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The use of molecular spectra simulation for diagnostics of reactive flows

Abstract: The C_2^* radical is used as a system probe tool to the reactive flow diagnostic, and it was chosen due to its large occurrence in plasma and combustion in aeronautics and aerospace applications. The rotational temperatures of C_2^* species were determined by the comparison between experimental and theoretical data. The simulation code was developed by the authors, using C++ language and the object oriented paradigm, and it includes a set of new tools that increase the efficacy of the C_2^* probe to determine the rotational temperature of the system. A brute force approach for the determination of spectral parameters was adopted in this version of the computer code. The statistical parameter c^2 was used as an objective criterion to determine the better match of experimental and synthesized spectra. The results showed that the program works even with low-quality experimental data, typically collected from in situ airborne compact apparatus. The technique was applied to flames of a Bunsen burner, and the rotational temperature of ca. 2100 K was calculated.

Keywords: Optical emission spectra, Computerized simulation, Combustion control, Computer programs, Rotational spectra.

INTRODUCTION

Combustion processes and plasma formation are inherent phenomena of the aerospace technology. Operation of propulsion artifacts, like gas turbine and rocket engine, and shock-wave formation, as observed in hypersonic flight and re-entrance procedures, are the most common examples of this association. In general, both systems constitute a high-temperature environment, with electrons and ionized species produced by a complex chemistry and a time-varying turbulent flow field (Zel'dovich and Raizer, 2002). The measurement of any basic system data, such as temperature and concentration of major species and intermediated radicals and flow velocities, requires the employment of specific diagnostic techniques.

For some kinds of combustion systems, which includes aeronautic and aerospace applications, the use of conventional probes, such as thermocouples and gas analyzers, for combustion diagnostic is not possible or can cause structural

problem and aerodynamic instability, disturbing the system (Gregory, 2005). In these cases, the reliability of measurements is affected by the high-temperature observed in these flames and by perturbations in the flow stability caused by probe insertion. As alternative, these systems can be monitored by using nonintrusive methods, especially those based on optical properties, i.e., in the absorption or emission of radiation of the system (Docquier and Candel, 2002). Among them, the natural or spontaneous emission spectroscopy of radical species shows some remarkable advantages, as it only needs a simple, low-cost, and compact experimental apparatus. These features point to the use of the emission spectroscopy as an important and powerful tool to support active control of combustion processes, where a real time and *in situ* system monitoring is required (Ballester and Garcia-Armingol, 2010).

Emission of flames is due to chemical reactions that produce intermediate chemical species, like, for example, free radicals in excited states. The radiation emission of these radicals is known as chemiluminescence. In hydrocarbon fuel flames, the most intense emissions are associated with C_2^*, CH^* e OH^* species (Durie, 1952; Kane and Broida, 1953). The asterisk denotes the electronic state associated

with each molecule. Apart from spectroscopic constants and experimental factors, the intensity of radical emission spectra is related to the system temperature. This relation is expressed by the Boltzmann's equation, which is given by:

$$I = CS_{J'J''}\lambda^{-4} \exp\left(\frac{-E_{J'}}{kT}\right) \qquad (1)$$

where $S_{J'J''}$ is the line strength of a transition from the upper (J') to the lower (J'') rotational state of the excited species; $E_{J'}$ is the rotational energy of the upper rovibrational level; C is a proportionality constant related to experimental factors, like detector and diffraction grating responses, which attenuate or maximize the emission signal; λ is the line wavelength associated with the transition; and k is the Boltzmann factor (Herzberg, 1950). Thus, the challenge of combustion diagnosis is to establish a consistent relation between microscopic features of the system, spontaneous emission of chemical species generated by fuel burning, and macroscopic property, e.g., its kinetics temperature, which is the most common temperature concept adopted to characterize a flame.

Considering that the emission spectrum follows the Boltzmann distribution given by Eq. 1, a plot of the natural logarithm of line intensities *versus* the energy term shows a straight line whose slope is the inverse of the rotational temperature. In some cases, the rotational temperature of the intermediate species of hot gases is very close to the kinetic temperature, because the translational and rotational energies of a molecule equilibrate within a trajectory of a few mean free path length by collisional processes with other species (Lapworth, 1974). In nonequilibrium systems, as in hypersonic flows (Fujita et al., 2002; Ivanov et al., 2008; Tsuboi; Matsumoto, 2006), supersonic combustion (Do et al., 2008), or plasmas generated by spacecraft during reentry (Blackwell et al., 1997), the various forms of energy are not in equilibrium, and there is not one temperature value able to describe all physical chemistry processes in course (Reif et al., 1973). In these cases, rotational temperature is useful to provide information about chemical reactivity of the medium and produced thermal energy (Acquaviva, 2004).

