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Temperature Measurements in a Carbon Dioxide Flow using Laser-Induced Fluorescence

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Abstract

Growing interest in the investigation of the Martian atmosphere requires ground-based test facilities, which are able to simulate the thermochemical flow conditions prevailing during entry. At the Institute of Thermodynamics at the Bundeswehr University Munich an arc-heated plasma wind tunnel can provide steady-state, high-enthalpy flows. Being equipped with a high efficiency vacuum pump and being operable with CO_2 as the test gas, realistic entry conditions into Martian atmosphere can be provided. For the thermometry of the high-temperature flow, non-intrusive, optical test methods are well suited. In the present work, the high-temperature flow with an enthalpy of 3.1 MJ/kg and 3.8 MJ/kg is investigated with a two-photon excitation process of CO molecules. For the excitation of the Q-branch of the $B^1\Sigma^+ \leftarrow X^1\Sigma^+(0,0)$ transition, the laser is tuned from 230.05 nm to 230.11 nm. Simulated spectra were calculated in advance with a simulation tool called NOCO-Spectra. The rotational temperature of CO in a high-temperature flow is evaluated by a correlation automated rotational fitting method (CARF).

Keywords: *arc-heated plasma wind tunnel, carbon dioxide, laser-induced fluorescence*

Nomenclature

Latin
A_{xy} – Einstein coefficient for spontaneous emission
B_{xy} – Einstein coefficient for absorption / stimulated emission
b_{xy} – Rate constant for absorption / stimulated emission
h – Planck's constant
h_0 – Total enthalpy
I – Laser irradiance
Q_{diss} – Rate constant for predissociation
Q_{xy} – Rate constant for quenching
W_{2i} – Rate constant for photoionization
W_{xy} – Rate constant for absorption/stimulated emission
W_x – Energy level
Greek
σ_{2P} – Absorption cross section
ω_{xy} – Frequency
Subscripts
x, y – Notation for energy level

1. Introduction

Ground based facilities offer a valuable method for the investigation of high-enthalpy thermochemical effects and the simulation of flow properties prevailing during entry of vehicles flying through the atmosphere of planets e. g. Mars. An arc-heated plasma wind tunnel with a maximum total enthalpy of more than 20 MJ/kg is in operation at the Institute for Thermodynamics at the Bundeswehr University Munich. As this test facility is equipped with a high efficiency vacuum pump and as it can provide a

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4 Director, Institute for Thermodynamics, Bundeswehr University Munich, Werner-Heisenberg-Weg 39, 85577 Neubiberg (Germany), christian.mundt@unibw.de steady-state flow of CO_2 as the test gas, realistic entry conditions into Martian atmosphere can be provided.

The characterization of the flow field can be carried out by conventional diagnostic methods like thermocouples, pitot-static probes or gas extraction. However, arc-heated flows present an enormously challenging environment for probes due to the prevailing thermal, mechanical and chemical loads. For the investigation of these flows, non-intrusive measurement techniques were developed at the Institute for Thermodynamics. Besides pointwise techniques, which imply a high measurement effort, integrative measurement methods like Schlieren, emission spectroscopy and interferometry offer a two-dimensional view. Non-intrusive laser based techniques such as Raman spectroscopy [1, 2] and laser-induced fluorescence (LIF) [3] are promising methods, in which LIF offers the advantage of a very high signal-to-noise-ratio and a good spatial resolution. Furthermore, in the context of an arc-heated wind tunnel LIF provides qualitative and quantitative flow field visualization, hence numerical simulations and radiation models can be verified and validated.

Suitable tracer molecules for laser induced fluorescence measurements in high-enthalpy flow using air and CO_2 as test gas are N_2, O_2, NO or CO. A 1-D CO-LIF measurement system was developed at the Institute of Thermodynamics of the Bundeswehr University Munich [4], which is able to determine the rotational temperature of the CO molecule. The first successful measurements were carried out at a stagnation enthalpy of 2.1 MJ/kg. In this manuscript the results for a stagnation enthalpy of up to 3.8 MJ/kg are presented, showing the technical feasibility of the laser-induced fluorescence measurement setup at the plasma wind tunnel, even with the elevated stagnation enthalpy. Taking into account the possible sources of error, a total uncertainty of the temperature values of ± 150 K is estimated conservatively. From the results of this work, it is concluded that the existing test setup in combination with the optical measurement technique CO-LIF and the data reduction by CARF is well suited for the reproduction of the thermochemical environment during entry into Martian atmosphere.

