Mapping of H₂O concentration and temperature in a turbulent methane/air diffusion flame using multispectral absorption tomography

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Abstract

Concentrations of gaseous chemicals species like H_2O , CO_2 together with temperature data are key parameters to characterize different types and phases of combustion processes. Measurements of these parameters can be used in numerical simulations to improve engine design and efficiency. The current paper presents the work on extending Tunable Diode Laser Absorption Spectroscopy (TDLAS) to tomographic measurements in view of two dimensional mapping of concentration (with focus on H_2O) and other thermodynamic parameters (with focus on temperature) in turbulent flames. An adaptation of the Algebraic Reconstruction Technique (ART) have been tested on a methane/air McKenna flame with a scanning TDLAS setup operating at wavelengths around 1.4 μ m on H_2O absorption lines. The theoretical principles are explained in details and some results of experiments are presented and analyzed to determine the accuracy of the TDLAT technique followed by suggestions for improvement.

1. Introduction

The tunable diode laser absorption spectroscopy (TDLAS) is a well-known method which can be helpful to measure chemicals species and thermodynamic data like temperature in a combustion flame. But this technique has no spatial resolution as it gives only integrated results of species density and temperature along the laser line of sight. Resolution can potentially be recovered by tomographic methods exploiting many lines of sight at different locations or angles (called projections) in the probed medium. The method is then called Tunable Diode Laser Absorption Tomography (TDLAT). But because of the few number of optical accesses in most of combustion chambers, this feature is often difficult to implement leading to a limited spatial resolution gain. However, the limited number of projections can be counterbalanced by supplementary spectral information at other wavelengths with a tunable laser.

We are developing a multispectral absorption tomographic spectrometer to probe a turbulent methane/air diffusion flame with several lines of sight, either with separate laser beams or fast spatial scanning of a single beam. Each of the projection spectra contains absorption lines sampled by a few hundreds of wavelengths giving access to pressure, temperature and concentration. Numerical reconstruction methods are applied on these projections to obtain two dimensional mapping of the parameters. The Algebraic Reconstruction Technique (ART) is used and combined with a non-linear optimization algorithm to minimize a cost function proportional to the difference of measured absorptions to the ones simulated with the Beer-Lambert formulation.

2. Tunable diode laser absorption spectroscopy (TDLAS)

The tunable diode laser absorption spectroscopy is based on the Beer-Lambert law which describes, for a particular range of wave number, the attenuation of laser beam intensity through a region of interest (ROI) as a function of the density of chemicals species, temperature and pressure. Let us consider a slice of a flame for a TDLAS measurement where temperature and H_2O density are searched. These two parameters have usually non-uniform distributions over the flame cross-section. For tomographic processing, this slice is discretized into a 2D matrix of pixels as showed in Figure 1.

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Figure 1 : Diagram representing general TDLAT principle

Following the Beer-Lambert law, the expression of the transmitted laser intensity after crossing the ROI is given by:

$$I(\sigma) = I_0(\sigma) \exp\left(-\sum_{kr} \int S(\sigma_{krl}, T_l) \cdot X_{kl} \cdot f(\sigma - \sigma_{krl}, P, T_l) dl\right)$$
(1)

where σ is the wave number of the laser varying from $\sigma_1 \text{to } \sigma_N$, N is the total number of wave-numbers, k the H_2O isotope index, r the absorption line index, $l = 1 \dots L$ the index of the pixel crossed by laser beam with L the total number of pixels on a line of sight. $I(\sigma)$ is the transmitted intensity after the ROI, $I_0(\sigma)$ the incident intensity on the ROI, σ_{krl} the wave number (cm^{-1}) of the absorption line r of the H_2O k isotope present in the pixel l. The function f is the Voigt profile characteristic of the absorption line shape which is a function of the pressure P (assumed to be constant and equal to atmospheric pressure $P_{atm} = 0.1013 MPa$ all over the ROI) and of the temperature T_l along the line of sight. The parameter S is the line strength which has dependency on the temperature T_l and X_{kl} is the density of the isotope k of H_2O in the pixel l. For more practical notation, the different parameters will be noted as follow: $f(\sigma - \sigma_{krl}, P, T_l) = f_{krl}$ and $S(\sigma_{krl}, T_l) = S_{krl}$, etc.

