Plasma Chem Plasma Process (2017) 37:581–600
DOI 10.1007/s11090-016-9761-y

ORIGINAL PAPER

Effect of Metal Vapours on the Radiation Properties of Thermal Plasmas

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Received: 17 October 2016 / Accepted: 6 November 2016 / Published online: 16 November 2016

Abstract In metallurgic applications of thermal plasmas the presence of metal vapour, even in small proportion tends to increase the electron number density and to modify some basic properties such as the electrical conductivity and the radiation emission. In this paper we focus on the influence of these vapours on the radiation properties. After the definition of some necessary and basic functions and laws we briefly present the mechanisms responsible for emission and absorption of radiation in thermal plasmas. Then an important section is devoted to the role of metal vapours on the net emission coefficient which is the most popular parameter used to evaluate the radiation power losses in general models. It is shown that metal vapours increase the emission especially at low and intermediate temperatures ($T < 12{,}000$ K) and that their relative influence depends on the nature of the initial gas and of the metal itself. We list a rather important number of references presenting calculation of net emission in various gas–metal mixtures. Finally we show in a last section the influence of metal radiation on general plasma properties such as the energy transfer (other methods than the net emission coefficient), the cooling effect, the global energy balance and the heating of particulates injected in the plasma. The most spectacular effects are the increase of radiation losses in the energy balance and the complex role of the metal in the local cooling of the plasma.

Keywords Radiation · Thermal plasmas · Metal vapour · Net emission coefficient

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Introduction

Thermal plasmas are often used in metallurgical applications such as welding, cutting or plasma spraying (see other papers in this special issue). Obviously in these conditions, metallic materials are in contact with thermal plasmas whose temperature is of the order of 10,000 K leading to an injection of metallic vapours within the plasma. Furthermore the electrodes of electric arcs are in general ablated, particularly at high current, even if the arc duration is short as it is the case for the circuit-breakers arcs. Hence the presence of metallic vapours is often observed in thermal plasmas.

The composition, the thermodynamic properties and the transport coefficients of a plasma depend strongly on the nature of the gas or of the mixture of gases constituting the plasma, especially when the temperature is lower than 15,000 K [1]. Let us note that at higher temperature the plasma is strongly ionized and its properties depend slowly on the nature of gases. So, when the proportion of a gas in a mixture is important, all the material properties of a plasma are affected by the presence of this gas. On the contrary when this proportion is of the order of 1% the plasma properties are (in general) almost unchanged. But this is not true for metallic vapours. Indeed as it can be seen in Fig. 1 showing the evolution of the electron number density in Ar–metal plasmas [2], the presence of a small amount of metal vapour increases this function at low or intermediate temperature ($T \leq 10{,}000$ K). This kind of result has been obtained performing the equilibrium composition calculation whose bases are well known [1]. The increase of the electron number density n_e due to the presence of the metal can be explained by the low ionization energies of these species (see Table 1 showing this energy for various neutral species) favouring the ionization of the gas following the Saha's law [1]. Figure 1 shows clearly the important effect of the metal at low temperature even for proportions lower than 1%. As expected, the effect increases with the metal proportion, but tends to disappear at high temperature ($T > 12{,}000$ K) because all the species become rather strongly ionized. It must be added that the influence of the metal vapour on n_e, is less important in air plasma than in argon

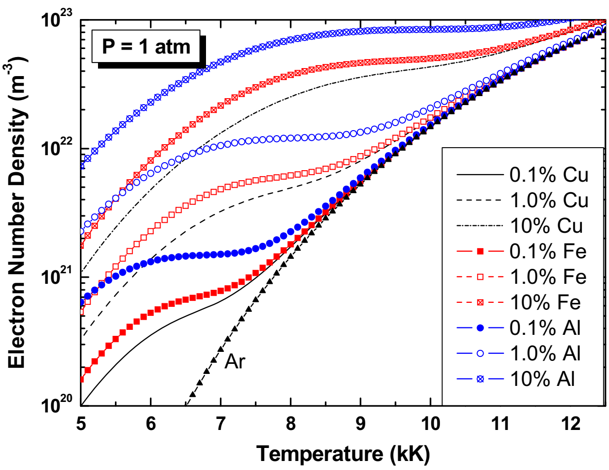

Fig. 1 Influence of the presence of metal vapours (mass proportions) on the variations of the electron number density, for an argon plasma (from [2]) at atmospheric pressure ($P = 1.013 \times 10^5$ Pa) (© IOP Publishing. Reproduced with permission. All rights reserved)

Table 1 Ionisation energy (in eV) of various neutral species

Ar	C	H	N	O	Ag	Al	Cu	Fe
15.759	11.260	13.598	14.534	13.618	7.576	5.985	7.726	7.902

plasma because the difference between the ionization energies of the metals and those of the gas components is smaller with air (N, O) than with argon, as it can be seen in Table 1.

As a consequence the effect of metal vapours on n_e at low temperature tends to modify specific properties in this temperature range. Among these properties, the radiation emission is very sensitive to the nature of the gas or vapour, reason why we present this paper on the influence of metal on the radiative transfer in thermal plasmas, influence that can be decisive even for small proportion of metal.

The main assumption used in this study and in literature cited in this paper is that the plasma is in Local Thermodynamic Equilibrium (LTE). This assumption allows first using the equilibrium composition giving the population of the number densities of the plasma components as illustrated in Fig. 2 for air–Al plasma [3]. The populations of the excited states can then be deduced using the Boltzmann's law and the partition functions previously calculated (see [1–3] for the methods). The second relation available in LTE conditions is the Kirchhoff's law, very important for calculating the radiative transfer and that will be defined in "Bases of Radiation Phenomena and Transport" section.

