

Gas-phase Thermometry and Heat Flux Measurements during Flame-Wall Interaction using Two-Line Atomic Fluorescence

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ABSTRACT

The feasibility of Two-Line Atomic Fluorescence (TLAF) of indium atoms to measure the gas-phase temperature in near-wall environments is investigated. Different methodologies to retrieve temperature are presented (conventional TLAF, OLAF, calibration-free TLAF) and discussed, aiming for this particular application. The platform AURYGA (Atomic fluorescence platform for thermometry in gases) has been developed to seed a premixed flame with indium atoms using trimethylindium and tunable external cavity diode lasers are used to allow the resolution of the hyperfine spectra of indium. Vertical and horizontal profiles of temperature in a free flame configuration using these methods are compared to thermocouple measurements, showing consistency if the signal-to-noise ratio is sufficient enough.

1. Introduction

Knowledge of temperature is essential in combustion studies. The heat release rate is highly dependent on this parameter, as are the reactions rates, which exhibit exponential behaviour with temperature. Furthermore, a wide range of temperature levels can be encountered in a reactive flow: from 300K in the fresh gases to more than 2000K in the burnt gases, with steep gradients inside the flame front. When a solid material is in contact with a reactive flow, additional processes have to be taken into account: significant temperature differences between the flame (around 1500-2000K) and the wall (below 1000K) are responsible for inefficient combustion (quenching events) and increased pollutants levels (unburnt fuel, CO, NO_x). Note that severe transient and heterogeneous wall heat transfer (up to 1 MW/m²) occur. To prevent from the alteration of the material properties (microstructure, surface roughness, mechanical properties...), thermal management, i.e. cooling of the wall, has to be properly designed. Temperature and heat transfer thus need to be carefully measured to obtain a comprehensive understanding of flame-wall interactions (FWI) (Dreizler & B  hm, 2015), being necessary to allow the optimization of current combustors, which is indispensable in the context of climate change.

Experimental temperature measurements in a confined zone close to the wall is challenging. Conventional intrusive methods such as thermocouple measurements are primarily limited by their size. Laser-based diagnostics can circumvent the drawback of disturbing the flow but still have to offer high spatial resolution. Coherent Anti-Stokes Raman Spectroscopy (CARS) will thus be limited by the minimum space required to cross the laser beams (Padhiary et al., 2024). Unwanted scattering from surfaces can also prevent techniques with collection and excitation at the same wavelengths such as Rayleigh scattering from providing coherent results without appropriate filtering (Brübach et al., 2006). Laser-induced fluorescence (LIF) could also be used on molecules naturally generated by the flame, e.g. OH, but in the case of local extinction, signal-to-noise ratio (SNR) would not be sufficient (Elmnefi et al., 2014). Moreover, molecular LIF is very sensitive to quenching, i.e. the non-radiative de-excitation of the previously laser-excited populations, adding uncertainties in the results.

Two-Line Atomic Fluorescence (TLAF) was first theoretically described by Alkemade (1970) and experimentally demonstrated shortly after (Omenetto et al., 1972). TLAF is based on the measurement of the relative populations of two states of an atom, giving the temperature via the Boltzmann distribution. The occupation of the upper energy level is induced by sequential excitations of two lower lying states by appropriate light sources. Temperature being calculated from a ratio of fluorescence, directly proportional to the population densities of the probed states, this technique is not subject to quenching, making TLAF measurement independent from the ambient gas composition. Besides the possibility of using TLAF in flames with various fuel and oxidizer, the technique has been demonstrated in particle laden environment (Borggren et al., 2018) and seems to be promising close to the wall where a complex chemistry can be observed. The temperature sensitivity is mainly defined by the energy difference ΔE_{21} between the two probed states. Indium is the most often used atom, due to its good sensitivity at elevated temperature levels (700-3000K) (Haraguchi et al., 1977; Dec & Keller, 1986), relevant in combustion processes. Various seeding systems have been reported in the literature, from indium chloride solutions (Chan et al., 2010) to the ablation of metallic indium (Gu et al., 2015). First introduced by Tian et al. (2005) and further developed by Whiddon et al. (2015), seeding a reactive flow using the sublimation of an organometallic substance, e.g. trimethylindium (TMI) for indium seeding, offers several advantages. A stable and perfectly-controlled concentration can be easily achieved inside the flow, atoms being released when TMI decomposes at high temperature (above 600K) (Damas et al., 2023). The change in the flame chemistry is negligible, with only a few ppm of TMI sufficient to induce significant LIF signals.

