Net emission coefficient of CO$_2$-Cu thermal plasmas: role of copper and molecules.

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Abstract
The present work was conducted to calculate the radiative transfer in CO$_2$–Cu mixtures at atmospheric pressure. The plasma is supposed to be in local thermodynamic equilibrium (LTE) for temperatures between 500 and 30000 K. In the first part of the work, we briefly present the plasma composition. The second part is devoted to the various mechanisms responsible for the radiation (atomic continuum, molecular continuum, atomic lines and molecular bands). A particular attention is paid to the radiation of molecular bands. These radiative properties are then introduced in the net emission coefficient (NEC) which is used to estimate the radiation in an isothermal and homogeneous medium. Finally, some results are proposed for various concentrations of copper and different plasma sizes. For optically thin plasmas, the radiation of molecular bands and metallic vapors strongly increase the NEC at low temperature. For higher plasma sizes, the copper lines do not only have high intensities but are also strongly self-absorbed contrarily to the radiation of the molecular bands which becomes the major part, indicating that we must not neglect their contribution in the total radiation.

1. Introduction
In a previous work [1], the plasma parameters of an electric arc discharge in a gas flow between copper electrodes were experimentally investigated. The electrical and thermal conductivities of Cu-Air and Cu-CO$_2$ plasma were calculated. Experimental temperature values were low (3000 K<T<6000 K) for a discharge at current 3.5A. To complete this experimental study and develop further numerical models, this paper deals with the radiation of CO$_2$–Cu plasmas. Some values can be found in the literature concerning pure CO$_2$ [2] and pure copper or mixtures with copper as Ar-Cu [3-6], N$_2$-Cu [4], SF$_6$-Cu [3,4,7], Air-Cu [8,9] and Ar-H$_2$-Cu [6]. Few data are available for this mixture. We then estimated the continuum radiation (molecular continuum, bremsstrahlung, radiative attachment) and the atomic line radiation spectra neglecting the atomic lines overlapping. According to the measured temperatures, a particular attention was focused on the emission of the molecular bands of CO, C$_2$, O$_2$ and CO$_2$ (with overlapping of molecular lines) since no data were available in the literature for the CuO species. Finally, we estimated the net emission at the centre of an isothermal and homogenous sphere, by applying the method of the Net Emission Coefficient [10].

2. Composition of the plasma
For thermal plasmas, the radiation strongly depends on the species present in the medium. In the present work, the computation of the equilibrium compositions of CO$_2$ plasmas with copper metallic vapors in the temperature range 500K – 30kK and at atmospheric pressure was realized with a method based upon the mass action law and the chemical base concept described by Godin and Trépanier [11]. We have considered 34 gaseous species: e’, C, O, Cu, C’, O’, Cu’, C$_2’$, O$_2’$, C$_3’$, CO$_2’$,
C⁺, O⁺, Cu⁺, C₂⁺, O₂⁺, CO⁺, C²⁺, O²⁺, Cu²⁺, C³⁺, O³⁺, Cu³⁺, C₂, O₂, Cu₂, CO, C₃, O₃, CO₂, C₂O, CuO, C₄, C₃O₂. Internal partition functions (IPF) of atoms and their positive ions were calculated with electronic level energies and degeneracies taken from the NIST database [12]; these IPF were systematically confronted with the Drawin and Felenbok’s IPF compilation [13]. IPF were assumed to be equal to the degeneracy of the ground state for negative atomic ions and they were calculated using the Morse potential minimization method for diatomic species. The spectroscopic data (Dunham coefficients, vibrational frequencies and degeneracies, rotational constants, moments of inertia and symmetry numbers) essential to the calculation of the internal partition functions of molecules were taken from Chase et al [14] and Huber and Herzberg [15]. First order corrections corresponding to deviations to the ideal gas law were included in the computation: the Virial correction [16] associated to interactions between neutral chemical species at low temperature and the Debye-Hückel correction [17] due to Coulombian interactions between charged particles at high temperature. An example with 99%CO₂-1%Cu (in mass proportions) is given in Figure 1.