A direct and simple method to determine the rotational temperature is the comparison of experimental spectra with a synthetic one. The latter can be computed by applying Eq. 1 to each rotational transition belonging to a specific natural emission band. The final spectrum profile is synthesized by the convolution of every spectral line using, for example, a Gaussian-type function. Most of the emitted chemiluminescent is due to intermediated radicals, generally diatomic molecules. Herein, the calculations are relatively simple, as the spectroscopy parameters are well-known experimentally. However, there are some mathematical aspects, as the number of points chosen for

theoretical calculation, which can generate nondesirable artifacts, as spectral line losses.

The aim of this paper is to present molecular spectra simulation as a combustion and plasma diagnostic tool. The species chosen as probe for rotational temperature determination was the C_2^* excited radical. This intermediated reaction product is largely present in all combustion processes containing fossil fuel. It is also present in plasmas formed by high-temperature exposure and ion irradiation of carbonic composites, which are materials with several aerospace applications (Arepalli et al., 2000; Paulmier et al., 2001).

SPECTRA SIMULATION

The computer code for spectra simulation used in this paper was developed by the authors using the C++ language and the object oriented paradigm, and it follows the procedure presented in Pellerin et al. (1996) and Acquaviva (2004) to synthesize the spectra. The computer code allows the visualization of the experimental spectra and of the theoretical one, of the computed emission lines, of both wavelength and intensity, and of the relative contribution of branches P, Q, and R for each C_2^* band. The comparison of theoretical and experimental spectra can be done by visual inspection, with both spectra superposed in one graph and showing the discrepancy between them for each wavelength in a second plot.

The theoretical spectrum curves are obtained following the steps: (i) calculation of the spectral lines position; (ii) determination of its relative intensities and finally (iii) application of a Gaussian-type broadening factor. The first step is carried out using spectroscopic data taken from literature (Pellerin et al., 1996). The two latter steps are dependent of the rotational temperature and total spectral resolution, respectively.

The spectra simulator allows the computation of a single spectrum from the temperature and resolution, and the solution of the inverse problem, determining an optimum pair (temperature, resolution) that minimizes differences between the simulated and experimental spectra. A brute force approach is adopted in this version of the computer code. A range for these two parameters is defined by the user, as well as the variation step of each of them. The implementation explores the multicore characteristic of the modern processors in order to determine both temperature and resolution in a very short time.

The number of computer cores is detected automatically by the computer program and an adequate number of threads is launched, parallelizing coarse grain computations. From these data, synthesized spectra for each pair combination,

temperature/resolution, is iteratively computed. Basically, each set of input data in the ranges defined by the user is used to simulate a spectrum. Every synthesized spectrum is compared with the experimental one to choose the synthesized spectrum that better matches the experimental one. The quality of each synthesized spectrum is defined by comparison with the experimental one, using the chi-squared statistical parameter (χ^2) as a figure of merit. The computer code adopts the full wavelength range as default, but this can be a bad choice if several bands are under consideration or if the contribution of other radicals is important in the experimental spectrum. The user can define a particular range of wavelength, for instance, a region where only a given band of a molecule contributes appreciably to the spectra, as basis for the chi-squared computation in order to reduce the influence of these factors and improve the parameter determination.

These tools are fundamental to reduce the error usually associated in literature with the determination of temperature. Although a detailed description of the tools implemented in our computer code is out of the scope of this work, some few special characteristics are presented when needed in the Results section.

SPECTRA ACQUISITION

Bunsen-type flames were investigated to illustrate employment and capability of the theoretical simulation to obtain the temperature related to the experimental spectra. The fuel burned was liquefied petroleum gas (LPG). The optical system consisted on a TRIAX 550 (Jobin Yvon) monochromator of 0.5 m focal length (f), equipped with a 1.200 lines.mm^{-1} diffraction grating, with blaze at 500 nm. The instrumental resolutions ($\Delta\lambda_{inst}$) resulting were 0.061, 0.095, and 0.13 nm, for exit slit apertures of 30, 60 and 90 μm, respectively. These values were calculated from the measurement of the full width at half maximum (FWHM) of the emission line at 546 nm of a low-pressure discharge Hg lamp. Three C_2^* spectra were acquired in the same experimental conditions for each instrumental resolution. Flame radiation was collected by a fiber optic bundle connected to the spectrometer light entrance. Emission signal was detected by a Hamamatsu R928P phototube, with 950 V as work voltage. Spectra were obtained in the 502 to 518 nm range, which corresponds to the $d^3\Pi_g \rightarrow a^3\Pi_u$ electronic transition, known as Swan band (Gaydon, 1957). All spectra were measured at the region corresponding to the end of the inner flame cone.