In this study, a newly built platform to seed indium atoms in flames is presented. Taking advantage of tunable narrow-band lasers, temperature profiles are measured using different approaches based on fluorescence of indium in a free flame configuration, thus allowing a comparison with corrected thermocouple measurements. The idea is to assess the possibility of using TLAF of indium to measure gas-phase temperature close to the wall for the first time to the authors' best

knowledge, aiming to reconstruct the local wall heat flux for in-situ measurements.

2. Detection schemes

Two linear methods are commonly used for temperature measurements using atomic LIF (Zizak et al., 1984). All methods required two excitation wavelengths and two fluorescence signals to be measured. For *conventional TLAF*, collection is performed at wavelengths different from the excitation ones. This first detection scheme is mostly employed in its non-linear or saturated forms with pulsed laser sources, to allow quasi-instantaneous measurements in turbulent environment (Kaminski et al., 1998; Manteghi et al., 2015). Nonetheless, this approach needs to be calibrated. Indeed, the constant C in Eq. (1) is accounting for the detection differences of the two detectors.

$$T = \frac{\Delta E_{21} / k_B}{\ln \left(\frac{F_{451} / I_{13}^0}{F_{410} / I_{23}^0} \right) + 4 \ln \left(\frac{\lambda_{23}}{\lambda_{13}} \right) + C} \quad (1)$$

where ΔE_{21} is the energy gap between the two probed states, k_b the Boltzmann constant, F_{451} and F_{410} respectively Stokes fluorescence (at 451nm) and Anti-Stokes fluorescence (at 410nm), I_{13}^0 and I_{23}^0 the excitation intensities and λ_{13} and λ_{23} the transition wavelength corresponding to transitions from the probed level ($5^2P_{1/2}$ is noted 1 or $5^2P_{3/2}$ is 2) to the upper level ($6^2S_{1/2}$ is 3).

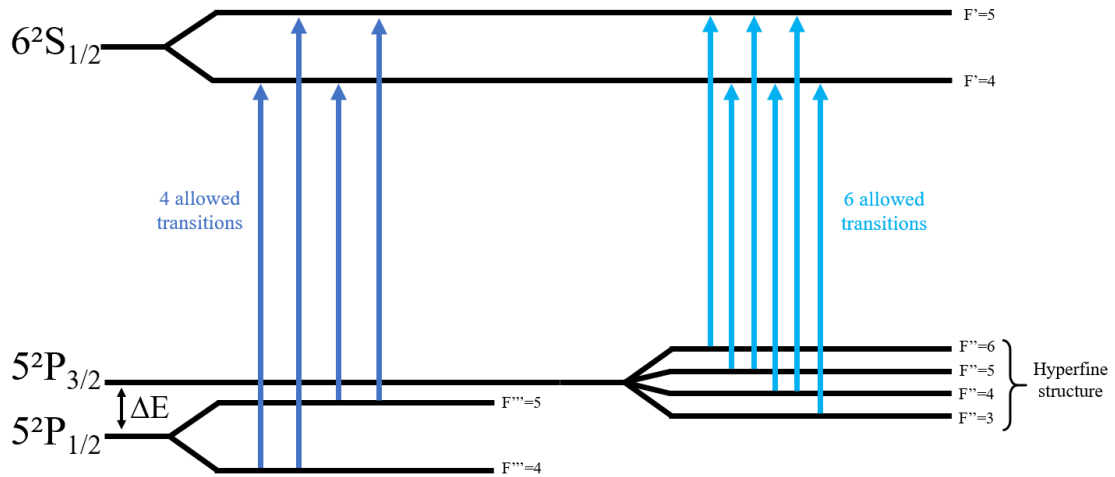


Figure 1. Energy levels and allowed transitions (only excitations are shown) for the indium atom. F is the quantum number due to the coupling of the net orbital angular momentum and the net nuclear spin. Left side corresponds to a transition at $\lambda \approx 410\text{nm}$, right side $\lambda \approx 451\text{ nm}$.