As this paper takes part of a special issue in homage of Prof. Pfender, it must be noted that E. Pfender was a pioneer in the field of thermal plasmas research and development and that among the many works he performed we must emphasis the study of energy transfer [4, 5] and specially the radiative transfer [6, 7] even in presence of metallic vapours [8, 9]. These two last references will be commented in "Net Emission Coefficient: Influence of Metal Vapours" and "Role of Metal Radiation on General Thermal Plasmas Properties" sections.

"Bases of Radiation Phenomena and Transport" section of this paper is devoted to a rather brief description of the bases of radiation transport and phenomena. The most

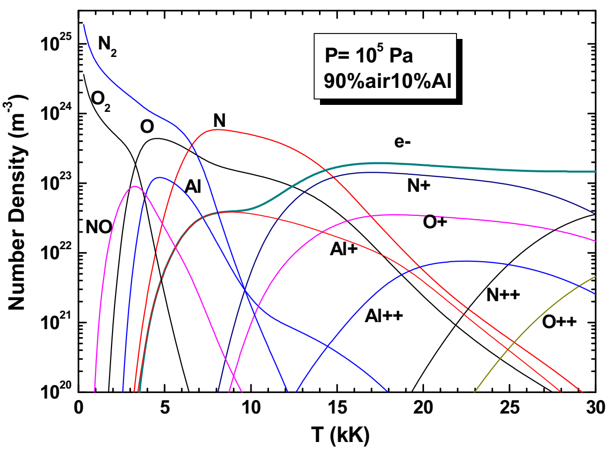

Fig. 2 Equilibrium composition of a 90%air–10%Al (mass proportions) plasma (from [3]) (P = 10^5 Pa)

popular and simplest method to compute the radiation transfer in thermal plasmas in presence of metallic vapours is the net emission coefficient (NEC). This method and the most characteristics results are detailed in "Net Emission Coefficient: Influence of Metal Vapours" section. Some recent results concerning other methods of radiative transfer calculation based on the use of mean absorption coefficients (MAC) are also presented in this section. "Role of Metal Radiation on General Thermal Plasmas Properties" section deals with the study of the influence of the metal vapours on the behaviour of thermal plasmas systems showing the important role of radiation on the plasma parameters (temperature for example) and on the global energy balance. The main part of this paper corresponds to a review from literature but some new and original results are included in "Net Emission Coefficient: Influence of Metal Vapours" section and in the presentation of the MAC.

It has to be noted that metallic species and their eventual by-products (oxides, carbides, nitrides, …) are in general in condensed form at low temperature (liquid or solid). As we are concerned with plasmas in gaseous phase, we will present results on radiation properties and radiative transfer in presence of metal vapours, for temperature values higher than roughly 2000 K.

Bases of Radiation Phenomena and Transport

General Laws and Definitions

The radiation intensity at a point P and in the direction $\vec{s}$ (see Fig. 3) is the power crossing the elementary surface area $dS_{\perp}$ (apparent surface perpendicular to the direction $\vec{s}$) and per solid angle unit. The spectral intensity I_{λ} refers to radiation at a given wavelength λ (in fact in an interval $d\lambda$ around λ) while the total intensity I is an integration of the previous one, over the total spectrum, i.e. including all wavelengths.

Let us remember that the intensity of a blackbody noted $I_{b\lambda}$ is isotropic. It is given by the *Planck's law* and the form depends on the spectral variable. For the frequency v:

$$I_{bv}(T) = \frac{2hv^3n^2}{c_o^2 \cdot (e^{hv/kT} - 1)} \tag{1}$$

where T is the temperature, k the Boltzmann constant and n the refractive index, very near from 1 for a plasma and often omitted. The expressions of the blackbody intensity for the other spectral variables are deduced from Eqs. (1) and from the relationship:

$$I_{bv}dv = I_{b\lambda}d\lambda = I_{b\eta}d\eta \tag{2}$$

We must distinguish the *radiation intensity* defined on the basis of projected area, and the *emissive power* which is the power emitted in a given direction per unit of actual

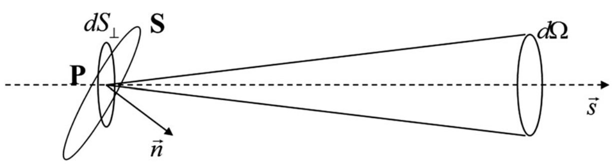

Fig. 3 Definition of the radiation intensity

(unprojected) surface area. From Fig. 1, and defining the angle θ between the directions $\vec{s}$ and $\vec{n}$, the emissive power E (see Modest [10] and Siegel and Howell [11] for a detailed analysis) is given by:

$$E_\lambda = I_\lambda \cos\theta \tag{3}$$

Hence, for the blackbody isotropic radiation, the *hemispherical spectral emissive power* $E_{b\lambda}$ is the power emitted by a black surface per unit area and per unit wavelength, integrating over all the solid angles of the hemisphere:

$$E_{b\lambda} = \pi \cdot I_{b\lambda} \quad \text{or} \quad E_{bv} = \pi \cdot I_{bv} \tag{4}$$

The *Planck's law* can be written on the form of the blackbody *hemispherical spectral emissive power*. As an example for the frequency:

$$E_{bv}(T) = \frac{2\pi h v^3 n^2}{c_o^2 \cdot \left(e^{hv/kT} - 1\right)} \tag{5}$$

The total blackbody emissive power or intensity can be calculated by integrating over all the frequencies (or wavelengths). The total hemispherical emissive power of a blackbody is then given by the well-known *Stefan–Boltzmann's law*:

$$E_b(T) = \sigma_S T^4 \tag{6}$$

where σ_S is the *Stefan–Boltzmann constant* equal to 5.6704×10^{-8} W m^{-2} K^{-4}.

