Calculation of the net emission coefficient of an air thermal plasma at very high pressure

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Abstract
The aim of this paper is to present an accurate evaluation of the phenomena appearing for high pressure air plasmas supposed to be in local thermodynamic equilibrium (LTE). In the past, we already calculated the net emission coefficient for air mixtures at atmospheric pressure and for temperatures up to 30kK (molecular contribution being restricted to 10kK). Unfortunately, the existence of high pressures does not allow us to use this database due to the non-ideality of the plasma (Viriel and Debye corrections, energy cut-off…), and due to the significant shifts of molecular reactions towards upper temperatures. Consequently, this paper proposes an improvement of our previous works with a consideration of high pressure corrections in the composition algorithm in order to take into account the pressure effects, and with a new calculation of all the contributions of the plasma radiation (atomic lines and continuum, molecular continuum, and molecular bands) using an updated database. A particular attention is paid to calculate the contribution of all the major molecular band systems to the radiation: O₂ (Schumann-Runge), N₂ (VUV, 1st and 2nd positive), NO (IR, β, γ, δ, ε) and N₂⁺ (1st negative and Meinel). The discrete atomic lines and molecular bands radiation including the overlapping are calculated by a line-by-line method up to 30kK and 100 bar. This updated database is validated in the case of optically thin plasmas and pressure of 1 bar by the comparison of our integrated emission strength with the published results. Finally, this work shows the necessity to extend the molecular radiation database up to 15kK at high pressure (bands and continuum) since their corresponding contributions could not be neglected at high temperature.

1. Introduction
In the earlier works of our team, the calculation of the net emission coefficient of air thermal plasmas was performed at atmospheric pressure for the temperature range 300-15000K [1]. The molecular contribution was limited to temperatures up to 10kK. In fact, several applications of air arcs may involve very high pressures such as special plasma torches (electric arc heaters) used in aerospace for the simulation of atmospheric re-entry behaviour of thermal protective materials [2]. Furthermore, another application in aerospace concerns the high enthalpy wind tunnel which provides flows at high velocity and pressures of several hundreds atmospheres [3]. Therefore, our previous results are not available in these conditions for two main reasons: the plasma is not ideal at pressures up to 100 bar and the plasma compositions did not take into account the pressure effects such as the shift of the dissociation of the diatomic molecules to higher temperatures. The role of the most important molecular species must thus be included for temperatures above 10kK. Consequently, we present here an improvement of the radiation data for air thermal plasmas at high pressures (up to a few hundreds
bars) and rather high temperatures (up to 30000K) paying a particular attention to the role of molecular species in the total plasma radiation. The first step of this work is an upgrade of the plasma composition algorithm used until now in order to consider the high pressure effects on the plasma behaviour. The second part concerns the calculation of the total atomic radiation which is the sum of the atomic continuum (photo-ionization, radiative recombination and atomic bremsstrahlung) and of the atomic lines due to transitions between two discrete states of the atomic species. The third part is devoted to the calculation of the molecular contribution to the total radiation. The molecular continuum is achieved by the compilation of the photoabsorption cross-sections of all the main diatomic molecules constituting air plasmas (O₂, N₂, NO, N₂⁺). A particular attention is also devoted to the calculation of each electronic band system which mainly contributes to the radiation of air plasmas. A systematic line-by-line calculation is achieved for each system, taking in account the overlapping of molecular discrete radiation lines. Finally, the total emission and absorption spectra of air plasmas is obtained by the summation of all the contributions (atomic continuum, atomic lines, molecular continuum and molecular bands) and using a line-by-line method with 4,470,000 wavelengths distributed in the range [30 – 4500 nm], the overlapping being accurately treated. It was then possible to evaluate the influence of each process on the total net emission for different temperatures and pressures. The total emission or absorption spectra can then be used to solve the radiative transfer equation either by an exact calculation or using some approached methods as the NEC (Net Emission Coefficient) [4] or the Mean Absorption Coefficients method (Pₙ models).