Hult et al. (2005) have introduced a calibration-free methodology using the second scheme, often referred as *calibration-free TLAF*. Both LIF-signals are collected at the same wavelength, avoiding the need of a calibration to compensate for the two detectors discrepancies in efficiency. Non-resonant and resonant fluorescence are therefore recorded, for indium, usually at $\approx 451\text{nm}$. The

technique is based on taking advantage of the arrangement of the energy levels of the atom which are composed of hyperfine structures, as it can be seen in Fig. 1. Hult et al. (2004) had previously shown that temperature could be retrieved from the shape of the hyperfine spectrum from one transition by scanning a narrow-band laser across the absorption profile of the atom. This technique is called One-Line Atomic Fluorescence (OLAF). For TLAf measurements, this scanning operation is performed twice (for the 410nm ($5^2P_{1/2} \rightarrow 6^2S_{1/2}$) and 451nm ($5^2P_{3/2} \rightarrow 6^2S_{1/2}$) transitions) while monitoring the excitation wavelength and laser power. Temperature is thus obtained from Eq. (2) :

$$T = \frac{\Delta E_{21}/k_B}{\ln \left(\frac{\int_0^\infty \frac{F_{nr}(\nu)}{I_{13}(\nu)} d\nu}{\int_0^\infty \frac{F_r(\nu)}{I_{13}(\nu)} d\nu} \right) + \ln \left(\frac{A_{32}}{A_{31}} \right) + 3 \ln \left(\frac{\lambda_{23}}{\lambda_{13}} \right)} \quad (2)$$

where $F_{nr}(\nu)$ and $F_r(\nu)$ are the spectral intensities of the non-resonant (excited at 410nm) and resonant fluorescences (excited at 451nm), A_{32} and A_{31} denote the Einstein coefficients for spontaneous emission for the corresponding transition.

It should be noted that various phenomena like flame luminescence or scattering can interfere with the measured fluorescence signals and hyperfine spectra are usually fitted on experimental data (Burns et al., 2011; Borggren et al., 2017). These theoretical spectra are calculated from the convolution of the Voigt profile of each hyperfine transition where their relative intensities and spacing are calculated from the Clebsch-Gordan coefficients. Indium is not a mononucleic element but (Labus et al., 2022) have shown that the difference induced by taking the isotopic shift into account is negligible at atmospheric conditions since In_{115} is much more abundant than In_{113} (95.71%/4.29%). In this study, only In_{115} will thus be considered.

3. Experimental setup and methodology

3.1. Indium seeding, burner and operating conditions

An experimental platform was designed at CORIA laboratory, named AURYGA (*Atomic fluorescence platform for thermometry in gases*), to seed premixed flames with indium atoms. A schematic and a photograph of the platform are presented in Fig. 2 and Fig. 3 respectively.

Indium atoms are generated by the decomposition of TMI vapour before the flame front at high temperature (see Section 1). A commercial bubbler (Nouryon TMIn SSG) composed of a tube dipping into TMI in its solid form (crystalline powder) is used. TMI having a high vapor pressure (around 230 Pa at 298.15 K) with a melting point of 361.15 K (Gerrard et al., 1992), sublimation is occurring at ambient conditions. A nitrogen flow (less than 1.1 % of the total flow rate) is sent into the bubbler to bring the TMI vapour through a vent to the mixing lines 250 mm before the flame,

and a non-return valve (Hamlet) is added before the injection to prevent any backflow from the main flow. Nitrogen is chosen to prevent TMI to ignite, this compound being pyrophoric. Seven pneumatic valves (RS-V) controlled by a LabView in-house program can bypass the bubbler, allowing to perform a purge procedure before and after every experiment to operate without risk. All traces of moisture and oxygen are eliminated by alternating a nitrogen flow and a vacuum pump. The seeding lines of the platform are monitored by several sensors (pressure and surface thermocouples) and are covered by heating cords to avoid TMI condensation, thus blockage concerns. A safety valve (Swagelok - 5.0 bar gauge) is also included after the bubbler to prevent overpressure in case unwanted combustion in the TMI tank would occur.