RESULTS AND DISCUSSION

Typical experimental C_2^* emission spectra acquired with three instrumental resolutions, 0.061, 0.096 and 0.13 nm are presented in Fig. 1. These resolutions are easily obtained

with commercial, low-cost, and compact spectrometers. These characteristics present special importance regarding the acquisition of data from aeronautics and aerospace applications. In most cases, the payload weight is a critical limiting variable, and the use of compact spectrometers of low-resolution is an interesting choice for *in situ* airborne diagnostic of plasma or combustion (Gord and Fiechtner, 2001; Knight et al., 2000).
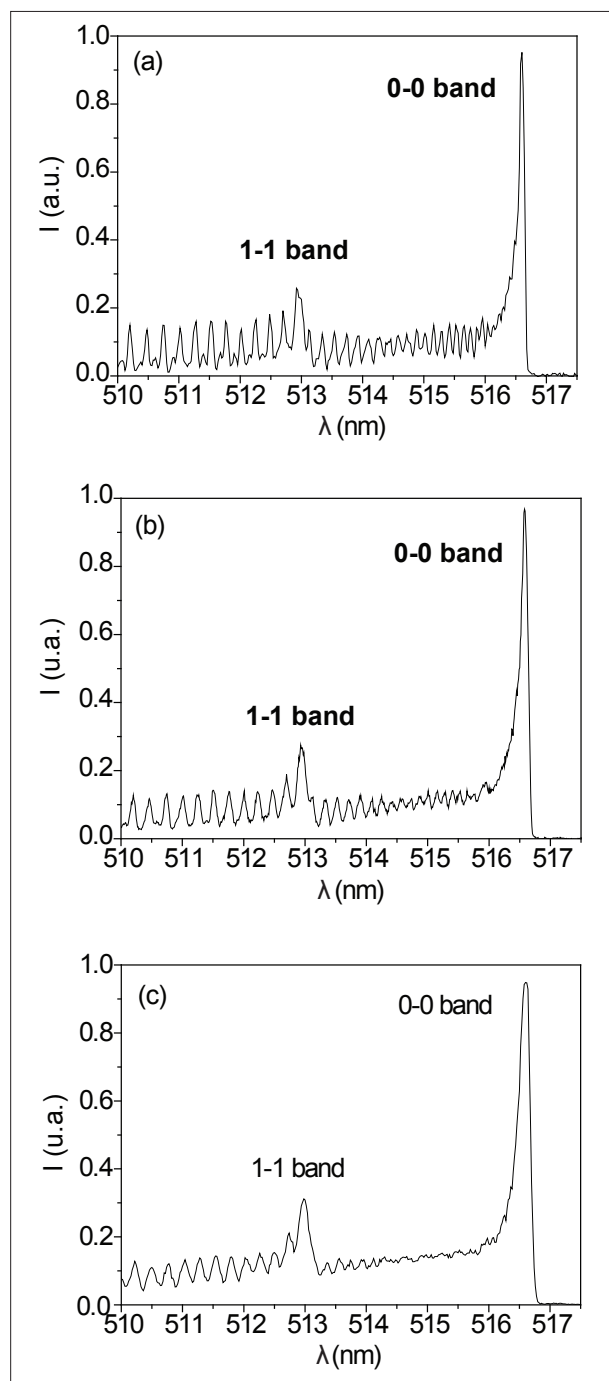

Figure 1: Emission spectrum of the Swan band of the C_2^* radical. Measured instrumental resolution were: (a) 0.061 nm, (b) 0.095 nm and (c) 0.13 nm.