2. Measurement technique

In this work, the CO molecules are excited by means of a laser from the ground state to an upper energy level. The energy levels of possible transitions of the Hopfield-Birge system $B - X(0,0)$ for the CO-molecule are shown in Fig. 1. For the calculation of the Morse potential function, the molecule constants were taken from literature [5, 6].

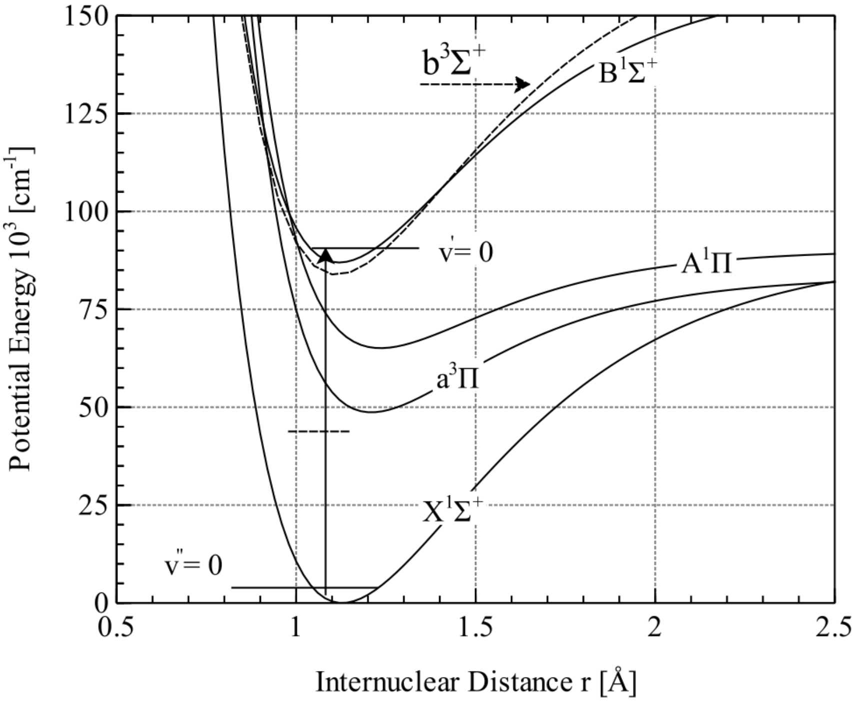

Fig 1. Schematic energy level diagram of the CO molecule

Because of the large energy difference between the ground state and the excited state, photons with a high energy are required for the excitation of the molecules. This implies a light source in the vacuum ultra-violet range, which complicates the experimental work enormously. Alternatively a two-photon process can be used, whereby the molecule is excited by two photons successively. Accordingly the required energy of these photons and their frequency is lower. In order to match the specific energy transitions to excite a molecule and to generate a sufficient number of two-photon excitation events, a tunable light source with a sufficient pulse energy is required.

For a two-photon process, the transfer of the molecule's state to a virtual intermediate state needs to be accounted for. In principle LIF consists of exciting a molecule from a lower to an upper level. The fluorescence signal can thereby be described by a two-level model, Fig. 2. The ground level is named with W_1 and the excited level with W_2. Each level can either be populated or depopulated by optical and collisional processes, which are mainly described by the rate constants b_{12} and b_{21} [5]. For a one-photon process these rate constants are related to the incident laser irradiance I and to the Einstein B coefficients by $b = B\, I\, c^{-1}$, whereas the spontaneous emission is described through the Einstein coefficient A_{21}. The two-photon excitation rate b_{12} is replaced by W_{12} and can be expressed as