![Figure 1](image_url)

**Figure 1.** Composition for a 99%CO₂-1%Cu plasma at atmospheric pressure.

### 3. Total absorption coefficient

The computation of the NEC requires the total absorption coefficient which was obtained after a preliminary study of the radiation resulting from the atomic continuum, the molecular continuum (6 molecules C₂, CO, CO₂, O₂ and O₃), the atomic lines and the molecular bands (Schumann-Runge system of O₂, 4th positive and IR systems of CO, Swan system of C₂ and IR bands of CO₂).

#### 3.1. Atomic continuum

For the atomic continuum radiation, we have to take into account three important phenomena: the radiative attachment, the radiative recombination and the Bremsstrahlung. Even if the radiative attachment is often negligible on the total continuum absorption coefficient, we nevertheless considered it because of the O⁺, C⁺ and Cu⁺ species in the plasma. The monochromatic emission coefficient is given by Gleizes et al [4] and requires the negative ion population number density and the photo-detachment cross-sections which were issued from Robinson and Geltman for oxygen [18], Yachkov for carbon [19] and Schultz-Gulde E and Gilde for copper [20].

The radiative recombination is often essential in the continuum emission of thermal plasmas and was calculated according to the method of the Scaled Thomas-Fermi potential [21]. By taking into consideration LTE assumption, the monochromatic emission coefficient is given for a temperature T and a wavelength λ by Gleizes et al [4] and requires the electron and ion number densities, the ion charge, the partition function and the ion ground state statistical weight and the Biberman-Schlüter factor taking into account the difference between the photoionization cross section of the energy levels...
and the corresponding hydrogen-like cross section [22,23]. This factor was calculated by Hofsaess for oxygen and carbon species [24]. As the Biberman-Schlüter factor is not available in the literature for the copper species, the photoionization cross sections is obtained according to the hydrogenoid-like atoms approximation developed by Okuda et al [25]. For this last method, the energy levels and principal quantum numbers of the considered species are issued from the tables of Moore [26], corrected by the recent data of the NIST [27] and completed if necessary by those of Kurucz and Peytremann [28].

The Bremsstrahlung coefficient due to the interaction between electrons and atoms is given by Gleizes et al [4] knowing the Gaunt factor introduced to correct the variation from the hydrogen-like approximation, and calculated from the total elastic cross section. We used the total cross sections issued from Neynaber et al and Robinson and Gelman [18,29] for oxygen and carbon, and from [18,30-32] for copper. The Bremsstrahlung coefficient due to the interaction between electrons and ions is obtained according to Gleizes et al [4] with the use of the Gaunt factor.

3.2. Molecular continuum
A special attention has been devoted to the description of the molecular continuum spectrum too since it represents one of the major contributions to the total emission at low temperature. We considered the C₂, CO, CO₂, O₂ and O₃ molecules assuming that the photoabsorption cross-section did not depend on temperature. Consequently, each molecular continuum could be obtained by the multiplication of this section by the corresponding molecular density. More than 50 references were used to reconstruct the photoabsorption cross-sections. Figure 2 shows the molecular continuum obtained at 300K, for 5 molecules and the atmospheric pressure, the number density for O₃ being negligible.

![Figure 2. Photoabsorption Cross-sections for C₂, O₂, CO and CO₂.](image)