The line strength [1] is given by the equation (2):

$$S_{krl}(T_l) = S_{krl}(T_0) \cdot \frac{Q_k(T_0)}{Q_k(T_l)} \cdot \frac{1 - \exp\left(-\frac{hc}{k_B} \cdot \frac{\sigma_{krl}}{T_l}\right)}{1 - \exp\left(-\frac{hc}{k_B} \cdot \frac{\sigma_{krl}}{T_0}\right)} \cdot \exp\left(-\frac{hc}{k_B} E_{kr}\left(\frac{1}{T_l} - \frac{1}{T_0}\right)\right)$$
(2)

with Q_k the partition function of the H_2O isotope k and r an index identifying the specific absorption line. The other parameters are T_l the temperature at index l in the ROI, $T_0 = 296K$, h the Planck constant, c the velocity of light, k_B the Boltzmann constant, E_{kr} the internal energy of line r for the isotope k. Values for S(TO) and Q are given for the H_2O molecule in the HITRAN database [2]. The Voigt profile is the convolution of a Lorentzian profile by a Doppler profile. For computation the numerical approximation of Hui et al is used [3]. In equation (1) the product inside the integral can be considered as a variable of the position in the l^{th} pixel. It can also be noted as the absorption coefficient with expression:

$$\alpha_l(\sigma) = \sum_{kr} S_{krl} \cdot X_{kl} \cdot f_{krl}$$
(3)

An absorption spectrum is obtained by measuring several ratios of transmitted and incident intensities on a wave-number range. Inversion of this spectrum consists of comparing it to spectra simulated with equation (1) and using a non-linear least square minimization in order to determine integrated temperature and H_2O density (Figure 2). The criterion C for this minimization is formulated as:

$$C = \left\| \overline{p_m(\sigma)} - \overline{p_c(\sigma, T_{rec}, X_{rec})} - \overline{J(T_{rec}, X_{rec})} \delta \vec{F} \right\|^2$$
(4)

where $\overline{p_m(\sigma)}$ is the measured absorption spectrum, $\overline{p_c(\sigma, T_{rec}, X_{rec})}$ is the computed absorption spectrum using the HITRAN coefficients and parametrized by the reconstructed temperature T_{rec} and the H_2O density X_{rec} along the line of sight. The Jacobian matrix $\overline{J(T_{rec}, X_{rec})}$ of the computed spectrum is constructed with the parameters T_{rec} and X_{rec} . The vector $\overline{\delta F} = (dT_{rec}, dX_{rec})$ contains the two steps for the variations of T_{rec} and X_{rec} determined by the minimization of the criterion C. Figure 2 presents an example of the best fit spectrum compared to its experimental counterpart. The chosen steps for temperature and density to calculate Jacobian matrix are 0,1 K and 1,0.10¹⁵ cm⁻³.



Figure 2 : Measured and fitted H_2O absorption spectrum through a flame



3. Experimental device

Figure 3 : Experimental TDLAS setup to probe McKenna burner

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The experimental device used to do this kind of measurements is constituted by three principal parts (Figure 3). All of them are connected by fiber to a laser diode that produce infrared light which can be finely tuned between 7179,5 cm^{-1} and 7183,5 cm^{-1} . A first fiber takes 10 % of emitted light. These 10% are then divided in two others parts in order to provide two particular spectrums used to calibrate the spectral resolution. The first part crosses a reference cell of 10 cm length, containing H_2O vapor at the pressure of 10 kPa at the temperature of 300 K. The diode emission is tuned to provide an absorption spectrum with well identified H_2O from the HITRAN database. In the second part, the light passes through a fibered Fabry-Perot cavity to produce a spectrum with an interference pattern (Figure 4). The peaks are separated in wave-number by a known value from cavity properties. Absolute spectral calibration can be obtained from combination of these spectral markers with the reference H₂O lines from the other spectrum.