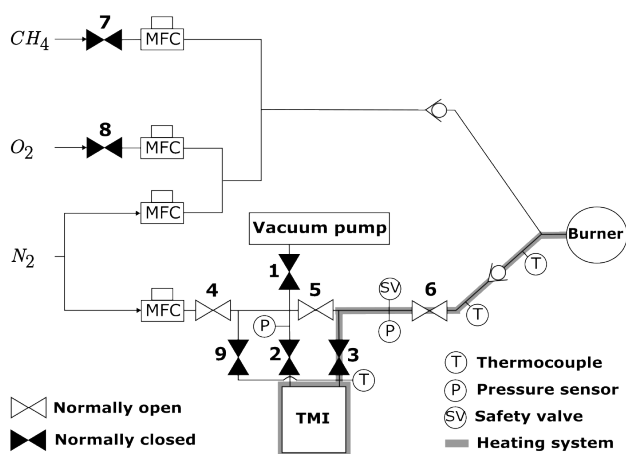


Figure 2. Scheme of the AURYGA platform showing mixing lines and seeding system.

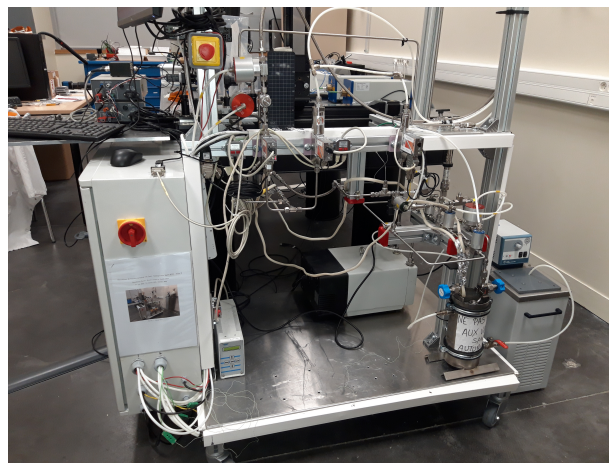


Figure 3. Indium seeding platform developed in CORIA laboratory.

The platform is designed to be able to easily replace the fuel/oxidizer mixture or modify the burner without changing the seeding system. For this study, a Bunsen burner (8 mm in diameter) is used to generate a conical laminar premixed methane/air flame (40 mm in length). Air is reconstructed (O_2 , N_2) to ensure an optimal dryness to avoid reaction of the moisture with TMI and to compensate the added nitrogen flow from the seeding lines. All flow rates are independently controlled with thermal mass flow meters (Bronkhorst) to reach an equivalence ratio of $\phi = 1.25$ (thermal power of 290 W).

3.2. Optical Setup

An optical setup was conceived to excite the indium atoms and collect the resulting fluorescence. A scheme and a photograph of the setup are shown in Fig. 4 and Fig. 5 respectively. Since tunable and narrow-band light sources are needed to perform calibration-free TLAF (see Section 2), external cavity diode lasers (ECDLs) are chosen to generate beams around 410 nm and 451 nm (Sacher Lasertechnik - LION Littman series TEC 500). The laser beams are passed through single-mode

optical fibres into adjustable collimators. After emission, the continuous beams are hashed using a mechanical chopper (Thorlabs MC2000B) rotating at 10 Hz and combined using a dichroic mirror. The wavelength of the lasers can be adjusted using a hex key (corse tuning) or a piezoelectric control (fine tuning) varied thanks to the LabView in-house program. A wavemeter is used for monitoring (High Finesse WS6-600). The incident laser irradiances are recorded on a photodiode (Thorlabs DET100A2), calibrated using a power-meter (Thorlabs S401C). Laser beams are focused above the burner using a focal lens ($f=200\text{m}$), for a focussed diameter of $\sim 300\mu\text{m}$. Indium fluorescence is collected using two spherical lenses ($f_1 = 75\text{ mm}$, $f_2 = 150\text{ mm}$), filtered at 451 nm and recorded on a photomultiplier tube (Hamamatsu). The burner is mounted on optomechanical translations so that the height and width of the measurement point can be adjusted.