3.3. Atomic lines
To calculate the emission of the atomic lines, we elaborated a complete data bank including the properties of 18069 lines (6672 lines for carbon species, 6217 lines for oxygen species and 5180 lines for copper species) whose characteristics (energy levels, quantum numbers, transition probabilities, oscillator strengths…) were issued from [26-28]. The radiation transfer requiring the knowledge of the line profiles, we took into account the Doppler broadening, the pressure effects (resonance and Van der Waals broadenings) and the Stark broadening [33]. The FWHM was obtained from the relation given by Laux [34] for the Doppler effect, from Ali and Griem [35] for the resonance broadening, from Walkup et al [36] for the Van der Waals broadening and from Griem [37] for the Stark broadening including the corrections of Griem [33,37,38] to take into account the influence of the ion. Due to the numerous lines, 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 $\Lambda_{\text{line}}$ introduced by Drawin and Emard [39]. This factor allowed us to simplify the treatment of the spectra and to spare calculation time. In the case of an isothermal plasma, this escape factor $\Lambda$ depends on the line profile and thus, of all the mechanisms of line broadening. It can be defined by the ratio between the radiative flux escaping an isothermal plasma of thickness $R_p$ with the consideration of the absorption and the radiative flux without absorption (optically thin plasma). Therefore, the resonance line escape factor lies between 0 and 1, and its value is equal to 1 for the other lines. The consequences are that the use of the escape factor tends to over-estimate the radiation whose values are however acceptable assuming the precision of the method. With the definition of the escape factor, the line radiation is written as:

$$
\varepsilon^\text{line} (T, R_p) = \left( \frac{e^2}{4\pi e_o} \right) \cdot \left( \frac{\pi}{m_e c} \right) \cdot L^\nu_u (T) \cdot f^\nu_u \cdot \bar{n}^\nu (T) \cdot \Lambda_{\text{line}} \cdot \left( 1 - \exp \frac{-hc}{\lambda k_b T} \right)
$$

where $L^\nu_u (T)$ 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, $\bar{n}$ is the average number density of the lower energy level, $f^\nu_u$ is the oscillator strength of this transition and $\Lambda_{\text{line}}$ is the escape factor of the line.

### 3.4. Molecular bands

Finally, the most important work of this paper is the consideration of the radiation issued from the molecular bands. In a previous work focus on the radiation of air plasmas, 9 molecular electronic systems corresponding to 1788 vibration bands and more than $10^6$ rotation lines [40] were considered in the visible and UV parts of the spectrum (Schumann-Runge system for $O_2$, first and second positive systems for $N_2$, $\beta$, $\gamma$, $\varepsilon$ and $\delta$ systems for NO, first negative and Meinel systems for $N_2^+$). This work had enabled to highlight the fundamental role of molecules in the emission of an air plasma for temperatures under 8000 K. Unfortunately, in the case of CO$_2$ plasmas, our team only has data for O$_2$ which are limited to temperatures inferior to 10000K, for an optically thin plasma and the atmospheric pressure. Therefore, it was necessary to develop new studies in order to consider the radiation of molecular bands of O$_2$, CO, CO$_2$ and C$_2$ species whatever the pressure, the temperature and with or without absorption. Molecular emission and absorption spectrums were described using 4.470.000 points covering the spectral range [30-4500] nm. All molecular systems were treated in the coupling Hund’s case b which is valid for $\Sigma$ states and which tends to be valid for $\Pi$ states when the rotational level increases. In the Hund’s case b, the quantum numbers which permit to describe the system are $\lambda$, $S$, $N$ and $J$. The overlapping of molecular bands was considered by summation in each wavelength point of all molecular systems spectrum before the evaluation of the self-absorption.

#### 3.4.1. Diatomic systems

The main diatomic molecular systems contributing to CO$_2$ mixture radiation [41] (IR and 4th positive systems of CO, Swan system of C$_2$ and Schumann-Runge system of O$_2$) were studied. To describe the radiation spectrums of molecular bands, it was necessary to calculate for each wavelength the spectral emission coefficient by the relation:

$$
\varepsilon^\nu (T) = \frac{hc}{4\pi} \left( E^\nu_{\text{em}} - E^\nu_{\text{abs}} \right) \Phi \left( \frac{N(T) \exp\left( -\frac{hcE^\nu_{\text{em}}}{kT} \right)}{Q^\nu_{\text{int}} (T)} \right) A^\nu_{\text{em}} S^{\nu}_{\lambda} \delta^{\nu \text{Voigt}}
$$