Obviously thermal plasma is not a blackbody and the radiation intensity within discharge thermal plasma is far from the Planck intensity. Nevertheless, as a good approximation, thermal plasmas are often considered in the state of local thermodynamic equilibrium (LTE) which allows using the Kirchhoff's law. To introduce this law two other functions must be defined, the absorption and the emission coefficients.

The absorption coefficient K corresponds to the attenuation of a radiation beam along a distance x, following the expression:

$$I = I_o \exp(-K \cdot x) \tag{7}$$

I_0 being the incident intensity. K is the inverse of a length (unit m^{-1}). This coefficient is a property of the gas or plasma and it depends strongly on the nature of the gases, on the physical conditions (temperature, pressure) and on the spectral variable.

The emission of an isothermal volume element is the radiation power emitted by this volume and can be defined as $4\pi\varepsilon$, ε being the emission coefficient and the term 4π representing the integration of the isotropic intensity over the total solid angle. The unit of $4\pi\varepsilon$ is W m^{-3} for the total radiation (integrated on the spectral variable) or W m^{-3} s for the spectral radiation (with frequency here).

Considering the microreversibility between emission and absorption in equilibrium conditions [10, 11], it can be demonstrated that the emission and absorption coefficients are linked with the blackbody intensity, by the following relation.

$$\varepsilon_v = K(v) \cdot I_{bv} \tag{8}$$

This form of Kirchhoff's law is very important for studying thermal plasmas because it is valid in LTE conditions and not only in complete equilibrium. Finally let us introduce the induced emission which corresponds to the emission of a photon stimulated by a photon from the radiation field. It acts as a negative absorption. For this reason, the true absorption coefficient $K(v)$ is corrected in order to take into account the induced emission. In LTE conditions, the corrected absorption coefficient $K'(v)$ is written:

$$K'(v) = K(v) \cdot \left(1 - \exp\left(-\frac{hv}{kT}\right)\right) \tag{9}$$

Emission and Absorption Phenomena in Thermal Plasmas

We present here some generalities on these phenomena, details can be found in the corresponding references. In thermal plasma radiation can be emitted or absorbed following various kinds of mechanisms that can be classified in 4 categories:

Atomic Continuum

The reactions involve free electrons and atomic particles (neutral or ions).

Bremsstrahlung A free electron in the field of an ion or an atom may be slowed down and therefore loses energy under the form of radiation. Approximate expressions are given in [12, 13]. As the mean kinetic energy of the electrons is of the order of 1–2 eV, the photons emitted by these mechanisms have in general a limited energy, of the order or less than 1 eV, so that this radiation is mainly located in the infrared (IR) part of the spectrum.

Radiative Recombination It corresponds to the interaction of a free electron with a positive atomic ion. The emission coefficient of recombination is given by Cabannes and Chapelle [12] and Gleizes et al. [13]. The non-hydrogenic structure of the elements is taken into account with the Biberman-Schlüter factor determined for a lot of species [14–16]. Data for metallic species can be found in [2, 3, 18]. A large amount of data on photoionisation cross sections is available from the Opacity Project and TOPbase site [17].

Radiative Attachment This phenomenon only occurs in presence of an electro-negative species and leads to a continuum emission, the emitted photons having energy higher than the attachment energy. The total emission due to this mechanism is always small in comparison with recombination.

Molecular Continuum

Due to the high temperature in central regions, the molecules, if they exist, are in general located in the external regions, and their role in the continuum component is mainly to absorb radiation through two mechanisms: photodissociation and photoionisation. For a given molecule these reactions are grouped and represented by a global absorption coefficient deduced from a global cross section [19, 20]. More complex examples are given in [21].

Atomic Lines

They correspond to radiative transitions between excited levels of a same atomic species and they often constitute the most important part of radiation transfer in thermal plasmas. The local total intensity of a line is simple to compute because it is proportional to the population of the upper state number density (deduced from the equilibrium composition) and of the spontaneous emission coefficient whose values can be found in literature, for a lot of neutral and ionized atoms. In particular a very large compilation and useful data from experimental and theoretical works are given by the NIST Institute that can be completed by Kurucz's database [22, 23].

But the main problem in the treatment of the lines is the line profile: because of the exponential form of the radiation transmission (Eq. 7) the central part of the line profile can be strongly attenuated. Hence the study of line broadenings is essential and various experimental or theoretical approximations must be used from literature (see specific references in [2, 3, 8, 18–20, 24]).

Molecular Bands

The molecular structure is much more complicated than the atomic one because more degrees of freedom are concerned. For example, for a diatomic molecule, vibration and rotation of the atoms are added to the electronic excitation involved in atomic structure. All the additional phenomena are quantified, and the allowed transitions between these energy levels lead to a great number of lines that are grouped in "bands". The fine description of this molecular structure is out of the scope of this paper and is described in special books, the most famous are those written by Herzberg [25].

For diatomic molecules some codes or databases useful to compute the spectral properties are available for specific species, based on scientific articles [26, 27]. It must be added that several hundreds of papers have been published on the spectral description of radiation properties of atoms and molecules.