2. Plasma composition

In order to characterize the radiative properties of thermal plasmas, the preliminary step consisted in an accurate determination of the mixture composition for each couple temperature / pressure. The computation of equilibrium compositions of air plasmas in the temperature range 300K – 3000K and pressure between 1 and 100 bar was performed according to the method described by Godin and Trépanier [5]. This calculation was based on the mass action law and the chemical base concept. At very high pressure, deviations from the ideal gas model had to be implemented, especially for the atomic species. Based on the works of Drellishak [6] and D’Angola [7] we added the Fermi criterion (high pressure density effect limiting the maximum atomic radius) and the Griem criterion (interactions between charged particles) in order to determine the total energy cut-off limiting the number of atomic levels in the calculation of the internal partition functions. More details are available in [8].

In this work, 31 gaseous species were considered in the calculation of the mixture composition: e, O, O⁺, O²⁺, O³⁺, O₂⁺, O₂, O², O₂⁻, O₂⁻, N, N⁺, N³⁺, N⁴⁺, N⁵⁺, N⁶⁺, N⁷⁺, NO, NO⁺, NO², NO₂, N₂O, N₂O⁺, N₂O⁺, N₂O⁺, N₂O₃, N₂O₄, N₂O₅, Ar, Ar⁺, Ar²⁺, Ar³⁺, Ar⁴⁺. Figure 1 shows the composition of air plasmas at 100 bar.

![Figure 1. Composition of air plasmas (P=100 bar).](image1)

![Figure 2. Total number densities of molecular and atomic species in air (P=1 and 100 bar).](image2)
It is well known that the increase of the pressure leads to shift the temperature dissociation of the molecules (around 2000 K in the case of $N_2$), and consequently gives a more important role to these species in a larger temperature range as illustrated in Figure 2. As consequence of that, we have to extend and improve our previous molecular database following the new pressure conditions.

3. Total absorption coefficient

The determination of the NECs for air plasmas and pressures from 1 bar to 100 bar required an accurate description of the total emission and absorption spectra which were obtained by adding all the radiative phenomena occurring in the mixture: the atomic continuum, the molecular continuum, the atomic lines and the molecular bands. These last radiative mechanisms are briefly described in the following sections.

3.1. Atomic continuum

The radiation of the atomic continuum results from the combination of three main phenomena: the radiative attachment, the radiative recombination and the Bremsstrahlung. The atomic continuum of air plasmas was thoroughly studied in previous papers [8,9].

3.2. Molecular continuum

This continuum contribution plays an important role at low and intermediate temperatures, where the molecular densities are predominant. We thus selected the major molecular species in air plasmas ($O_2$, $N_2$ NO, $N_2O$, …) and we elaborated an exhaustive review of the most accurate photoabsorption cross-sections published in the literature. Over a hundred partial photoabsorption cross-sections were found in the literature. The most exhaustive data were those of Fennely and Torr [10], Lee et al. [11] and the critical review of Hudson [12]. For the calculation, we assumed that the photoabsorption cross-sections did not depend on temperature. Consequently, the molecular continuum was approximated by the multiplication of each cross-section with molecular number density of the corresponding species. Figure 3 shows the molecular continuum of the main molecular species in air plasmas ($O_2$, $N_2$ and NO) for atmospheric pressure and temperature of 300K.

3.3. Atomic lines

The calculation of atomic lines emissions required a complete data bank including the properties of 15530 lines (6217 lines for oxygen species and 9313 lines for nitrogen species) whose characteristics (energy levels, quantum numbers, transition probabilities, oscillator strengths, …) were issued from [13-15]. In a previous work [1], we assumed that the lines’ overlapping had a negligible influence on the radiation allowing us to treat each line by means of an escape factor. In this work, we decided to use the exact description of the atomic line spectrum and to compare the results with those obtained using the escape factor. We then calculated the atomic line radiation for isothermal plasmas of

![Figure 3. Photoabsorption cross-sections for O$_2$, N$_2$ and NO.](image)
thickness $R_p$ as follows:

$$
\varepsilon_{\lambda}^{\text{line}}(T, R_p) = \left( \frac{e^2}{4\pi^2}\right) \left( \frac{\pi}{m_e c} \right) \cdot L^\nu_\lambda(T) \cdot f_{\lambda u} \cdot \bar{n}_f(T) \cdot \exp(-K'_\lambda R_p) \cdot \left( 1 - \exp\left( -\frac{hc}{\lambda_0 k_b T} \right) \right)
$$