The spectra were normalized with respect to the band-head of the 0-0 vibrational band. The use of different instrumental resolutions provides spectra with remarkable differences. Characteristic intense peaks are observed at ca. 516.6 and 513.0 nm, corresponding to the band-heads of 0-0 and 1-1 vibrational bands, respectively. In the present paper, only the peaks of 0-0 band between 513.5 -516 nm were used in the determination of temperature. This range contains an appreciable quantity of "thermometric peaks", i.e., peaks whose relative intensities are more sensitive to changes in the rotational temperature (Carinhana, 2006). However, the degradation of spectrum profiles caused by peak broadening is stronger in this region.

In the following subsections, we illustrate the use of the simulation tool to aid the temperature determination for the case of higher resolution spectra (0.061 nm).

A - Procedure for the rotational temperature determination

The first issue for temperature determination is the choice of initial values of the rotational temperature (T_{rot}) and of the spectral resolution ($\Delta\lambda_{spec}$), which will be used as input data for computing the theoretical spectra. The initial input data were set as T_{rot} = 1850 K and $\Delta\lambda_{inst}$ = 0.061 nm. The chosen temperature corresponds to the temperature for Bunsen-type flames at the investigated region, determined by McPherson and Henderson (1927). The initial value of spectral resolution was assumed to be equal to the instrumental resolution determined with the Hg lamp.

Figure 2 shows the experimental spectrum overlapped with the synthesized one. It can be noticed differences in the wavelength of corresponding peaks in the experimental and synthesized spectra. These differences can be explained by small deviations in spectral calibration of the experimental apparatus.

One of the tools implemented in the simulator allows a wavelength adjustment in order to match both experimental and synthesized spectra in a region between two defined peaks. This process is done to correct these deviations, improving the procedure taken in the next step to determine the temperature. To carry out the matching, the correspondence of two known pair of peaks of experimental and theoretical spectra, defined by the user, is set in a specific graphic window. In this example, a match was accomplished choosing as reference peaks the band-head of the 0-0 and 1-1 bands, that is, the limits of the region of interest regarding the 0-0 band (Fig. 3). Apparently, a reasonable match between both spectra is shown. Nevertheless, some remarkable differences in the peak intensities can be observed.

The next step is the optimized values determination of the simulation parameters. In fact, as previously explained, a brute force approach is used to solve an inverse problem. For that, a range of T_{rot} and of $\Delta\lambda$, and the number of steps for each of them, is defined by the user. The program computes a sequence of spectra for each (T_{rot}, $\Delta\lambda_{spec}$) pair, and also calculates the respective value of χ^2 comparing the synthesized spectra with the experimental one. These results are expressed graphically as a family of curves of χ^2 *versus* T_{rot} for each value of $\Delta\lambda_{spec}$. The optimized parameters of the theoretical spectrum are given by the

Table 1: T_{rot} and Dl_{spec} calculated from experimental spectra. Temperature uncertainties corresponds to the standard deviation of the measurements

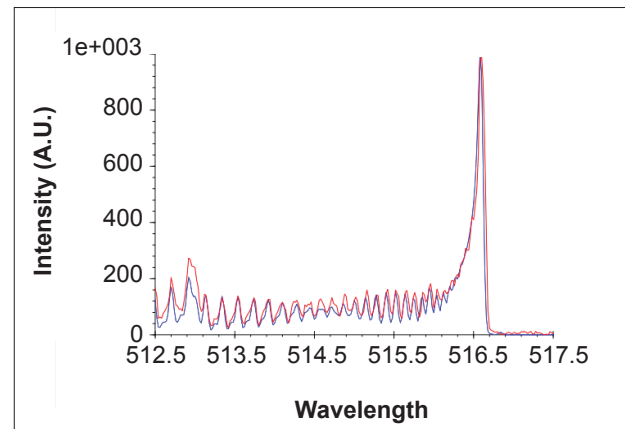| $\Delta\lambda_{inst}$ (nm)	T_{rot} (K)	$\Delta\lambda_{spec}$ (nm)
0.061	2113 (77)	0.069(2)
0.095	2173 (34)	0.0961(16)
0.13	2616 (77)	0.1451(31)