Total Spectra

The most useful description of radiation for a gas or plasma is the spectral variation of the absorption coefficient for a given general composition. The best method to take into account the atomic lines and the molecular bands is called the "line by line" method. This kind of calculation has been made for plasmas in atomic gases such as argon and metallic vapor [8, 9] but the results are more spectacular with molecular gases as it is the case for air plasmas [19, 20, 28].

The influence of the presence of copper vapour on the total spectrum of an air plasma is illustrated in Fig. 4 from [29]: this influence is marked at low and intermediate temperature due in particular to resonance lines. At high temperature the relative influence is not so important. We can see in Fig. 5 from [30] that the quantitative influence of the metal depends on the nature of this metal. In this example the spectrum of tungsten contains much more lines than that of copper. We have seen in "Introduction" section that the ionisation potential is another parameter explaining the role of vapour radiation.

Net Emission Coefficient: Influence of Metal Vapours

Definitions

Assuming a non-scattering medium, the equation of radiative transfer along the direction s, can be written [11]:

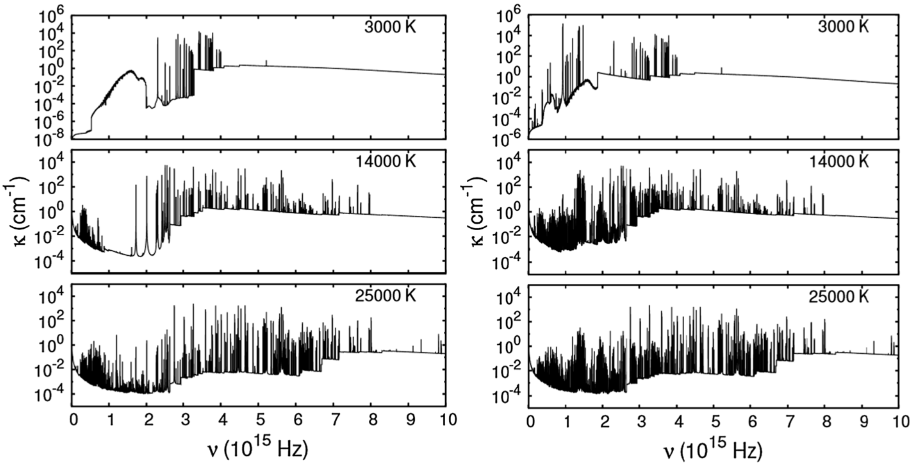

Fig. 4 Absorption coefficient of pure air (*left hand side*) and air with 20mass% copper vapour (*right hand side*) as the function of frequency and temperature (from [29])

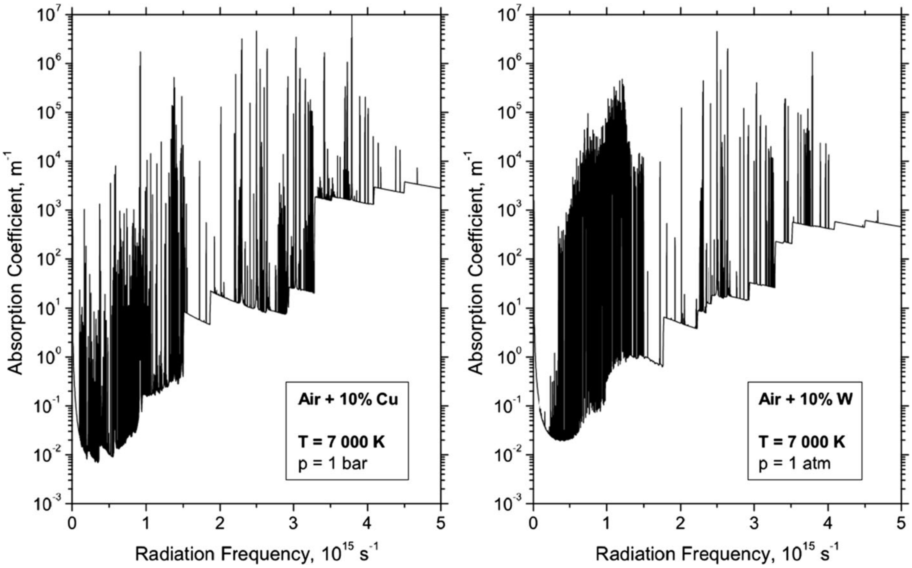

Fig. 5 Total absorption coefficient for the air plasma with admixtures of Cu and W at temperatures of 7000 K, from [30] (© IOP Publishing. Reproduced with permission. All rights reserved)

$$\frac{dI_\lambda}{ds} = -K(\lambda, s) \cdot I_\lambda(s) + K(\lambda) \cdot I_{b\lambda}(s) \tag{10}$$

This equation has been written here for a monochromatic radiation and assuming that the refractive index of the plasma is 1. $K(\lambda)$ is the absorption coefficient corrected with the induced emission: for simplifying K' is replaced by K. Let us consider now the radiative term in the energy conservation equation: in an elementary volume dV per time unit, this term is the difference between the power deposited in dV and the power emitted by this volume ("net" power). The radiative flux vector $\vec{q}_r$ can be defined in the direction $\vec{s}$ across an elementary area dA taking into account all the directions of the incident intensities [11]:

$$\vec{q}_r = \int_{\omega=o}^{4\pi} I(x,y,z) \cdot \vec{s} \cdot d\omega \tag{11}$$

Hence the radiation energy term required in the energy conservation equation is the net radiative balance which is the divergence of the radiative flux that can be expressed by:

$$-\vec{\nabla} \cdot \vec{q}_r = \int_{\lambda=o}^{\infty} K(\lambda) \left(\int_{\omega=o}^{4\pi} I_\lambda(\omega) \cdot d\omega \right) \cdot d\lambda - 4\pi \int_{\lambda=o}^{\infty} K(\lambda) \cdot I_{b\lambda} \cdot d\lambda \tag{12}$$

The calculation of the divergence of the radiation flux is very complex in practical conditions. This calculation can be performed assuming severe added assumptions. Two kinds of treatment can be schematically defined: the first one is a simplification of the spatial or geometrical conditions; the second one is a simplification in the spectral description of radiation.