(1)

where $L^\nu_\lambda(T)$ is the Planck function at the centre of the line, $e$ is the electron charge, $\lambda$ is the wavelength corresponding to the transition between the upper energy level $E_u$ and the lower energy level $E_l$, $m_e$ is the mass of the electron, $\bar{n}_f$ is the number density of the lower energy level, $f_{\lambda u}$ is the oscillator strength of this transition and $K'_\lambda$ is the total absorption coefficient in m$^{-1}$. As the spectral description of the atomic lines’ radiation required the knowledge of the line profiles, each line was described by a Voigt profile:

$$
P^{\text{Voigt}}_\lambda = \frac{\gamma_D}{\gamma_D^2 \pi^2} \int_0^\infty \exp\left( -y^2 \right) \left( \frac{\gamma_L \sqrt{\ln 2}}{\gamma_D} \right)^2 \left( \frac{(\lambda - \lambda_0) \sqrt{\ln 2}}{\gamma_D} - y \right)^2 dy
$$

(2)

with $\gamma_D$ and $\gamma_L$ the Doppler and Lorentz (natural, resonance, Van Der Waal and Stark) broadenings respectively. The HWHM (Half Widths at Half Maximum) were given by Laux [16] for the Doppler effect, from Ali and Griem [17] for the resonance broadening, from Walkup [18] for the Van der Waals broadening and finally from Griem [19] for the Stark broadening including the corrections of Griem [19,20,21] to take into account the ionic influence. Due to the overlapping and a line by line treatment, we used two algorithms to spare time: a quicksort-insertion algorithm to sort the data before summation, and the Drayson algorithm [22] to integrate the relation (2).

**Figure 4.** Influence of the atomic line’s overlapping for different $R_p$ and 1 bar.

**Figure 5.** Influence of the atomic line’s overlapping for optically thin air plasmas at 100 bar.

The last step to describe the total emission spectrum was a direct summation in the wavelengths range (30-4500 nm) with a 10$^{-3}$ nm spectral step. The latter was validated by a comparison between the integration of the emission spectrum and the sum of the integrated atomic lines. As shown in Figure 4, the NECs obtained with and without overlapping are similar in the case of optically thin plasmas. We can also remark that the escape factor does not lead to an important error at low temperatures and low plasma’s thicknesses $R_p$. At contrary, the use of this factor can lead to an overestimation of factor 2 for higher thicknesses at 1 bar. Moreover, the Figure 5 highlights that discrepancies could overtake a factor 10 with the rise of pressure and/or thickness. This difference is mainly due to the total HWHM
of the atomic lines which increases with temperature and/or pressure, causing a more important overlapping phenomenon.

### Table 1. Molecular electronic systems considered in this study

| Electronic States | Calculated bands | Number of rotational lines |
|-------------------|------------------|---------------------------|
| O₂ Schumann-Runge | (19,21)           | ≈ 200.000                 |
| NO Beta           | (15,22)           | ≈ 550.000                 |
| Gamma             | (8,20)            | ≈ 300.000                 |
| Delta             | (4,21)            | ≈ 160.000                 |
| Epsilon           | (5,22)            | ≈ 90.000                  |
| Infrared          | (26,25)           | ≈ 120.000                 |
| N₂⁺ 1ˢᵗ negative | (12,20)           | ≈ 200.000                 |
| Meinel            | (9,10)            | ≈ 110.000                 |

| Electronic States | Calculated bands | Number of rotational lines |
|-------------------|-------------------|---------------------------|
| N₂ 1ˢᵗ positive   | (21,21)           | ≈ 300.000                 |
| 2ⁿᵈ positive      | (4,19)            | ≈ 50.000                  |
| Birge-Hopfield 1  | (19,0)            | ≈ 1.000                   |
| Birge-Hopfield 2  | (28,0)            | ≈ 1.000                   |
| Caroll-Yoshino    | (8,0)             | ≈ 400                     |
| Worley-Jenkins    | (4,0)             | ≈ 200                     |
| Worley            | (4,0)             | ≈ 200                     |