Figure 4 : Spectral calibration

The third part of the experimental device takes 90% of signal from the laser source for TDLAT measurements in the flame. Optical fibers coupled to off-axis collimators are used to make the laser beam cross the flow. The two collimators on the flow side are fixed on a translation rail enabling a spatial scanning of the laser beam across the flame (Figure 5).





(a)

(b)

Figure 5 : McKenna Burner studied by TDLAS method : (a) optical device and (b) shape of the flame with O_2 flow of 20mL/min, CH_4 flow of 2mL/min and with a N_2 guard around the flame of 15mL/min

4. Tomographic measurements: Algebraic Reconstruction Technique (ART)

The algebraic reconstruction technique is a numerical tomographic method which consists of reconstructing a two-dimensional function from limited projections. Figure 6 shows the discretization of the absorption coefficients α_1 in the ROI at a given wave-number σ . Each pixel is supposed to contain a uniform value α_l with $l = 1 \dots L^2$. This ROI is crossed by different lasers beams at different positions and angles. Each laser beam provides an integrated projection of the contributions of the absorption coefficient in each pixel l crossed by this beam. These projections are indexed p_i with $j = 1 \dots 2L$ the index of the projection.



Figure 6 : Discretization of the ROI

To calculate a projection, the method consists of using a matrix noted W and called a system matrix with coefficients indexed w_{jl} corresponding to the intersection length of the beams j with the pixel l (Figure 7).



The projections of the absorption coefficients are expressed as:

$$p_j = w_{jl} \alpha_l \tag{5}$$

A tomographic processing consists of recovering the α_l on each pixel from acquired projections p_j with a series of lasers beams at different angles and the matrix w_{jl} corresponding to the geometry of the system. A least square minimization equation:

$$\left\|p_{i} - w_{il}\alpha_{l}\right\|^{2} = 0$$
(6)

is used to come to the best fits for the α_l . This is the basic ART method created by **Kaczmarz** and is also a typical case of inverse problem [4].

5. Application to Tunable diode laser absorption tomography (TDLAT)

5. a. Calculation of multiple projected spectra

In the case of a TDLAT measurement, a projection is not a single point but a function of the wavenumber $p_l(\sigma)$ as showed in Figure 8.



Figure 8 : Scanning of the ROI and acquisition of projected spectrums

The principle of the TDLAT method is to combine the TDLAS spectral inversion with the ART. In other words, the objective is to fit simultaneously every spectra acquire through the ROI by computing local values of temperature T_l^{rec} and H_2O density X_l^{rec} . This requires in turn the computation of the projection of the absorption coefficient for each wave-number σ . If the ROI is discretized as showed in Figure 1, it is possible to compute in each pixel the absorption coefficients $\alpha_l(\sigma)$ according to equation (3). The results have the shape of the Figure 9.

$\alpha_1(\sigma_1)$	$\alpha_2(\sigma_1)$	$\alpha_3(\sigma_1)$		$\alpha_L(\sigma_1$)		
$\alpha_{L+1}(\sigma_1)$	$\alpha_1(\sigma_2)$	$\alpha_2(\sigma_2)$	$\alpha_3(\sigma_3)$			$\alpha_L(\sigma_3)$	
	α _{L+1} (σ ₂)	α1(σΝ) α ₂ (σ ₁	_N) α ₃ (σ	N)		$\alpha_L(\sigma_N)$
:		α _{L+1} (σ	$\alpha_{L+2}(\alpha_{L+2})$	σ_N) α_{L+3}	(σ _N)		$\alpha_{2L}(\sigma_N)$
$\alpha_{L(L-1)+1}(\sigma_1)$	$\alpha_{L(L-1)+1}(\sigma_2)$						
N maps		$\alpha_{L(L-1)+1}($	$\sigma_N \alpha_{L(L-1)+2}$	(σ _N)	••••		$\alpha_{LL}(\sigma_N)$

Figure 9 : Computed of absorbance σ_l for each wave-number σ from each T_l and X_l

Once this absorption coefficient function of the wave number is calculated for each pixel, projected spectra can be computed by integration of these functions along the line of sight of each beam. It is done by using the projection matrix w_{il} for each wave number as showed in equation (7):

$$p_j(\sigma) = w_{jl}\alpha_l(\sigma)$$
⁽⁷⁾

With $p_j(\sigma)$ the projection spectrum of the beam j, w_{jl} the projection matrix with coefficients indexed w_{jl} corresponding to the intersection length of the beams j with the pixel l (Figure 7) and $\alpha_l(\sigma)$ the absorption coefficient for the pixel l as a function of σ .