Figure 2: Experimental spectrum with instrumental resolution of 0.061 nm superimposed to a synthesized spectrum computed with rotational temperature of 1850 K and spectral resolution of 0.061 nm.
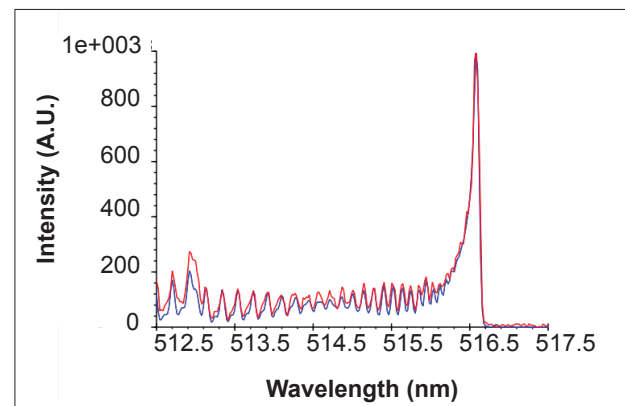

Figure 3: Experimental spectrum with instrumental resolution of 0.061 nm superimposed to a synthesized spectrum computed with rotational temperature of 1850 K and spectral resolution of 0.061 nm after the matching operation.

best spectra match, i.e., by the point where the minimum value of χ^2 is observed. Examples of these calculated curves are shown in Figs. 4 and 5.

With respect to Fig. 4, the testing range of Dl was arbitrarily assigned as 0.061 ± 0.01 nm, i.e., ca. 15% for lower and upper values. A similar criterion was adopted for the T_{rot} range, which was chosen as $1,850 \pm 270$ K. The number of steps for both ranges was 32. Of course, these were not good choices because it is not certain that the family of curves have reached a minimum. The optimized temperature can be out of the chosen range.

Figure 5 shows a new set of curves obtained for $2000 \leq T_{rot} \leq 2,700$ K and $0.061 \leq \Delta\lambda_{spec} \leq 0.081$ nm. From this family of curves, the minimum of c^2 corresponds to the pair $T_{rot} = 2113$ K and $\Delta\lambda_{spec} = 0.069$ nm. Note that the determined rotational temperature was considerably higher than the original input data.

The same procedure was applied to determine the temperature associated with the other experimental spectra. Values of T_{rot} and $\Delta\lambda_{spec}$ presented in Table 1 correspond to the mean values computed from the set of spectra acquired for each instrumental resolution. The errors, presented inside the parenthesis, correspond to the standard deviation.

Figure 6 shows the spectrum synthesized with the parameters presented in Table 1 for $\Delta\lambda_{inst}$ 0.061 nm superposed to one of the experimental spectra. Figure 7 shows the deviation between the same experimental spectrum and the synthesized spectra referring to Figs. 2, 3, and 6. The deviation graph is other of the tools implemented in the computational program, and it is very useful in the superposing spectra evaluation. The optimum synthesized spectrum is in very good agreement with the experimental one. Only small differences are observed in the region between 513.2 and 516 nm, distributed around zero, i.e., in the region of the thermometric peaks.

Here, it is important to say that instrumental resolution ($\Delta\lambda_{inst}$) is not the unique contribution to the peak profile of the emission spectra. There are physical-chemistry phenomena, like Doppler Effect and collisional processes involving the chemical species present in the system, which also cause line broadening of the spectrum (Gaydon, 1957; Griem, 1997). As both factors are mainly dependent on the temperature and pressure of the system, respectively, it can be accepted that they are negligible

Table 2: T_{rot} and $\Delta\lambda_{spec}$ calculated from experimental spectra with nominal resolution of 0.13 nm (lower resolution)

	Spectrum 1	Spectrum 2	Spectrum 3
T_{rot} (K)	2555	2592	2814
$\Delta\lambda_{spec}$ (nm)	0.146	0.141	0.146

for low-pressure Hg lamp, and that $\Delta\lambda_{inst}$ is given by the peak profile of atomic lines (Griem, 1997). However, for atmospheric LPG flames, the Doppler and the collisional
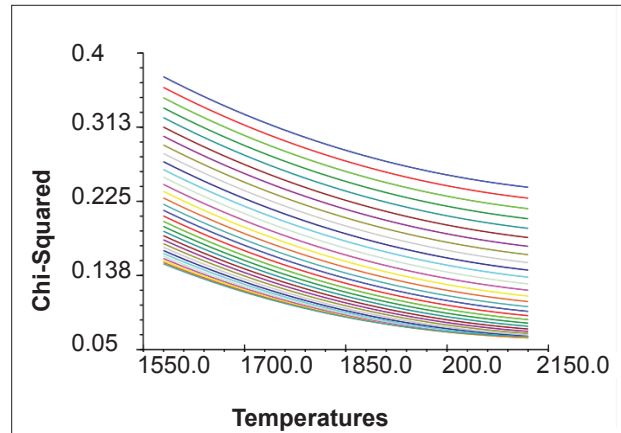

Figure 4: χ^2 versus T_{rot} curves used in optimization procedure of spectral parameters. Each one corresponds to a value of $\Delta\lambda_{spec}$ between 0.05 and 0.07 nm. The minimum value of χ^2 corresponds to the pair $T_{rot} = 2120$ K and $\Delta\lambda_{spec} = 0.07$ nm.

