fitted for multiple of these molecules. The obtained temporal temperature trends will be interpreted in the context of the CO_2 conversion process.

References:

- 1. Ceppelli et al. 2021 Plasma Sources Sci. Technol.
- 2. Budde et al 2022 Plasma Sources Sci. Technol. 31 055002

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Temperature measurements in low-temperature plasma by infrared absorption spectroscopy

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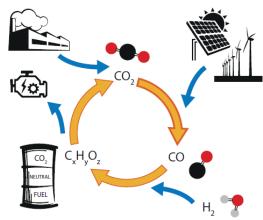
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Study on the positive influence of N₂ on the CO₂ conversion process in a plasma by optical emission spectroscopy



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of CO_2 [Mark Damen PhD Thesis 2020].

It is known that N_2 can promote the vibrational excitation of CO_2 . This is for instance used in CO_2 lasers. The common explanation is that vibrational energy can be effectively transferred from N₂ to CO₂. This is however rarely demonstrated quantitatively. On option to do so is by comparing the vibrational temperature of CO₂, for instance from IR absorption spectroscopy, with the vibrational temperature of N₂. The latter cannot be determined from IR spectroscopy but from optical emission spectroscopy (OES).

Project description: Time-resolved OES will by applied to CO₂ plasma with N₂ admixture. From a fit of the vibrational lines of nitrogen its vibrational temperature will be determined and then compared with the vibrational temperatures of CO₂. This will allow us to assess the importance of vibrational energy transfer between the two molecules concerning the conversion process.

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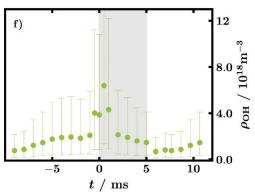
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Ex situ infrared absorption spectroscopy study on H₂O₂ from a CO₂ conversion plasmas with H₂O admixture



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Obviously, the production of hydrocarbons required the introduction of hydrogen to the system. Water could be a cheap option. However, the ongoing chemistry in the plasma becomes much more complicated. Highly reactive species like OH and H_2O_2 might be produced and with an unknown effect on the conversion process.

Figure 2: Density of OH in a CO_2 - H_2O glow discharge determined from LIF spectroscopy. Grey background indicates the time the plasma is on [1].

In a quantitative study on OH we obtained the time-dependent absolute OH number density shown in figure 1. Note that even milliseconds after the plasma is turned off, OH is still observed even though its lifetime should be much shorter. H_2O_2 dissociation might serve as a source for OH.

Project description: With H_2O_2 being a fairly stable molecule is sufficient to look for it in the post-discharge region. Fourier-transform infrared (FTIR) spectroscopy will be used to quantify the amount of H_2O_2 produced in the plasma and what parameters influence that amount.

References:

1. Budde et al 2022 Plasma Sources Sci. Technol. 31 055002

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Electrical characterization of a CO₂ conversion plasma

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A fundamental parameter deciding which processes get excited is the so-called reduced electric field E/N, where E is the electric field and N the absolute gas number density. E/N is also crucial when trying to model the plasma. With a dedicated plasma reactor the measurement of E/N is realized through the measurement of the voltage drop between two electrodes introduced into the plasma the pressure, and the gas temperature.

Project description: With the available gas feed system low-temperature plasma will be created from various gas mixtures including CO₂ and the reduced electric field measured. In the end a little database for plasma modeling will be obtained. Moreover, a look at electron kinetic modeling will be possible.

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