$$W_{12} = \frac{\sigma_{2P}\, I^2}{(h\, \omega_{12})^2} \tag{1}$$

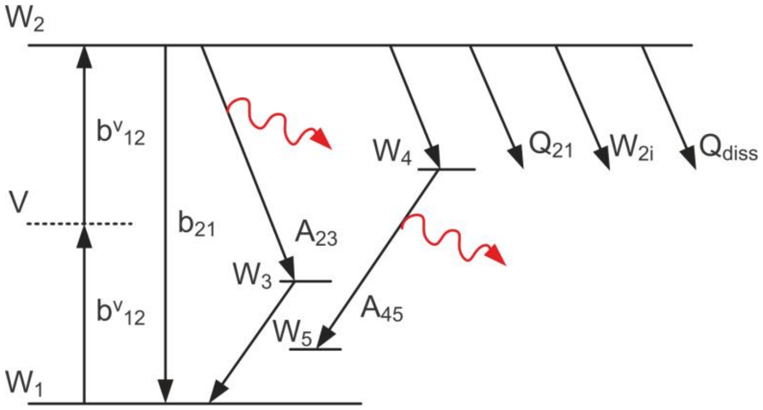

Fig 2. Energy levels of a two-photon excitation process

The strength of the absorption depends on the product of the absorption cross section, σ_{2P} and the square of the irradiance [7]. Fluorescence can occur in different ways, either by an energy transfer of the molecule from a higher level W_2 to the lower level W_3, followed by a non-radiating process, or the by an energy transfer by a collision process to an intermediate state W_4 subsequently followed by a fluorescence process to W_5. Further processes such as collisional quenching and photoionization are represented through the rate constants Q_{21} and W_{2i}, respectively. In the case of an open two level model, the loss of population from the upper level to a predissociative level is expressed by Q_{diss}. The population of the quantum states of the ground level depends on temperature and can be described by the Boltzmann distribution related to the temperature, electronic, vibrational, and rotational quantum numbers.

3. Experimental setup

The arc-heated plasma wind tunnel is equipped with Y-shaped annular electrodes and offers long-duration testing, basically limited only by the amount of available test gas. The electric field between cathode and anode accelerates the electrons leaving the cathode and thermal energy is transferred through collisions with heavy species. In the stagnation chamber, a stagnation temperature in the order of 6000 K can be achieved. In a settling chamber additional gas can be injected, cooling the flow and hence covering a wider enthalpy range. Subsequently, the test gas is accelerated by expansion in a Laval nozzle. To achieve a broad spectrum of Mach numbers in the sub- or supersonic region, different nozzle contours can be used. The test facility is equipped with probes to determine the bulk enthalpy and the stagnation temperature. By using a vacuum pump, the facility is able to produce specific enthalpies and pressures to simulate conditions prevailing during the entry into Martian atmosphere. Because of the dissociation of CO_2, a considerable amount of CO is produced, which may form an explosive mixture inside the high-pressure part of the vacuum pump. To stay below the explosive limits of CO, an additional amount of nitrogen is added close to the exhaust gas for dilution. Assuming a 100% conversion of CO_2 to CO, a dilution ratio for N_2:CO_2 of 5:1 is proposed [8].