A very simple but useful treatment of the radiation term in thermal plasma modeling is called the "net emission coefficient" (NEC) introduced by Lowke [31]. The main assumption is that the plasma is considered isothermal and homogeneous (in case of mixtures). The second condition is a simplified geometry. Let us consider a sphere of radius R_p, the NEC ε_N is defined as the divergence of the radiative flux per solid angle unit that means that the radiation term in the energy equation will be $-4\pi\varepsilon_N$. The minus sign is due to the fact that in the hottest regions of the plasma the radiation is an energy loss phenomenon.

From Eq. 12, considering an isothermal spherical plasma, the NEC is written:

$$\begin{aligned} \varepsilon_N(T, R_p) \equiv \frac{\vec{\nabla} \cdot \vec{q}_r}{4\pi} &= \int_{\lambda=o}^{\infty} K(\lambda) \cdot I_{b\lambda} \cdot d\lambda - \int_{\lambda=o}^{\infty} K(\lambda) \cdot I_{b\lambda} \cdot \left(1 - e^{-K(\lambda)\cdot R_p}\right) \cdot d\lambda \\ &= \int_{\lambda=o}^{\infty} K(\lambda) \cdot I_{b\lambda} \cdot e^{-K(\lambda)\cdot R_p} \cdot d\lambda \end{aligned} \tag{13}$$

The interests of the use of the NEC in modelling are its simplicity and its good accuracy for evaluating the radiative losses from the hottest regions. Its computation using Eq. 13 is rather long but it can be performed independently of the general modelling and the NEC can be tabulated as a function of the temperature T and of the radius R_p for a given gas (or mixture of gases) at a given pressure.

Properties of the Net Emission Coefficient

The NEC can be calculated using two methods:

- Direct integration (Eq. 13) with a very small wavelength step. It takes between 200,000 spectral points (for Ar) to more than one million points (for molecular plasmas with diatomic bands) to have a fine description ("line by line") of the absorption coefficient corresponding to Figs. 4 and 5.
- The second is a method requiring much less cpu time for the computation and assuming that the line overlapping has no influence on the radiative transfer [2–24], using an escape factor for each line.

The use of escape factors tends to overestimate the NEC because the overlapping of the lines not taken into account through the escape factors, tends to attenuate the transferred radiation. At atmospheric pressure and for a plasma size of the order of a few mm, the error is acceptable, but it increases at very high pressure [32] and for high values of R_p [8, 32].

As the object of this paper is to show the influence of metal vapors on radiation properties we will present the variations of the NEC in presence of these vapors. Figure 6 from [30] shows the variations of the NEC for a mixture 90%air–10%Cu (mole or volume proportions). The case $R_p = 0$ is a fictitious case assuming that radiation is not absorbed (exponential term in (12) is equal to 1). With this Fig. 6, we can see two general properties of the NEC whatever the gases or vapours involved: fast increase of the NEC with temperature up to 10–12 kK, this threshold depending on the nature of the species; strong absorption of radiation, even in the first tenth of mm.

The influence of the presence of metal vapour on the NEC is illustrated in Figs. 7 and 8, showing that the radiation of a plasma (here argon or air) is strongly enhanced by the metal vapour at low and intermediate temperature. This is due to the low ionisation potentials of the metals (see Table 1) leading to an increase of the number densities of the electrons and of the atomic (neutral or ions) excited levels at a given temperature in comparison with those of the pure gas.

Apart from the quantitative values of the NEC, Figs. 7 and 8 give some general information:

- The NEC of the metal does not only depend on the ionisation potential, but also on the atomic structure: Fe is more emissive than Ag or Cu because of its rich line spectrum.
- The relative influence of the metal depends also on the initial gas. For example, air is more emissive than Ar (at a given temperature) so that the same amount of a metal vapour has a relative effect greater with Ar than with air.
- At low temperature where the NEC of the pure gas is negligible in front of the metal NEC, the NEC of the mixture is roughly proportional to the metal partial pressure (this is not an exact law but it could be quite convenient in practical applications).

Figure 9a shows the influence of pressure on the variations of the NEC given by Aubrecht et al. [30]. The presence of metal does not change the general behaviour observed for pure

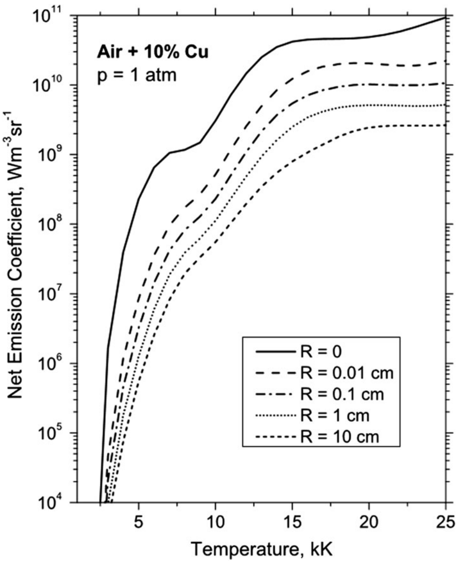

Fig. 6 Net emission coefficient of a mixture 90%air–10%Cu (molar proportions) for various values of plasma radius (from [30]) (P = 10^5 Pa) (© IOP Publishing. Reproduced with permission. All rights reserved)