3.4. Molecular bands

Finally, a particular attention was devoted to the radiation of the molecular bands. In one of our previous works [1], 9 molecular electronic systems (1788 vibration bands, 10⁶ rotational lines) were considered in the visible and UV parts of the spectrum, taking into account 9 bound-bound molecular systems (Schumann-Runge system for O₂; first and second positive systems for N₂; β, γ, ε and δ systems for NO; first negative and Meinel systems for N₂⁺). This work highlighted the fundamental role of molecules in the total emission of air plasmas. However, it was restricted to temperatures below 10kK, to optically thin plasmas, to Doppler broadening and to atmospheric pressure. Therefore, it was indispensable to extend this work so as to consider the radiation of molecular bands for higher pressures, higher temperatures and assuming or not the self-absorption. In this work we updated our spectroscopic database by adding the contribution of N₂ VUV (Birge-Hopfield 1 and 2, Caroll-Yoshino, Worley-Jenkins and Worley systems) and the NO Infrared system. All molecular systems were treated in the coupling Hund’s case b.

We have to precise here that this case b is valid for Σ states, and tends to be valid when the rotational level increases for Π states. The molecular systems were then described by the quantum numbers Λ, S, N and J [23]. The total discrete molecular bands radiation was calculated from the main diatomic molecular systems referenced in Table 1 [24]. Therefore, we considered 15 bound-bound diatomic molecular systems in this study.

In order to describe the radiation spectrum of molecular bands by a line-by-line method, it was necessary to calculate the corresponding spectral emission coefficient for each wavelength as follows:

\[
e_{\lambda} = \frac{h \epsilon (E_{\text{up}}^{\nu \nu'J} - E_{\text{down}}^{\nu'\nu J})}{4\pi} \Phi \left( \frac{h \epsilon E_{\text{up}}^{\nu \nu'J}}{kT} \right) \frac{N(T) \exp\left( \frac{-h \epsilon E_{\text{up}}^{\nu \nu'J}}{kT} \right)}{Q_{\text{int}}^{\nu}(T)} A_{\lambda}^{\nu \nu'J} S_{\nu'J}^{\nu J} P_{\lambda}^{\nu \nu'J} \right]
\]

where \( E_{\text{up}}^{\nu \nu'J} \) are the total energies expressed in cm⁻¹, \( N(T) \) is the total population number density of the molecule in m⁻³, \( Q_{\text{int}}^{\nu}(T) \) is the total internal partition, \( \Phi \) is the nuclear spin degeneracy factor.
of the emitting state [1], \( A_{v'v''}^{e} \) is the vibrational transition probability in s\(^{-1}\), \( S_{N'J'}^{N'J} \) is the Hönl-London factor corresponding to the rotational transition involved and \( P_{2}^{\text{Voigt}} \) is the Voigt profile in m\(^{2}\). In the formalism of Hund’s case b, the numbers \( e, v, N \) and \( J \) correspond to electronic, vibrational, rotational and sub-rotational levels (spin splitting) respectively.

The line-by-line method used to describe the emission spectrum consists in determining for each rotational transition the emitting population number density, the line position, its integrated intensity and the line shape.

**The population number density** of the rotational states was assumed to be a Boltzmann distribution since the plasma was supposed to be in LTE (Local Thermodynamic Equilibrium).

**The line position** is related (in cm\(^{-1}\)) to the energy difference between two rotational levels and was calculated in the Klein-Dunham formalism (equilibrium spectroscopic constants for each electronic state). In the Born-Oppenheimer approximation, the expression of the energy levels is given by the expression:

\[
E_{\epsilon,v,N,J} = \sum_{i,j} Y_{ij}(\nu + 1/2)\left[F(N,J)\right]^{i,j}
\]

(4)

The rotational term dependency \( F(N,J) = N(N+1) \) only depends on \( N \) in the case of a singlet transition (\( S=0 \)). For multiplet states (spin-splitting) it not only depends on the parameter \( N \) but also on the sublevel \( J \). Expressions for each electronic state were given in the literature [1,16,24]. Lambda-doubling was neglected for the \( \Pi \) states. For each electronic state (Table 1), we chose the most accurate and most recent spectroscopic constants given by Naghizadeh-Kashani [1], Chauveau [24] and Laux [16].