5. b. Global fitting of projection spectrum

This method is based on coupling **direct absorption spectroscopy with non-linear tomography** [5]. The principle is to acquire several projection spectrums around the ROI. Then the same spectrums at the same angles and positions are computed numerically and fitted with the measured spectrums by global adjustments of the temperature and H_2O density in every pixel simultaneously. The calculation principle is so to minimize the criterion C_s :

$$C_{S} = \left\|\overline{P_{j}(\sigma)} - \overline{W}.\overline{\alpha_{l}(\sigma,\overline{\theta_{kl}^{rec}})} - \overline{J(\overline{\theta_{kl}^{rec}})}\overline{\delta\theta_{kl}}\right\|^{2}$$
(8)

with $\overline{P_j(\sigma)}$ the projections spectrums obtain experimentally, $\overline{\theta_{kl}^{rec}} = (\overline{T_l^{rec}}, \overline{X_{kl}^{rec}})$ a two dimensional matrix containing all reconstructed temperatures and densities in the pixels of the ROI, $\overline{\alpha_l(\sigma, \overline{\theta_{kl}^{rec}})}$ the reconstructed absorbance coefficients in all pixels of the ROI, \overline{W} the system matrix equivalent to w_{ji} used to calculate the projection of $\overline{\alpha_l(\sigma, \overline{\theta_{kl}^{rec}})}$, $\overline{J(\overline{\theta_{kl}^{rec}})}$ the Jacobian matrix of \overline{W} . $\overline{\alpha_l(\sigma, \overline{\theta_{kl}^{rec}})}$ in relation to θ_{kl}^{rec} , $\overline{\delta\theta_{kl}}$ the variations of $\overline{\theta_{kl}^{rec}}$ calculated to minimize the criterion C_s .

During this study, two kinds of ROI geometry have been used. The first is a square ROI as showed in Figure 8. This kind of geometry needs a lot of projections at different angles to provide correct results. However, because of the circular symmetry of the flame studied experimentally (McKenna burner) a circular geometry constituted of concentric rings can also be used as showed in Figure 10. Tomographic reconstruction is nearly the same as the one for a square ROI using equation (8), the difference being in the building of the system matrix \overline{W} now calculated with intersections of the laser beams and rings at a chosen step. An advantage of this geometry is that only one scan on the half of the flame is needed.



Figure 10 : Calculation of the system matrix with $l = 1 \dots L$ the index of a slice and the index of a laser beam position and L the total number of slices and of laser beams

These two geometries have been tested in simulations and both of them have their particularity that influences the results.

6. Simulations results

6. a. Square region of interest

In this case, a first simulation has been made by using a lot of different projections at different angles. Let's consider a grid of 5 by 5 pixels. A two-dimensional profile of temperature and H_2O density is simulated as shown in Figure 12 a and c. The beams used to obtain absorption spectra are simulated as 5 parallels beams placed at 12 different angles around the ROI from 0° to 180° (Figure 11).



Figure 11 : Examples of projections simulated between 0° and 180°

An absorption spectrum is computed with equation (7) for each beam and the tomographic reconstruction by global fitting of equation (8) gives the results presented in Figure 12 b and d.



Figure 12 : (a) simulated and (b) rebuilt temperature, (c) simulated and (d) rebuilt H_2O profile

This reconstruction leads to rebuilt temperatures and densities that are really underestimated when compared to the initial simulated ones. However, the shapes are recovered. The projected spectra with the recovered maps are also nearly identical to the projected spectra claculated with the initial maps as shown in Figure 13.