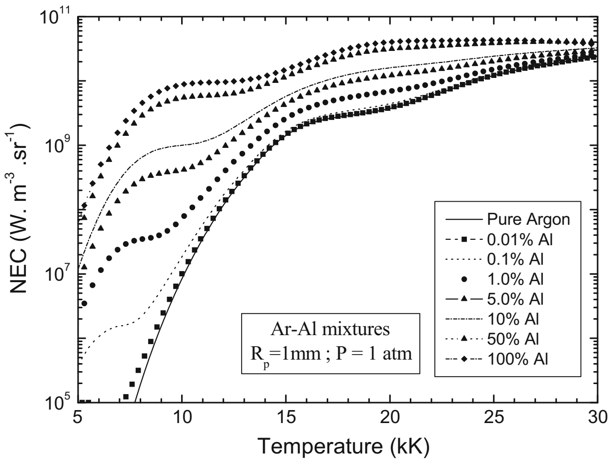

Fig. 7 Net emission coefficient for Ar–Al mixtures (mass proportions) [2] (© IOP Publishing. Reproduced with permission. All rights reserved)

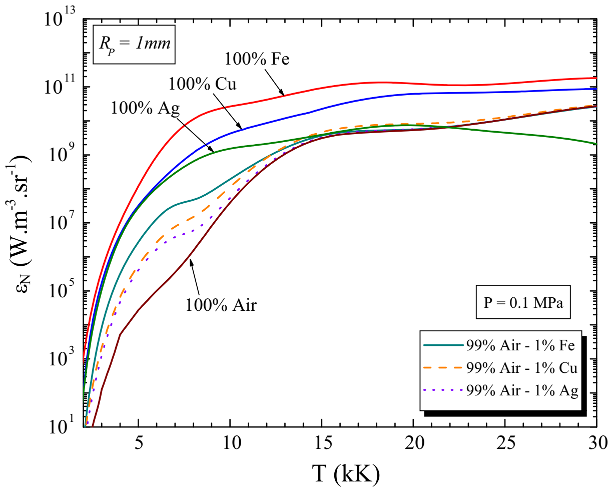

Fig. 8 Net emission coefficient for air–metal mixtures (mass proportions) [18] (© IOP Publishing. Reproduced with permission. All rights reserved)

gases. In the low and intermediate temperature range (T < 15 kK) the NEC is rather proportional to pressure which is not exactly the case at higher temperature (easy to see at 20 kK on Fig. 9). This evolution is connected to the variations of the equilibrium composition with pressure, and in particular to the variations of the electron number density.

We present on Fig. 9b some original unpublished results showing the influence of pressure on the NEC of an air–5%iron mixture (mass proportion). They have been obtained following the bases of paper [18], and they present a similar behaviour.

Several teams have calculated the NEC for metal containing thermal plasmas and we have indicated in Table 2 the corresponding references classifying them following the metal nature. In general the temperature range lies between 2 and 30 kK.