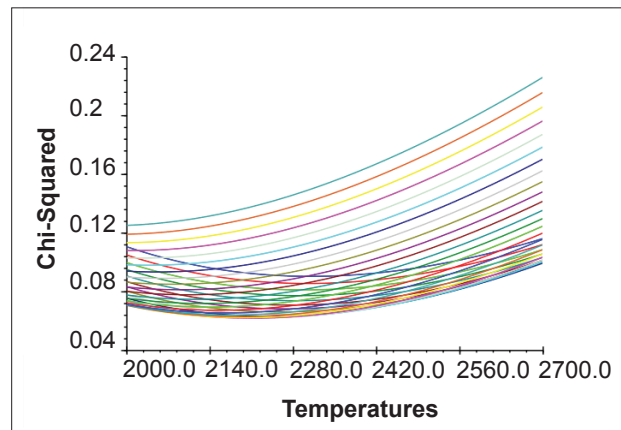Figure 5: Final χ^2 versus T_{rot} curves used in optimization procedure of spectral parameters. The minimum value of χ^2 corresponds to the pair $T_{rot} = 2,113$ K and $\Delta\lambda_{spec} = 0.069$ nm.
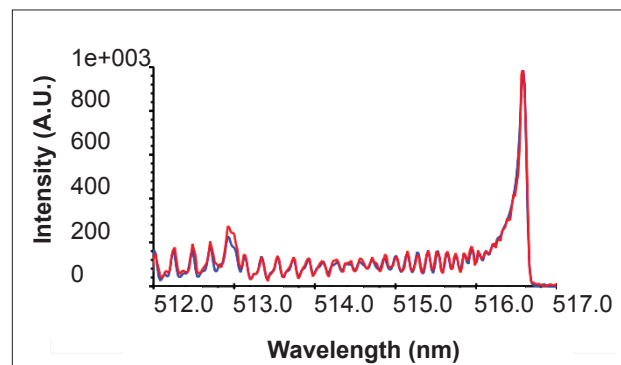

Figure 6: Final synthesized spectrum overlapped with an experimental one. $T_{rot} = 2,113$ K and $\Delta\lambda_{spec} = 0.069$ nm.

broadening show appreciable influence. Thus, the spectral resolution of C_2^* emission spectra is expected to be larger than the $\Delta\lambda_{inst}$.

In all cases, the results regarding $\Delta\lambda_{spec}$ were consistent with the expected behavior: all of them were higher than the $\Delta\lambda_{inst}$. The temperature obtained from the spectra with nominal resolution of 0.061 and 0.095 nm agrees, considering the standard deviation.
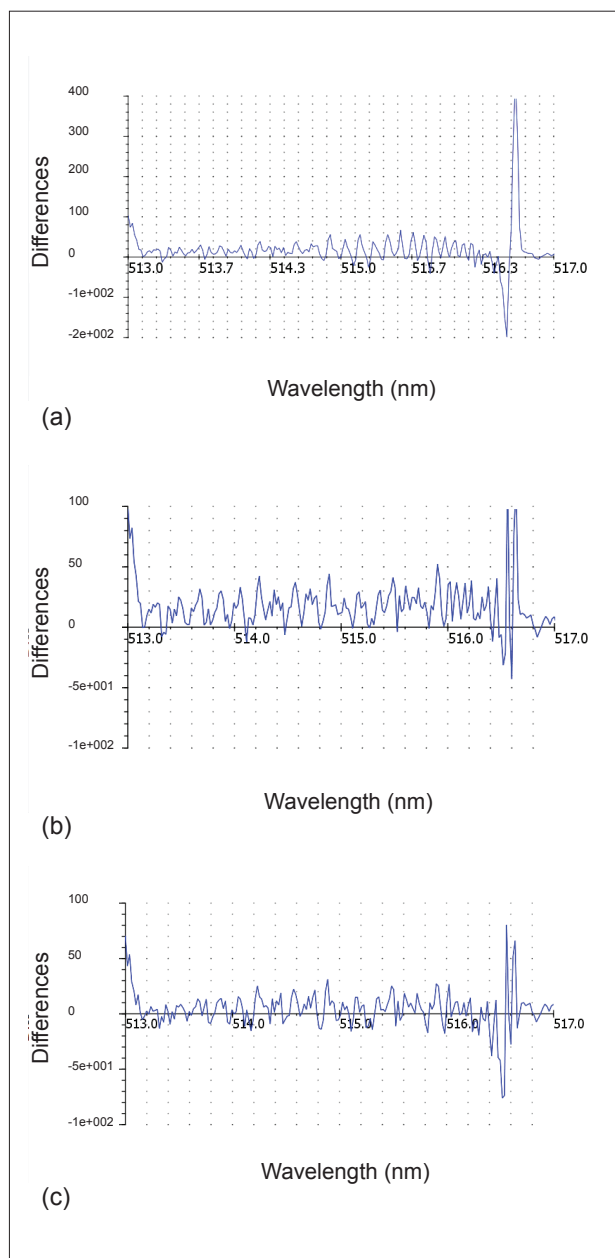