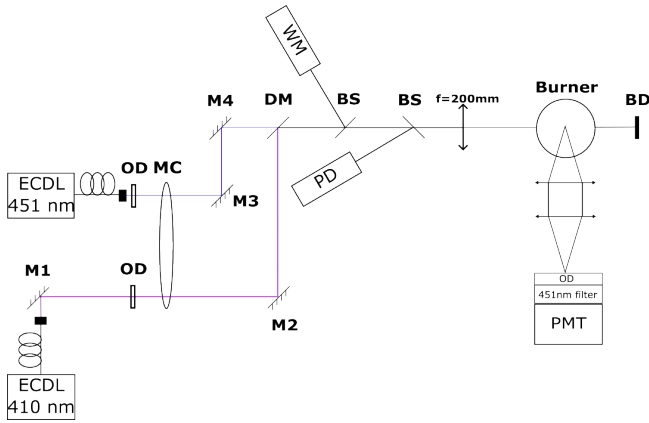


Figure 4. Scheme of the optical setup. ECDL: External Cavity Diode Laser, OD: Optical density, M: mirror, MC: mechanical chopper, DM: dichroic mirror, BS: beam splitter, WM: wavemeter, PD: photodiode, f: focal lens, PMT: photomultiplier tube, BD: beam dump.

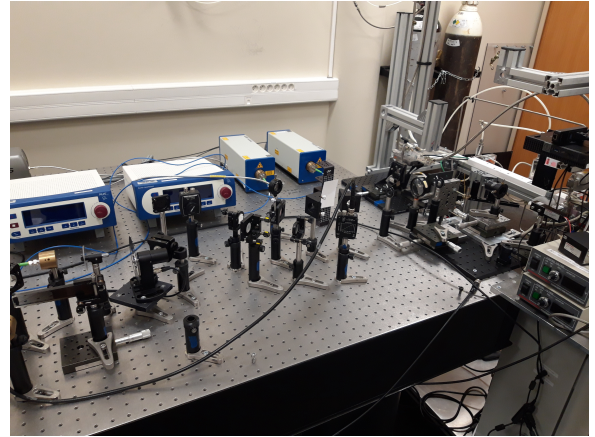


Figure 5. Photograph of the optical setup.

3.3. Temperature determination method

Experimental spectra are obtained by scanning alternately both lasers across the absorption profiles of the indium atoms. The limited mode-hop free range of the ECDLs prevents from having fully resolved spectra and if the background is not taken into account, deviations in the temperature will be observed. Therefore, to evaluate the temperature using calibration-free TLAF, theoretical excitation spectra are fitted on experimental data (least-squared method) and used for the numerical integration in Eq. 2, as described in Sec. 2. The adjustment parameters are the temperature, the intensity, the background, the absolute position of the first hyperfine compound and the spectral broadening (effect from temperature with Gaussian broadening, effect from pressure with Lorentzian broadening). At atmospheric pressure, the effect of both broadening are of the same order of magnitude and a two-step process is necessary to ensure an appropriate determination of

the parameters. In the first step, all the parameters are varied except the Lorentzian broadening. In the second step, only the latter is determined using the temperature previously calculated as an input. The final result is obtained using multiple iterations as proposed by Borggren et al. (2017), but often two iterations were sufficient.

4. Results and discussion

4.1. Verifications: Impact of the Laser Fluence and of the TMI concentration

Both equations presented in Section 2 are only valid in the linear regime of fluorescence. To experimentally characterize the regime induced by each laser, laser fluence is varied by adjusting an optical density to preserve laser stability (injection current of the lasers is kept constant). Fluorescence emission is recorded with ~ 20 ppm of seeded TMI. Fig. 6 shows the fluorescence intensity, with an excitation wavelength at 451 nm. Each point represents a mean of 500 measurements. Linear regime is verified and saturation regime is not reached with our optical setup. Similar trend is obtained for an excitation at 410 nm (not shown).