Figure 13 : Simulated absorption spectra for the angle 0° and fitted spectra (projected spectra from simulations in blue and fitted spectra in green)

These results show that this tomographic technique allows more than one minimum for the criterion C_S meaning more one possible reconstruction for density and temperature mapping from a same series of projections. This phenomenon is more obvious if fewer angles are used. These results show that for a squared ROI it is necessary to have a lot of projections angles. However this setup is difficult to implement because of the lack of spatial access used combustion bench.

6. b. Circular region of interest

In a case of a flame with a circular symmetry, the ROI is constituted by concentric rings. The global fitting is sufficient on only half of the ROI. Simulations results are shown in Figure 14.





Figure 14 : Reconstruction results on circular region of interest of (a) H_2O density and (b) temperature

These results show that rebuilding and accuracy are better with circular geometry than with square geometry repetition. We used this method on our experimental setup described above.

7. Experimental results and discussion

For experimental constraints, the scanning of the McKenna burner is performed in only one direction but the stability of this kind of burner allows the hypothesis of circular geometry for the ROI repetition. In the configuration of gas flow as shown in Figure 5, the expected temperature is a profile approximatively constant inside the flame at around 2000 *K*. The density is expected to have the same shape and a constant value around $1,0.10^{19}cm^{-3}$.

The measurements come from 11 parallel beams along a half part of the flame and the distance between two beams is $0.3 \ cm$. These density and temperature reconstructions are showed in Figure 15. The results are presented for one cross-section of the flame at the height of 2 mm from the burner surface and as a function of the radius between 0 cm and 3,0 cm. A small part outside the flame is also shown for radius between 3.0 cm and 3.5 cm.



Figure 15 : Experimental results of (a) and (b) rebuilt H_2O density in cm^{-3} along the radius of the flame and (c) and (d) rebuilt temperature in K along the radius of the flame

The H_2O density values are around the mean value of $8,0.10^{17}cm^{-3}$ and the mean value of the temperature is around 2000 K inside the flame. The temperature is close to the expected value for this kind of burner, but this is not the case for H_2O density. Moreover, the algorithm is not really robust because of too many varying parameters and fixing very restrictive boundaries is required to attain convergence. The symmetry and the shape of the flame provide *a priori* information to determine some boundaries. For example, for the temperature, the boundaries were limited between 300 K and 800 K outside the flame and between 1800 K and 2200 K for the rings inside the flame. But very often, the results of temperature are systematically locked on one of these boundaries, showing the difficulties for this algorithm to converge to the best solution. However, the solutions found for the H_2O density are not locked on boundaries which are $3,0.10^{17} cm^{-3}$ and $1,0.10^{19} cm^{-3}$, but it is not as regular as expected for this type of burner. A representation of acquired spectra and fitted spectra along the scan is showed in on figure 16. These graphics show that the rebuilt temperature and H_2O density provide spectra relatively close to the experimental ones.



Figure 16 : Measured spectra and fitted spectra along the scan of the ROI for the 11 positions y_l of the laser beam with $l = 1 \dots 11$

These results need to be improved and different ways are now being considered. Indeed, even if the fitting is realized directly on absorption spectrums, several solutions can exist for the inverse problem. One way to choose the best solution is to impose boundary limits for the values to recover on each ring or in zones where a-priori knowledge is available (temperature outside the flame, etc.). Another way is through a method described in reference [5], where the minimization of the criterion C_S is made using a simulated annealing algorithm to select the best minimum to C_S .

8. Conclusion and perspectives

This first work on the TDLAT method has given some results that show that it is possible to obtain spatial resolution with absorption spectroscopy measurements. Noisy spectra and cold temperature intense absorption lines outside the combustion flow have brought perturbations and occultation on the lines used to probe the flow and thus lead to results with poor accuracy. Improvements are now being sought through a larger number of projections (more laser beams and more viewing angles) to come to better spatial resolution. Absorption lines with higher variability to temperature as well as with higher sensitivity will also be helpful. A setup in the mid infrared to operate on H_2O and CO_2 absorbing lines offering these advantages will be shortly used in our next experiments. Inclusion of pressure small variations in the spectrum inversion is also expected to improve recovery of spatial resolution for the sought aerothermal parameters.

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