where the energies $E^\nu_{\text{em}}$ and $E^\nu_{\text{abs}}$ are expressed in cm$^{-1}$, $N(T)$ is the total density of the molecule in m$^{-3}$, $Q^\nu_{\text{int}} (T)$ is the internal partition function, $\Phi$ is the nuclear spin degeneracy of the emitting state, $A^\nu_{\text{em}}$ is the vibrational transition probability in s$^{-1}$, $S^{\nu}_{\lambda}$ the Hönl-London factor corresponding to the rotational transition involved and $\delta^{\nu \text{Voigt}}$ the Voigt line profile in m$^{-1}$. In the formalism of humd’s case b, the numbers $\nu$, $N$ and $J$ corresponds respectively to electronic, vibrational, rotational and subrotational levels (spin splitting). Spectrums were computed using a line by line simulation spectrum.
method which consists in determining for each rotational transition involved the emitting density population, the line position, its integrated intensity and the line shape.

The line position was related (in cm⁻¹) to the energy difference between two rotational levels, which were calculated in the Klein-Dunham formalism (equilibrium spectroscopic constants for each electronic state). In the Born-Oppenheimer approximation, the expression of energy levels is given as follows (3):

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

(3)

F(N)=N(N+1) being the rotational term dependency in the case of singlet transition. In the case of multiple transitions, a more complex expression must be found to describe spin multiplicity [42]. Lambda-doubling have been neglected for the \( \Sigma \) states. All spectroscopic constants for each electronic states used for this work are referenced in Table 1.

### Table 1. Molecular systems and spectroscopic constants used for each electronic state

| Mol States | Band system | Transitions | Spectroscopic constants | Bands \((v',v'')\) | Number of rotational lines |
|------------|-------------|-------------|-------------------------|------------------|---------------------------|
| \( \text{O}_2 \) | \( X^3\Sigma^+_g \) \( B^3\Sigma^+_u \) | Schumann-Runge | \( B^3\Sigma^+_u - X^3\Sigma^+_g \) | [34] [43] | (19,21) 200.000 |
| \( \text{CO} \) | \( X^1\Sigma^+ \) \( A^1\Pi \) | Infrared 4th positive | \( X^1\Sigma^+ - X^1\Sigma^+ \) \( A^1\Pi - X^1\Sigma^+ \) | [44] [45] | (30,29) (20,20) 200.000 110.000 |
| \( \text{C}_2 \) | \( a^3\Pi_u \) \( d^3\Pi^+_g \) | Swan | \( d^3\Pi^+_g - a^3\Pi_u \) | [46] [47] | (20,20) 250.000 |

The integrated line intensity is proportional to the product of the vibrational transition probability (Einstein coefficients \( A_{v',v''} \)) and the Hönl-London factors \( S^{N,j'}_{N,j} \) which define the relative rotational intensity between transitions allowed by the selection rules. Hönl-London factors used for this work are referenced in Table 2. All are normalized with the Whiting convention:

\[ \sum_{j'} S^{N,j'}_{N,j} = (2 - \delta_{0,A} \delta_{0,A^*})(2S'+1)(2N'+1) \]  

(4)