**The integrated line intensity** is proportional to the product of the vibrational transition probability (Einstein coefficient) and the Hönl-London factor which defines the relative rotational intensity between transitions allowed by the selection rules. The Hönl-London factors used in this work were those of Tatum [25], Schadee [26] and Hertzberg [23]. All factors were normalized according to the Whiting convention:

\[
\sum_{J'} S_{N'J'}^{N'J} = (2-\delta_{0,N+1,0})(2S'+1)(2N'+1)
\]

(5)

We used the Einstein coefficients published by Laux [16] for most of the molecular systems. For the \( \text{N}_2 \) VUV systems (Birge-Hopfield 1 and 2, Worley-Jenkins, Worley and Caroll-Yoshino), we used the absorption oscillator strengths compiled by Chauveau [24]. The table published by Langhoff [27] was valuable for the infrared system of NO. Finally, we had to develop the classical method for the calculation of the vibrational transition probabilities for the electronic systems of the \( \text{O}_2 \) Schumann-Runge and NO-gamma systems since we obtained significant differences with the results published for these cases. Accordingly, the first step was the reconstruction by the RKR method [28] of the internuclear potential curves for each state. We extrapolated the RKR potential with a repulsive potential \( V_{\text{rep}}(r)=a.r^{-b} \) for the shorter internuclear distances, and with a Hulburt-Hirschfelder potential (HH) for the longer internuclear distances. In order to calculate the vibrational wave functions \( \psi_{v'}(r) \), this potential was injected in the radial Schrödinger equation and the differential equation was then resolved by a method proposed by Cooley [29] using a Numerov iteration algorithm (two-step). This calculation was achieved for all couple \((v',v'')\). Knowing the upper and lower rotationless vibrational wave functions, the vibrational transition probabilities were obtained as follows:

\[
A_{v'v''}^{e} = 2.026 \times 10^{-6} \left( \sigma_{v'} \right)^{2} \left\{ \frac{2-\delta_{0,N+1,0}}{2-\delta_{0,N'}} \right\} \left[ \frac{1}{0} \psi_{v'}(r)R_{e}(r)\psi_{v'}(r)dr \right]^{2}
\]

(6)

The electronic transition dipole moments \( R_{e}(r) \) used in the calculation were published by Friedman for the \( \text{O}_2 \) Schumann-Runge [30] and Langhoff for NO-gamma systems [31]. Our Einstein coefficients
were compared with published values [16], and the NECs obtained with our tables of coefficients were in good agreement with the literature for those two molecular systems [24].

Finally, the line shape of each line was calculated by a Voigt profile taking into account the Doppler and the collisional broadenings. The Doppler broadening for molecules obeys to the same formula as atomic case [16]. For collisional broadenings, we did not find enough information to cover our large pressure and temperature ranges. Consequently, for all diatomic molecular bands, we systematically used the semi-empirical approximation law to determine the molecular collisional broadenings HWHM (Half widths at Half Maximum) as a function of pressure:

$$\gamma_c(T, P) = \gamma(T_{ref}, P, T_{ref}, P)^n$$

with $\gamma_{Tref} = 0.1$ and $n=0.7$ for all molecules as recommended by Breene [32].

![Figure 6. Comparison with the literature for the particular system N$_2^+$ 1$^\text{st}$ Negative system](image)

![Figure 7. Contribution of the molecular diatomic species to the NEC.](image)

The NECs of all molecular bands systems were validated by comparing them to previous works for a pressure of 1 bar (Figure 6). At most 80% of discrepancy was observed with Naghizadeh-Kashani [1], 30% with Chauveau [24] and 70% with Laux [16]. Our results almost systematically overestimated those of Naghizadeh and Laux and underestimated those of Chauveau. These discrepancies are mainly caused by the number of vibrational levels considered in the Tables of the Einstein coefficients. Indeed, this number differs according to the used methods for the limitation of the rotational levels for each vibrational level, and/or according to the spectroscopic constants chosen to describe the energy levels. In this study, we defined this maximum number by comparing the vibrational energy with the dissociation limit. The limitation of the rotational levels for each vibrational state was performed by comparison of the total rotational states energies $E_{r,v,N,J}$ with the local maximum of the rotational-vibrational potential curve [33]. Figure 7 highlights that N$_2^+$ 1$^\text{st}$ negative system is highly responsible for the molecular radiation at 10kK whereas O$_2$, NO and N$_2$ systems are above 7kK.