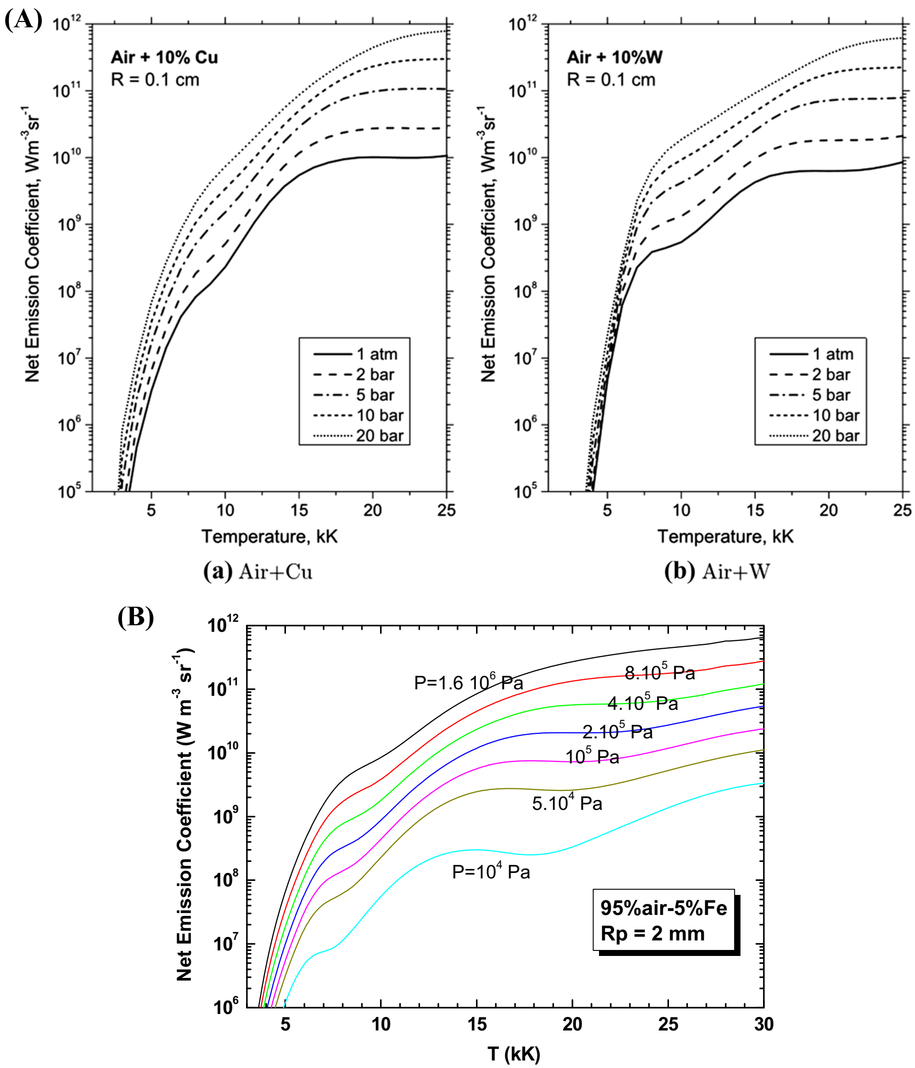

Fig. 9 **a** Influence of pressure on the NEC (from [30]) (in units of bar with 1 bar = 10^5 Pa) **b** Influence of pressure on the NEC for air–Fe mixture

Role of Metal Radiation on General Thermal Plasmas Properties

Radiative Transfer

The NEC allows evaluating the radiation power escaping the hottest regions of the plasma but it is not able to determine the total radiative transfer, and in particular not able to calculate the part of radiation absorbed in the external regions. This knowledge is often not necessary because in general radiation does not play an essential role in the energy balance of the external regions, but in certain cases it is important to calculate this absorption (high

Table 2 Bibliography on the calculation of net emission coefficients for metal containing thermal plasmas

Metal	Mixtures	References
Ag	Air–Ag	[18]
Al	Ar–Al	[2, 35]
	Air–Al	[3]
Ca	Complex mixture	[40]
Cu	Ar–Cu	[2, 8, 34, 44]
	N_2–Cu	[34, 50]
	Air–Cu	[18, 29, 30]
	SF_6–Cu	[34, 37]
	CO_2–Cu	[42]
	Ar–H_2–Cu	[38]
	Air–PA66–Cu	[41]
Fe	Fe	[45]
	Ar–Fe	[2, 33–36, 47, 48]
	Air–Fe	[18]
	Ar–H_2–Fe	[36]
	He–Fe	[46, 47]
	N_2–Fe	[49]
Mg	Complex mixture	[40]
Na	Complex mixture	[40]
Si	Ar–Si	[35]
W	Air–W	[30]
	N_2–W	[50]
Sr, Tl, Dy	With Ar–Hg	[43]
Co, Ni	With C–He–N_2	[65]

voltage circuit-breaker for example) and/or to estimate the radiation flux escaping the plasma, responsible for material ablation rather far from the plasma. This problem has been solved for several molecular gases such as air and SF_6 using in particular P1 method or Discrete Ordinates Method (DOM) (other methods have been explored for thermal plasmas but are less efficient). But the study of radiative transfer in presence of metal vapour has not been strongly developed. This is mainly due to the fact that the presence of metal enhanced the emission from the hottest regions but changes only weakly the absorption of radiation in the other zones, and the use of the NEC is often sufficient to take this effect into account. Nevertheless we may mention first, the work proposed by the team of profs. Pfender and Heberlein (Menart et al. [9]) using a line by line method rigorously applied to a pure argon arc and applied to an Ar–Cu arc with simplifications (same temperature field and uniform repartition of Cu).

In iterative procedure used in a general modelling, the computation of the radiative flux and of its divergence cannot be performed for all the spectral points (of the order of 1 million points for a given temperature!). Hence a simplified spectral description must be used and in thermal plasmas it consists to calculate the Mean Absorption Coefficient (MAC) on a few spectral intervals, which is not trivial [21, 32, 51, 52]. These values of the MAC can then be applied to compute the radiative transfer using the P1-method as it was the case for metallic halogenates lamp [39] in a general modelling. For mixtures with metals others than that studied in [39] only a few variation of the MAC have been published (and sometimes associated with the P1-model in simplified cases of imposed temperature field) such as Ar–Fe [53], SF_6–Cu [54] and complex mixtures with salted water [55].

We present in Figs. 10 and 11 some new results relative to the MAC we have calculated for air–iron mixtures following paper [18] for the bases of radiation properties calculation. We have used the Planck MAC defined in a given spectral interval $[\lambda_1 - \lambda_2]$ by:

$$\bar{K} = \frac{\int_{\lambda_1}^{\lambda_2} K_\lambda I_{b\lambda} d\lambda}{\int_{\lambda_1}^{\lambda_2} I_{b\lambda} d\lambda} \tag{14}$$

where $I_{b\lambda}$ is the blackbody intensity. We have determined 6 spectral intervals noted 1–6 in the figures with limits given in Table 3.

We can see from Fig. 10 that for the two first intervals corresponding to the highest photon energies, the values of the MAC are not influenced by the presence of iron vapour. This is not the case for the other intervals in particular for the large interval number 5 covering all the visible part and the non-absorbed UV part, where is emitted a very large number of iron lines. The relative influence of iron proportion for intervals 3–6 is detailed in Fig. 11.