For the excitation of the CO molecule in the spectral range from 230.05 nm to 230.11 nm, a laser system consisting of a Nd:YAG (wavelength 355 nm, pulse energy 300 mJ, pulse duration 20 ns, repetition rate 10 Hz) laser and a dye laser (dye coumarin 47, spectral linewidth 0.06 cm^{-1}, pulse energy 1 mJ) is used. The laser beam enters the test section through a fused silica window. The two-photon process requires a high energy density. Therefore, a spherical lens with a focal length of 500 mm focuses the laser beam into the measurement volume enabling a 1-D analysis of the high-enthalpy flow. The focus of the beam is adjusted to the center of the flow. A beam dump at the top of the test section absorbs the laser beam to avoid reflections. The fluorescence signal is collected perpendicular to the axis of the laser beam by an intensified CCD camera and a UV lens (f = 105 mm). The resolution of the camera (1024 px × 1024 px) is reduced by pixel binning (2 px × 2 px) in order to increase the signal-to-noise ratio and the readout rate of the detector leading to a pixel density of 8.6 px/mm. To reduce the influence of laser energy fluctuations and low signal intensity, the detector is exposed 100 times for each image. Furthermore, the accumulation of the data is done before reading out the detector leading to a significantly increased signal-to-noise ratio. A long-pass filter transmitting light from 425 nm to 760 nm is used to reduce reflections of the laser beam and Rayleigh scattering without any considerable weakening of the emission detection following the transition $B^1\Sigma^+ \rightarrow A^1\Pi(0, v'')$. The scanning of the wavelength is realized by an adjustment of the grating of the dye laser during measurements, which requires a permanent lateral alignment of the laser beam by tilting the compensator. However, this process and the aging process of the dye during measurements lead to significant fluctuations of the pulse energy. In order to compensate for these fluctuations, an energy meter is placed in reflection of a fused silica glass (reflection approximately 9 %). The measured signals are then normalized with the recorded energy of each single laser pulse. The Q-switch output of the Nd:YAG laser is used as master of the synchronization. Control of the micro channel plate (MCP) of the camera, synchronization of the laser pulse and the MCP, the dye laser as well as the energy meter is performed with the internal delay generator of the camera. The schematic of the experimental set up is shown in Fig. 3.

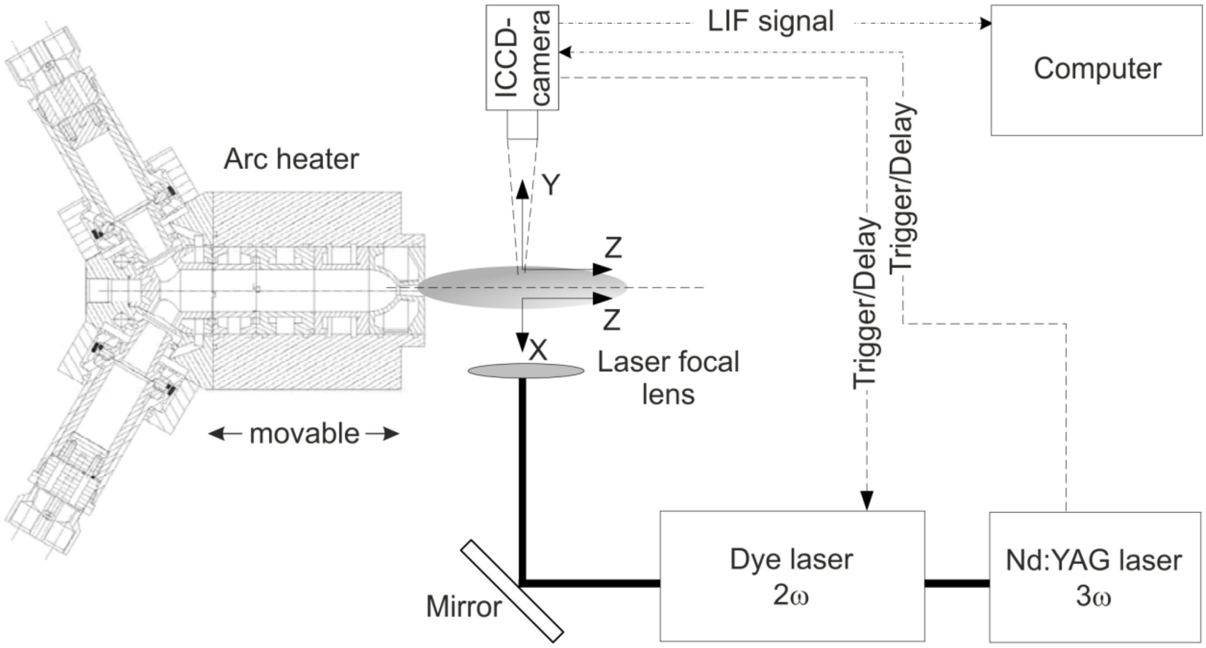

Fig 3. Optical setup for temperature measurements in the arc-heated flow

In contrast to the former works, the operational mode of the ICCD camera was changed to 'integrate-on-chip', i.e. to accumulating the signal before reading out the detector. Avoiding the readout noise of every single frame, the signal-to-noise ratio of the measured spectra could thereby be improved significantly. Besides improving the image quality, this operational mode of the camera enables an independent variation of spectral measurement points and data accumulation, i.e. an increase of measurement points and accumulation number at the same time. This effect will be very helpful for severe measurement conditions with a lower signal intensity.