Some Einstein coefficients and absorption oscillator strengths are available in the literature and were deduced from calculations or experiment measurements. Unfortunately, the comparison of the references showed some discrepancies at rather high vibrational levels, except for the case of \( \text{C}_2 \) Swan system. Consequently, we decided to use the published values for \( \text{C}_2 \) Swan system and to set up the classical method of calculation vibrational transition probabilities for the electronic systems IR and 4th positive of \( \text{CO} \) and for the Schumann-Runge system of \( \text{O}_2 \). We used the well-known RKR method [47] in the formalism of Zare [48], which consists in the reconstruction of the internuclear potentials by inversion of the equilibrium spectroscopic constants for each electronic state. Out of the experimental range of internuclear distance validity, we extrapolated the RKR potential with a repulsive potential \( V_{\text{rep}}(r) = ar^{-b} \) at low distance, and with a Hulbert-Hirschfelder (HH) at large distance. Then, this potential was injected in the radial Schrödinger equation to calculate the electronic wave functions using a Numerov iteration algorithm (two-step) by a method based on the works of Cooley [49]. This calculation was achieved for all couple \((v',v'')\) up to the limit of dissociation (a cut off could be done when the high vibrational levels have no significant interest for this work). Knowing the rotationless vibrational wave functions for all upper vibrational levels \( \psi_{v'}(r) \) and all lower \( \psi_{v''}(r) \) vibrational, the vibrational transition probability between two vibrational states is obtained by the formula:
The electronic transition dipole moments $R_e(r)$ used in the calculation of each system are referenced in Table 2. Einstein coefficients obtained were validated by comparison with experimental lifetimes or partial Einstein coefficients of absorption oscillator strength published in the literature. Furthermore, we obtained very accurate results for vibrational energy levels eigenvalues by our method compared to the experimental energy obtained with the Klein-Dunham formalism, which tends to valid the wave functions and the RKR potentials obtained.

Finally, the line shape of each line was calculated by a Voigt profile, taking into account the Doppler and the collisional broadenings. Doppler broadening for molecules obeys to the same formula as the atomic case [34]. For collisional broadening and species involved in this work, we did not find information in the literature with a sufficient pressure and temperature validity range. Consequently, we used systematically, for all the diatomic molecular bands, the semi-empirical approximation law for molecular collisional broadening HWHM (Half width at Half Maximum) $\gamma_c(T,P) = \gamma_{\text{ref}} \cdot P(T_{\text{ref}}/T)^n$ extended to be pressure dependent. We used in a first approximation the order of magnitude defined by Breene [50] ($\gamma_{\text{ref}} = 0.1$ and $n=0.7$) for the broadening parameters of all molecules.

**Table 2. Molecular systems**

| Molecule | Band system | Einstein coefficient | Hönl-London factors | Dipole moment function Re(r) |
|----------|-------------|----------------------|---------------------|----------------------------|
| O₂       | Schumann-runge | This work [52]       | [55]                |                            |
| CO       | Infrared    | This work [53]       | [56]                |                            |
| C₂       | 4th positive | This work [53]       | [57]                |                            |
|          | Swan        | [51]                 | [54]                | -                          |

### 3.4.2. Linear triatomic contribution: CO₂

The emission of CO₂ infrared bands was obtained using two databases: CDSD-1000 for the temperature range 500-2000K and CDSD-4000 for the temperature range 2000-5000K [58]. Those databases give us the required parameters to compute the emission and the absorption spectra of CO₂. It contains for each rovibrational line: the band position, its integrated intensity (in cm⁻¹ / (molecule.cm⁻²) at 296 K), the rovibrational transition probability in s⁻¹, the energy of the lower state, and the broadenings parameters. The CDSD-4000 is not convenient to be used in routine because it contains 628.324.454 integrated rovibrational line entries, which require at least a thousand billions of points to compute. Therefore, we decided to set up partial databases by steps of 500K to keep only lines contributing significantly to the radiation in this range of temperature. It can be reduced to around 15.000.000 integrated lines for each temperature, leading to a difference of more than 0.1% of the total emission.

### 4. The Net Emission Coefficient (NEC)

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

$$\varepsilon_N(T, R_p) = \int_0^\infty L_\lambda^0(T) \cdot K_\lambda(T) \cdot \exp \left( -K_\lambda(T) \cdot R_p \right) \, d\lambda$$

where $\lambda$ (m) is the wavelength, T and P the local temperature and pressure respectively, $L_\lambda^0$ (W/m²/sr/m) the Planck function, $R_p$ is the size of the sphere assimilated to the plasma’s size and $K_\lambda$ (m⁻¹) the monochromatic absorption coefficient corrected by the induced emission and correlated with
the local emission coefficient by the Kirchhoff law. Moreover, due to the exponential term, self-absorption of radiation in the emitting regions is rather well estimated. In numerical modelling, the NEC is a good method to characterize the radiation losses in hot regions while taking into account the absorption. Our last studies also showed that this coefficient depends not only on the temperature, the pressure and the plasma size but also on the presence of the molecules or metallic vapours for low temperatures. Indeed, Figure 3 shows the NEC and the different contributions obtained in the case of pure CO2 at atmospheric pressure and two plasma’s sizes: optically thin plasma and 1mm of thickness.