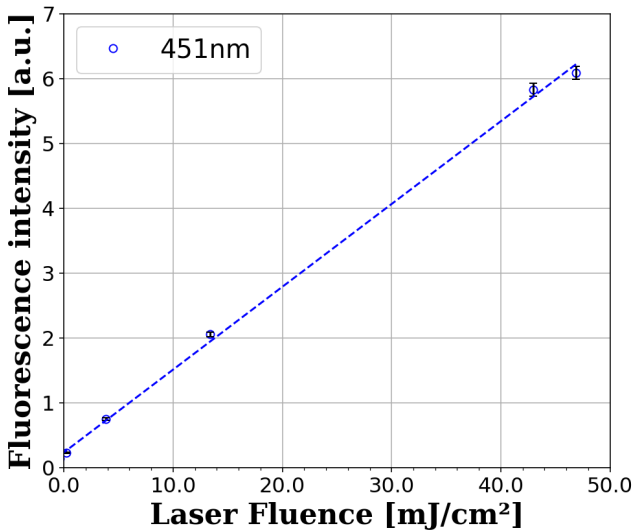


Figure 6. Fluorescence intensity as a function of the laser fluence at 451nm and ~ 20 ppm seeded TMI. Dash line represents linear regression.

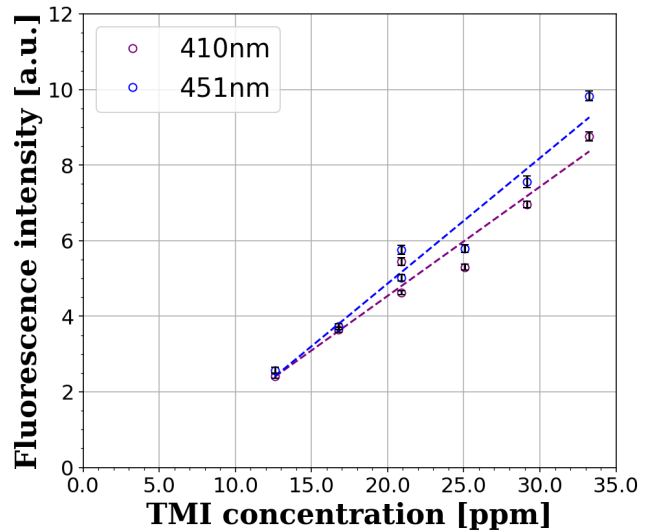


Figure 7. Fluorescence intensities at both wavelength as a function of the TMI concentration. Dash lines represent linear regressions.

Stable and repeatable seeding must also be checked. TMI concentration inside the flame is varied by adjusting the nitrogen flowrate and is calculated assuming the reactive mixture behave like an ideal gas mixture and using Eq. 3 (Whiddon et al., 2015):

$$C_{TMI} = 10^6 \frac{P_{TMI}}{P_{atm}} \frac{Q_{v_c}}{Q_{v_t}} \quad (3)$$

where Q_{v_c} is the volume flow rate of the carrier flow of nitrogen and Q_{v_t} is the total volume flow rate. P_{TMI} , the vapour pressure of TMI in the carrier flow of nitrogen is determined using Eq. 4 (Shenai-Khatkhate et al., 2008):

$$\log(P_{TMI}) = 10.98 - \frac{3204}{T}. \quad (4)$$

where P_{TMI} is in torr, and T is the temperature of the gas (K). Fluorescence intensities for both excitation wavelengths are represented on Fig. 7. Linear regimes can be observed in the test region (12-32 ppm).

4.2. Temperature profiles in free flame

In order to assess our TLAf setup, temperature profiles have been performed in a free flame configuration (without a wall) to be able to compare with thermocouples measurement. Two thermocouples of type K with different diameter (50 and 500 μm) were used to perform a correction of the radiative contribution using the methodology presented by Brohez et al. (2004). A vertical profile, at the flame centre, and a horizontal profile, at HAB=15 mm are presented in Fig. 8. The corrected thermocouple measurements (mean of 1000 points) are shown along with the adiabatic flame temperature for a methane/air reactive flow at $\phi = 1.25$, calculated by the simulation software CANTERA, and the OLAF and TLAf measurements. Each OLAF and TLAf points is the mean of 3 independent series of 50 spectra.