CARF Method

One possible method to determine the temperature from measured spectra is a cross correlation with simulated spectra, which directly fits the data without prior adoptions. This spectral rotational fitting method is realized with an automated algorithm also called correlation automated rotational fitting (CARF) and is mainly used in physical chemistry [9, 10] and is described in detail in [4, 11].

For the calculation of CO excitation spectra, a tool called NOCO-Spectra has been used, which was developed in [11]. For an optimum fitting process multiple excitation spectra with different temperatures have been simulated in advance. The temperature step width was set to 10 K, which represents the minimum temperature resolution.

The next step requires the measured excitation spectra from the free stream of the plasma wind tunnel. To take into account variations from the laser, the measured spectra are corrected by the laser energy. In addition, the spectra are also corrected by the background noise, which provides a constant baseline for the excitation spectra.

NOCO-Spectra correlates each simulated spectrum with the measured ones. As a result of the comparison, a correlation factor is calculated, considering the used temperature for the simulated spectra. In Fig. 4 the temperature dependent slope of the correlation factor can be seen. The most probable temperature is displayed by the highest correlation factor.

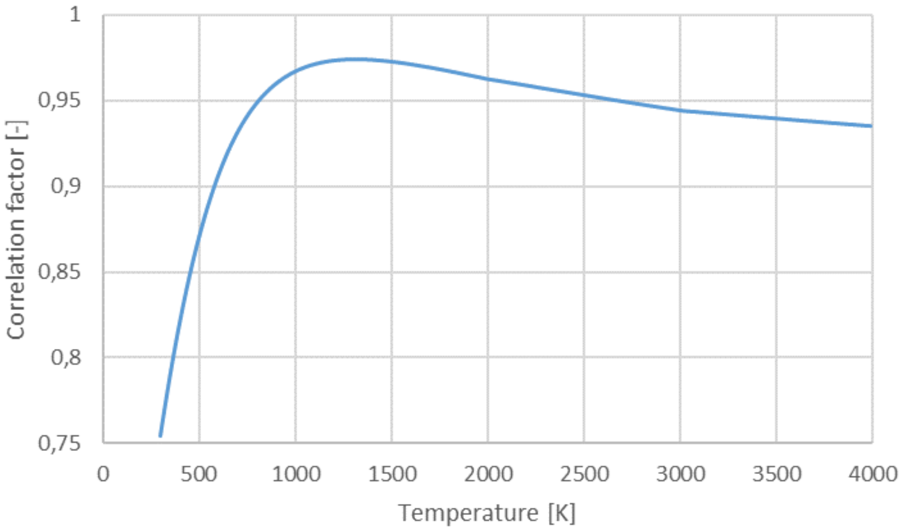

Fig 4. Correlation curve (h_0 = 3.1 MJ/kg, y = 0 mm)

During the measurement process the wavelength is adjusted continuously in order to get excitation spectra. Due to this fact and the variation of the stagnation enthalpy of +/- 12 % within time, the temperature uncertainty is conservatively estimated at ± 150 K.

4. Measurement results

In own former works the described test facility was successfully used to measure the temperature in a CO_2 flow with a stagnation enthalpy of 2.1 MJ/kg [4]. In this work, the measurements were carried out with a total enthalpy of 3.1 MJ/kg and 3.8 MJ/kg. The Laval nozzle expanding the flow to a Mach number of 3.4 at a pressure of 0.3 kPa used here is the same as in [4].