### 4. Results obtained with the Net Emission Coefficient (NEC)

The plasma’s radiation was obtained according to the Net Emission Coefficient (NEC) which consists in calculating the net emission at the center of an isothermal and homogenous sphere of radius $R_p$:

$$\varepsilon_N(T, R_p) = \int_0^\infty L_\lambda(T) \cdot K_\lambda(T) \cdot \exp(-K_\lambda(T) \cdot R_p) \cdot d\lambda$$

(8)
where $\lambda$ (m) is the wavelength, $T$ and $P$ are the local temperature and pressure respectively, $L^\lambda(T)$ (W/m$^2$/sr/m) is the Planck function, $R_p$ is the size of the sphere assimilated to the plasma’s size and $K^\lambda(T)$ (m$^{-1}$) is the monochromatic absorption coefficient corrected by the induced emission and correlated with the local emission coefficient by the Kirchhoff law. Furthermore, due to the exponential term, radiation’s self-absorption in the emitting regions is rather well estimated.

In numerical modelling, the NEC is a good approximation to characterize the radiation losses in hot regions while taking into consideration the absorption. Our previous studies also showed that this coefficient not only depended on temperature and pressure but also on the plasma size.

**Figure 8.** NEC for optically thin air plasmas at 1 bar.

**Figure 9.** NEC for optically thin air plasmas at 100 bar.

Figures 8 and 9 represent the NEC and the contribution of each radiative mechanism for optically thin air plasmas at 1 bar and 100 bar. We observe that the pressure increase does not significantly modify the relative contribution of each process. We also remark that molecular bands are predominant at low temperatures, up to 5kK for 1 bar and up to 7.5kK for 100 bar. In this case, atomic lines are not self-absorbed and consequently contribute to the radiation at high temperature.

**Figure 10** shows the contribution of each process to the NEC for various plasma sizes at 100 bar. We note that the self-absorption of the atomic lines hardly impacts on the NEC due to the absorption coefficient in the exponential term (8) which increases with pressure. The contribution of the atomic lines is hardly predominant for optically thin plasmas at high temperatures and low pressures. When the pressure increases, the absorption coefficient and the broadening parameters of each line increases too. The self-absorption becomes so important that the molecular continuum and molecular bands significantly extend their ascendency domains when the thickness of plasma increases.

**Figure 11** illustrates this phenomenon showing the contribution of all molecular bands to the NEC. We observe a shift of the molecular contribution towards higher temperatures depending on the pressure and the plasma’s thickness. The molecular bound-bound contribution could be neglected above 10kK only for optically thin plasmas. For those reasons, a particular attention has to be paid to realise an accurate calculation of each diatomic electronic system involved in the radiation of air plasmas.
5. Conclusions

The aim of this paper was to calculate the Net Emission Coefficient of air plasmas for pressures up to 100 bar. First, we upgraded our composition algorithm to better estimate the effects of high pressures. Then, we compared our NECs obtained by a line-by-line calculation or using the escape factor. We showed that the differences between these two approaches rise with the pressure due to the overlapping of the atomic lines. Finally, we showed that the molecular contribution, often neglected in this kind of works, has to be accurately calculated until approximately 15kK. A particular attention has to be paid to model these bound-bound molecular bands in the case of air plasmas.

6. References

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Figure 10. Contribution of the radiative processes to the NEC for two \( R_p \) and \( P=100 \) bar. (AC : Atomic continuum, MC : Molecular continuum, MB : Molecular Bands, AL : Atomic lines)

Figure 11. Total contribution of the molecular bands to the NEC for different pressures and plasma thicknesses (\( R_p \)).
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