(a)

(b)

(c)

Figure 7: Deviation between synthesized spectrum and the experimental one: (a) original data – 1,850 K; (b) after spectra matching - 1,850 K; and (c) optimized spectrum – 2,113 K. Note that the amplitude of the deviation between the spectra decreases systematically from (a) to (c).

The difference observed in temperature between the computed and the original input data (1850 K) is not related to the computer code response, but to the deactivation processes of the C_2^* radical. In fact, C_2^* radical rotational temperatures have been described in literature as being higher than the theoretical equilibrium flame temperature (Gaydon and Wolfhard, 1970). This means that the rotational degrees of freedom are not in equilibrium and Boltzmann-Maxwell distribution of velocities is not strictly followed. The average number of collisions in the excited state is not sufficient to establish a rotational distribution equivalent to the temperature associated with the translational, or kinetic, mode. The emission spectra, therefore, correspond to a description of the rotationally excited states of the C_2^* species. This feature of C_2^* radical is useful to describe the physical chemistry processes of reactive flows with the presence of organic reactants (Rond et al., 2007). In these systems, the nonequilibrium situation produces a strong radiative emission from the excited species, and this radical can be used as a probe to indicate the temporal evolution of rotational temperature.

B - Limits of the proposed approach

As already stated, the temperature determination by spectra simulation is based on the comparison of experimental and computed spectra profiles. The analyzed range of wavelengths has to contain an appreciable quantity of peaks, whose relative intensities are more sensitive to changes in the rotational temperature. As the resolution decreases (greater values of $\Delta\lambda_{spec}$), a degradation of spectrum profiles caused by peak broadening is expected, reducing the elements for the pair determination $(T_{rot}, \Delta\lambda_{spec})$. In this case, the determination of the temperature can be a problem.

Spectra with 0.13 nm instrumental resolution are examples of cases where the spectra broadening results in a lack of thermometric peaks (Fig. 8b). Table 1 shows that the rotational temperature obtained for the spectra acquired with $\Delta\lambda_{inst}$ = 0.13 nm is much higher than the value presented in other cases. The computer code was able to reproduce consistently the temperature and $\Delta\lambda_{spec}$ even for this set of experimental spectra, resulting in a rotational temperature above 2,600 K with a standard deviation of the same magnitude as the other cases.

A simple procedure was performed to test the former result consistency: the experimental spectra were compared with the simulated ones with T_{rot} of 2,150 K as input data. This value corresponds to the average T_{rot} presented in Table 1 computed from the other two more resolved spectra. The superposed spectra in Figs. 8a and 8b suggest that the experimental one with larger nominal resolution do not present the same T_{rot} of

the others, because the synthesized spectrum is systematically below the experimental in the region of interest, from 513.2 to 517 nm. The temperature determined for each spectrum, in this case, is presented in Table 2, which can indicate that the mixture of oxygen and LPG was not anymore stable when these spectra were acquired.

Additional tests have to be carried out in order to determine the actual limitations of the proposed approach.