For optically thin plasmas (R_p = 0mm), we observe that the radiation of the molecular bands represents the only contribution up to 5kK, and shares the radiation with the atomic lines up to 10kK with more than 50% at 6.9kK. For this size of plasma, the continuum is negligible. In the case of R_p≠0mm, the lines are strongly self-absorbed in the first millimeter which can explain the reason why their contributions decrease with only 60% at 10kK against 97% for R_p=0mm. The consequence of this absorption is that the contribution of the molecular bands, which are less absorbed, always plays an important role at 7.5kK and that the contribution of the continuum can represent up to 40% of the NEC between 10kK and 15kK.

Always in the case of pure CO2, we compare on Figure 4 the contribution of the 4 molecular systems (O_2, CO, CO_2 and C_2 molecules) to the total radiation of the molecular bands for pure CO2 and two values of R_p. We observe that the radiation of the molecular bands is mainly due to the CO_2 system for temperatures lower than 3.5K-4kK, CO IR for temperatures between 3.5kK-5kK, and 4th CO for higher temperatures. However, if the plasma’s size increases, the contributions of CO_IR and 4th CO seem to decrease to the detriment of O_2 (SR) and CO_2. Nevertheless, if the plasma’s size is inferior to 5mm, the molecular systems of C_2 (Swan) and O_2 (SR) do not play an important part in pure CO2 radiation.

Finally, Figure 5 compares the NEC of pure CO2 plasma with the NEC of mixtures containing various concentrations of copper for R_p=5mm. We observe that a small concentration of metallic vapours (few percents) increases the radiation of the plasma for temperatures ranging from 5000K to 10000K. This increase is more visible for low sizes of plasma since the absorption minimizes the influence of the
resonance lines of the metallic vapours species. At lower temperature, the increase of the plasma’s size tends to diminish the important role of the metallic lines to the detriment of the molecular bands radiation which is less absorbed.

![Diagram](image)

**Figure 4.** Contribution of the molecular systems to the total radiation of the molecular bands for pure CO₂, the atmospheric pressure and two values of \( R_p \).

![Diagram](image)

**Figure 5.** NEC for different concentration of metallic vapors in CO₂-Cu mixtures at \( R_p=5\text{mm} \) and atmospheric pressure.

5. **Conclusions**

This paper proposed a study of the radiative properties for CO₂-Cu thermal plasmas calculated according to the method of the Net Emission Coefficient, for temperatures between 500K and 30000K,
and at the atmospheric pressure. The results showed that the NEC was mainly due to the emission of the molecular bands which should be considered in the NEC calculation for temperatures inferior to 4000K and all plasma sizes, then due to the emission of the atomic lines for higher temperatures and optically thin plasmas, and finally due to the emissions of the atomic lines and of the continuum for higher temperatures and higher plasma’s sizes.

In pure CO₂, we also showed that C₂ emission was negligible whatever the plasma size and that CO and CO₂ emissions were the main contributions to the molecular bands emission for optically thin plasmas. For higher thickness of plasma Rₚ, the emission of CO became less important to the detriment of the O₂ SR emission. In CO₂-Cu mixtures, we highlighted that the continuum could represent up to 10% of the NEC for concentrations of vapours inferior to 10% and optically thin plasmas. We also put in evidence the decrease of the molecular bands emission with the increase of the metal vapours in the plasma. When the thickness of plasma increases, the results showed that the molecular bands and the continuum contributed more significantly to the NEC due to a strong self-absorption of the emission of the resonance atomic lines. In this case, the continuum could represent up to 45% of the NEC for the intermediate temperatures.

6. References

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