In the first measurement the investigated spectrum extended from 230.07 nm to 230.108 nm, the spectral increment was 0.0005 nm leading to 77 measurement points in the described spectral range. The CO_2 massflow was 2.3 g/s and the stagnation enthalpy 3.8 MJ/kg. The result of this test run is shown in Fig. 5. It can be seen that the temperature of the flow reaches a maximum of approximately 1600 K, which is nearly constant from the center to a radial distance of 6 mm. At a radial position of -8 mm the temperature drops to 1410 K and 1180 K at +8 mm showing a slightly asymmetrical behaviour. A asymmetry also can be seen at -10 mm (720 K) and +10 mm (600 K). At a distance of 15 mm to the center of the flow the temperature symmetrically drops to 350 K.

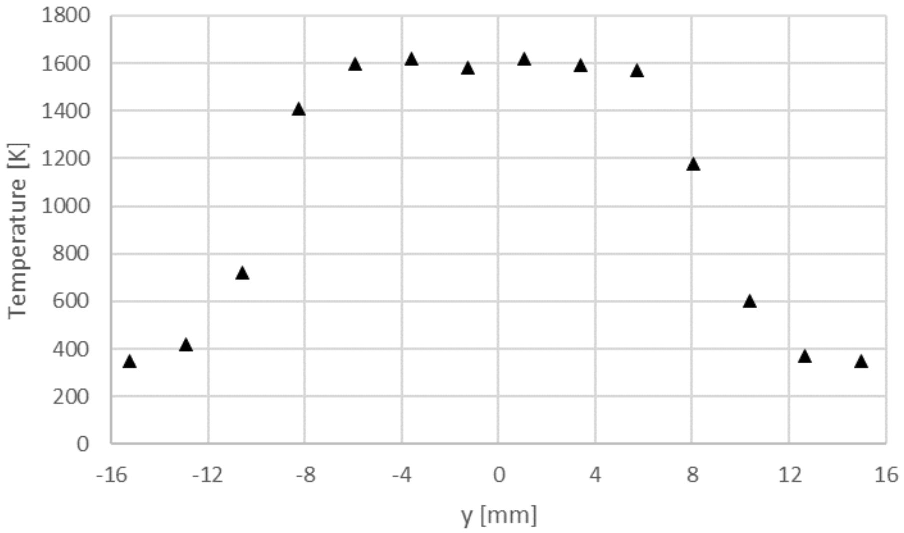

Fig 5. Temperature profile in the arc-heated flow (h_0= 3.8 MJ/kg, x = 10 mm)

For the second measurement the spectral range was increased to 230.05 nm to 230.11 nm. Using the same spectral increment of 0.0005 nm 121 measurement points were scanned. While the CO_2 mass flow was kept constant (2.3 g/s), the stagnation enthalpy was 3.1 MJ/kg. Fig. 6 depicts the temperature profile in the flow of this test run. The temperature shows constant maximum values of approximately 1300 K from the center of the flow to a radial distance of 6 mm, which is in good agreement with the first measurement. According to the lower electrical power, the maximum temperature is about 300 K below the temperature of the first test run. At a radial position of -8 mm the temperature drops to 960 K and 1110 K at +8 mm again showing a slightly asymmetrical behavior. At -10 mm the temperature is 590 K and at +10 mm 700 K. At a radial distance of 15 mm the temperature drops to below 400 K. Note that the asymmetry observed is different in the presented data of both test runs. While in the first data set at a negative radial position higher temperature values are measured, the second data shows higher values at a positive radial position. As this asymmetry also was observed with different test parameters in [4], it is assumed that the reason for this observation is an uncertainty in the evaluation of the data especially at the measurement points with a strong gradient in temperature and not necessarily a systematic asymmetry of the temperature profile of the high-enthalpy flow.

Possible sources of error of the temperature measurement using the presented procedure are the variation of the stagnation enthalpy during the scanning process, which takes about 30 minutes depending on the wavelength increment. In general, the stagnation enthalpy can vary ±12%, which leads to variations in temperature. In addition, reflections of the fluorescence signal on metallic surfaces inside the measurement chamber may lead to signal interferences. Furthermore, even keeping the test conditions of the arc constant, fluctuations of electrical power and of the arc position can lead to temperature differences during the measuring time [3, 14]. Therefore, for a time sequential measurement technique, single peaks within the excitation spectra slightly deviate from the simulated spectra reducing the accuracy of the CARF algorithm.