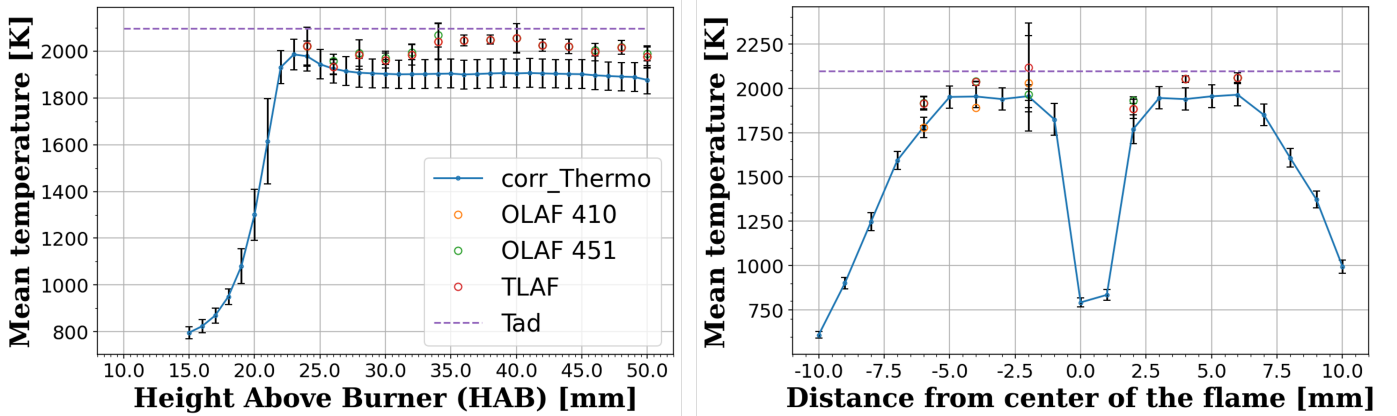


Figure 8. Temperature profiles: vertical variation at the flame centre (on the left), horizontal variation at HAB=15mm (on the right). Corrected thermocouples data are represented by the solid line, OLAF at 410nm, 451nm and TLAf by circles, adiabatic temperature calculated by CANTERA for $\phi = 1.25$ by dash line.

The simulated temperature is the highest temperature, consistent with its adiabatic nature, condition that is never fulfilled in real flames. It should be noted that fluorescence spectra with a too low SNR were removed, recorded signals being insufficient to obtain coherent temperature measurements. The flame front, identified by the steep temperature rise around HAB=20 mm in the

corrected thermocouple measurements, is not resolved with OLAF nor TLAF. Same phenomenon is observed on the edges of the flame for a radius greater than 5 mm. One can note that the point at -2 mm, has a greater standard deviation, probably due to the proximity of the flame front. TMI being decomposed around ~ 600 K, that is inside the flame front, it might not decompose fast enough to ensure enough signal in a close region around it. The three techniques using indium fluorescence collected around 451 nm, i.e. OLAF excited at λ_{13} (410 nm), OLAF at λ_{23} (451 nm) and calibration-free TLAF (made on the same spectra than the OLAF), are consistent with each others. Temperatures obtained are globally about 50-100 K greater than the corrected thermocouple measurements but show the same tendency (plateau). An example of the recorded and fitted spectra for a point at the flame centre and HAB=40 mm is shown in Fig. 9.