CONCLUSIONS

The use of C_2^* molecular emission spectroscopy allied with spectra simulation for reactive flows diagnostic was presented. The presented methodology was able to determine consistently the temperature for C_2^* spectra acquired within a large range of spectral resolution, including low-quality spectrum data. The developed computational program allows a quick determination of the C_2^* rotational temperature associated with the 0-0 band. A brute force approach to solve the inverse problem, allied to a C_2^* spectra simulator, which explores the multicore capability of the modern computers, proved to be a good strategy for determining the optimum pair $(T_{rot}, \square\square_{spec})$. The time frame to obtain the optimum pair is compatible with the spectrum acquisition time, making this one useful tool to flame diagnostics in laboratory.

This has remarkable importance for aeronautics and aerospace applications, where the use of light and compact spectrometers of low-resolution can reduce the payload weight. The computed values of C_2^* rotational temperatures, considering the higher resolution spectra, were ca. 2150 K for a Bunsen burner flame. This value is ca. 6% higher than the one reported in literature, which indicates, as expected, that the Bunsen-type flames are not completely in equilibrium state.

Nowadays, the computer code computes up to five Swan bands, but the determination of the rotational temperature is based only on the thermometric peaks of the 0-0 band. New tools to explore the additional bands are under development.

ACKNOWLEDGMENTS

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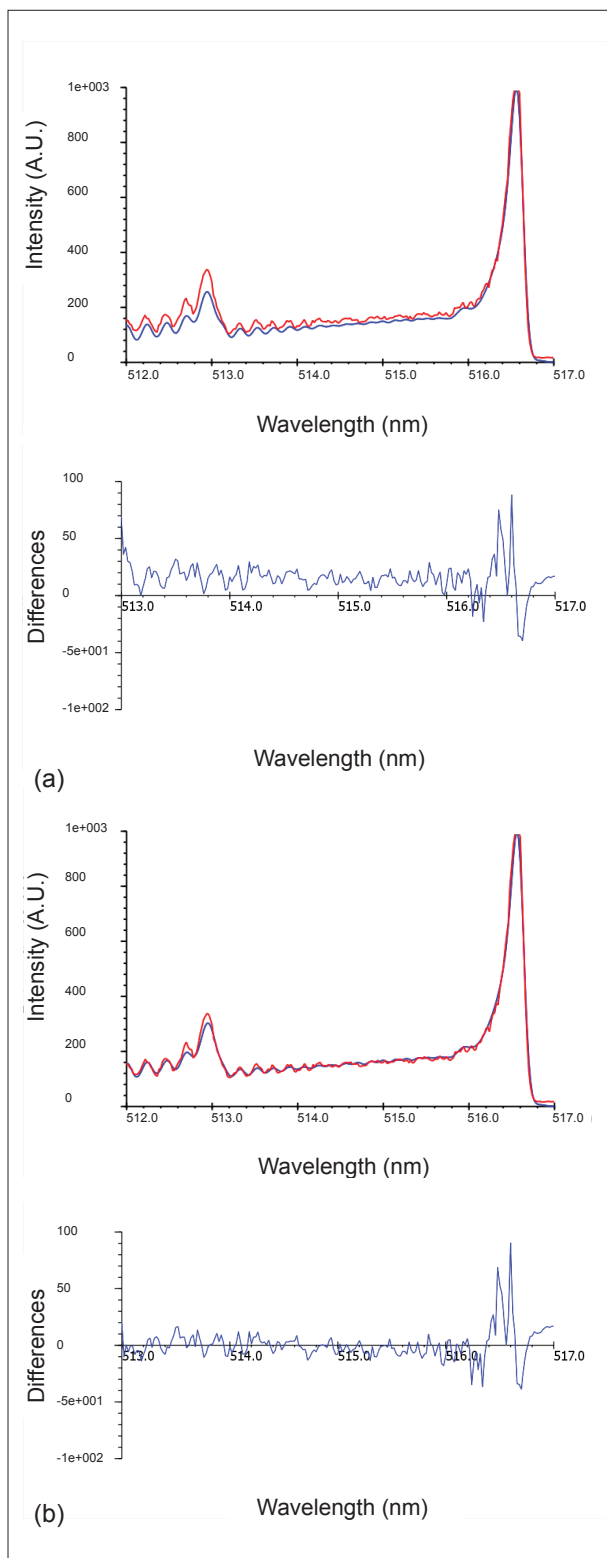

Figure 8: Comparison of synthesized and experimental spectra for two different set of input parameters : (a) T_{rot} = 2,150 K and (b) T_{rot} = 2,616 K. The same $\square\square$ = 0.145 nm is used in both cases.

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