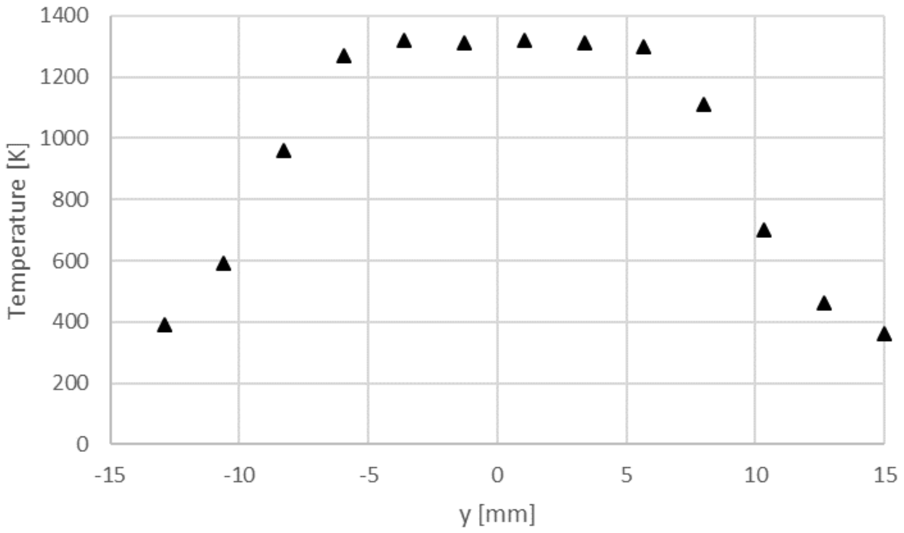

Fig 6. Temperature profile in the arc-heated flow (h_0= 3.1 MJ/kg, x = 10 mm)

5. Conclusions

In this work, spectroscopic investigations of CO in a high-enthalpy flow generated by an arc driven plasma wind tunnel and CO_2 as test gas were conducted. The measurements were carried out with laser-induced fluorescence (LIF) of the CO molecule, enabling non-intrusive measurements of the supersonic flow. As the energy difference between the ground and the excited state of CO is very high, a light source with emission in the vacuum ultraviolet range is required. To avoid the extremely short wavelengths, the approach in this work was to use a two-photon process including a dye laser enabling wavelength scanning and thus the recording of excitation spectra. For the excitation spectra of CO, the Q-branch (B-X) was used. Based on former own investigations with this test bench [3, 4, 11] with a total flow enthalpy of 2.1 MJ/kg, the enthalpy was increased to 3.1 MJ/kg and 3.8 MJ/kg, respectively.

The signal-to-noise ratio of the measured spectra could be improved significantly compared to former measurements by changing the operational mode of the camera. Furthermore, this mode enables an increase of measurement points and accumulation number at the same time. This effect will be very helpful for conditions with a lower signal intensity.

For the simulation of the excitation spectra of CO, the in-house developed tool NOCO-Spectra was used, which enables the simulation of excitation spectra of CO. In the mentioned former investigations, it could be shown that the spectra simulated with NOCO-Spectra agree well with the values from literature and also with spectra measured at a flat flame burner. There a rise in the population of higher rotational quantum numbers of the B-X(0,0) transition with increasing temperature successfully could be shown. This approach for spectra simulation could be used in this work to establish a method for rotational temperature measurements using the Boltzmann distribution. To evaluate the rotational temperature of CO, the calculated spectra were fitted to the measured ones using a correlation automated rotational fitting procedure (CARF).

The total uncertainty of the presented temperature values is conservatively estimated at ± 150 K. It can be concluded that the presented measurement procedure at the plasma wind tunnel and data reduction is well suited for the reproduction of the thermochemical environment during entry into Martian atmosphere.

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