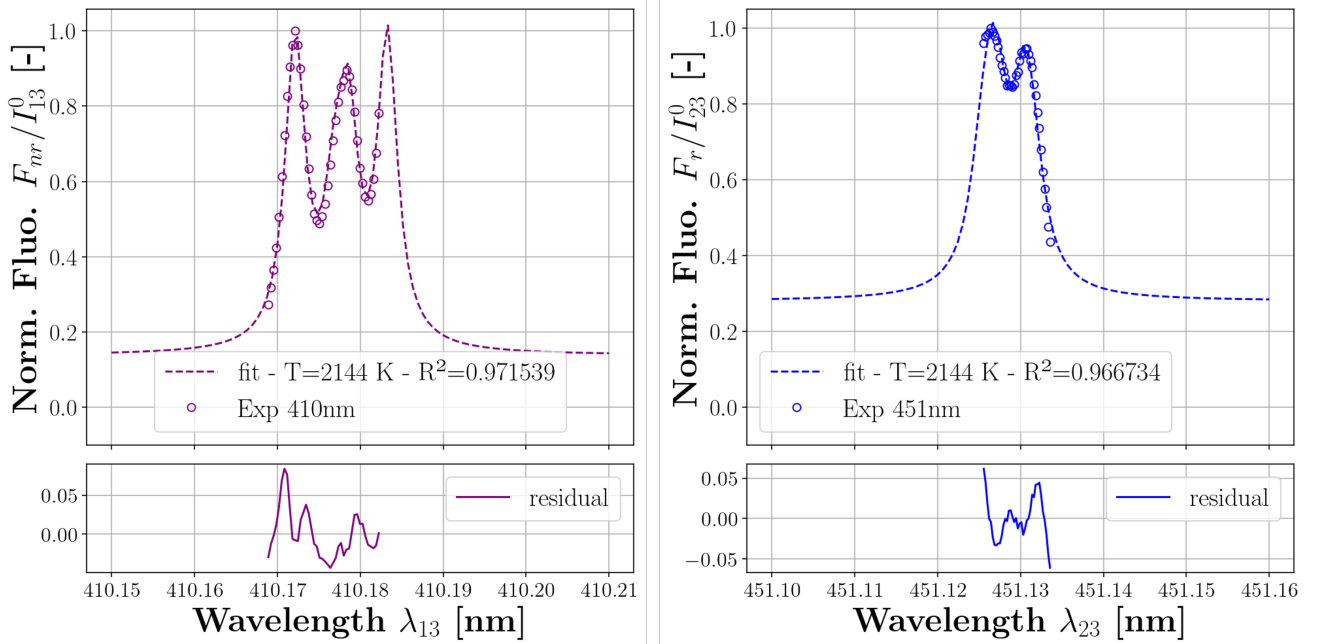


Figure 9. Fluorescence intensity as a function of the excitation wavelength for both transitions ($5^2P_{1/2} \rightarrow 6^2S_{1/2}$ on the left and $5^2P_{3/2} \rightarrow 6^2S_{1/2}$ on the right). Experimental data are represented by dots, fitted theoretical spectra by dash lines. Spectra were recorded at the flame centre at HAB=40mm.

5. Conclusion and perspectives

Calibration-free TLAF of indium has been demonstrated in a free flame (methane/air) configuration and compared to corrected thermocouple measurements (two-thermocouples method). Consistent results are observed between the indium fluorescence methodologies (OLAF at both excitation wavelengths and TLAF) when SNR is sufficient (temperature and indium seeding high enough). The next step in our study is to perform measurements during a flame-wall interaction to assess the possibility of using TLAF in a near-wall configuration for the first time. Once the vertical profile of temperature perpendicular to the plate is measured, heat flux can be calculated

and compared to Gordon gauge measurements. Using the developed methodology, the influence of the burner-wall distance will be studied.

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Nomenclature

A_{ji}	Einstein coefficient of spontaneous emission of the ij transition [s ⁻¹]
C	Calibration constant in the conventional TLAf model [-]
C_{TMI}	Concentration of TMI [ppm]
ΔE_{21}	Energy difference between the two probed states [J]
F_{ij}	Fluorescence intensity of the ij transition [A.U.]
I_{ij}^0	Incident laser irradiance at the ij transition [W/m ²]
k_B	Boltzmann constant [J/K]
λ_{ij}	Excitation wavelength at the ij transition [nm]
ϕ	Equivalence ratio [-]
P_{atm}	Atmospheric pressure [torr]
P_{TMI}	Vapor pressure of TMI [torr]
Q_{vc}	Volume flow rate of the carrier gas [m ³ /s]
Q_{vt}	Total volume flow rate [m ³ /s]
T	Temperature [K]
T_g	Temperature of the carrier gas [K]

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