Cooling Effect

As seen in Figs. 7 and 8 a weak amount of metal vapour may greatly enhance the radiation emission. But in practical conditions such as arcs in metallurgy the theoretical curves of Figs. 7 and 8 must be coupled with the true temperature field because in general metal vapour tends to cool down the plasma for a fixed current intensity (if an arc) or a fixed input power. In fact two mechanisms are involved in the cooling. The first one is the increase of radiation power losses leading directly to the cooling. The second phenomenon is indirect and is correlated to the increase of the electrical conductivity (due to the low ionization potential of the metal) which tends to enhance the conduction radius of the arc and then to decrease the temperature in the hottest regions. These effects have been largely

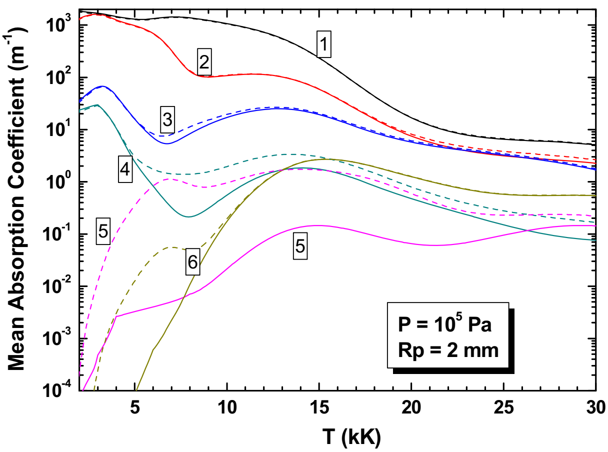

Fig. 10 Influence of iron on the mean absorption coefficient of air for 6 spectral intervals (see Table 3 for the intervals). Solid line: pure air; dashed lines: 95%air–5%Fe (in mass)

Fig. 11 Influence of the mass proportion of iron on the MAC of air–Fe mixtures (*solid* 1%Fe; *dash* 5%Fe; *dot* 20%Fe)

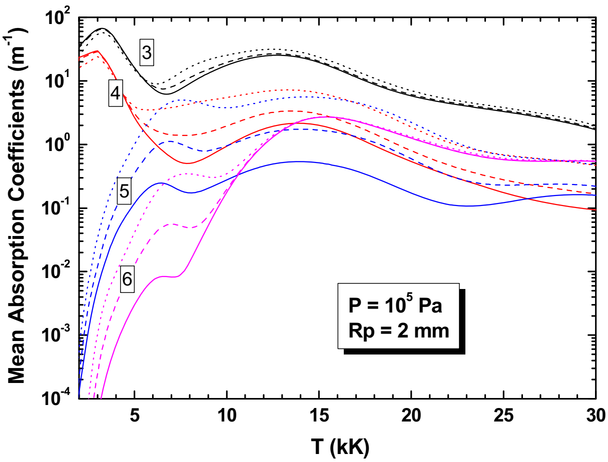

Table 3 Spectral intervals used for calculating MAC represented in Figs. 10 and 11

Interval Number	λ_{min} (nm)	λ_{max} (nm)
1	300	852.2
2	852.2	1019.1
3	1019.1	1129.9
4	1129.9	1946.7
5	1946.7	8565.5
6	8565.5	45,000

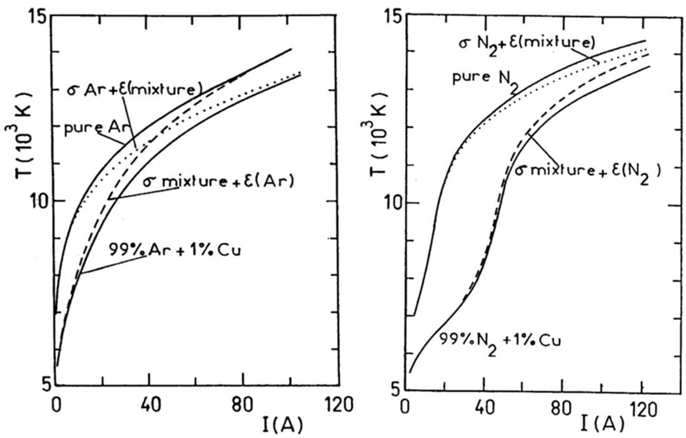

Fig. 12 Theoretical temperature profiles calculated for a wall-stabilized arc (5 mm diameter) with Ar–Cu (*left*) and N_2–Cu (*right*) plasmas. *Solid curves* correspond to temperature profiles computed with the correct values of electrical conductivity and radiation whereas the other curves are fictitious using mixed properties (from [57])

studied in literature (see [56] for example) and the relative importance of these effects in arc devices depends on the current intensity and on the nature of the gas as it has been demonstrated in [57] illustrated by Fig. 12 showing theoretical axis temperature for a wall-stabilized arc. Solid curves correspond to temperature computed with the correct values of electrical conductivity and of the NEC for the various gases or mixtures. Dashed curves were computed using the electrical conductivity of the mixture and the NEC of the pure gas whereas dotted curves were obtained using the radiation of the mixture and the conductivity of the gas. From Ar–Cu curves we can deduce that the effect of the electrical conductivity is important at low current intensity and negligible at high current whereas the contrary is observed for radiation. For N_2–Cu curves the same tendency is observed but the crossing of the fictitious curves is produced at higher current. This is due to the fact that radiation losses in nitrogen plasmas are more important than those of a pure argon plasma, and thus the relative influence of copper vapour on the NEC is lower in nitrogen than in argon.

Hence the presence of metal vapour tends to increase the radiation losses but not as strongly as Figs. 7 and 8 could suggest it because of the effective decrease of temperature. It must be added that some resonance metallic lines are emitted in the UV range but not in the VUV range and hence are not absorbed by the cold surrounding gas such as air, which tends to increase the radiation escaping the arc.

Global Energy Balance

Various authors have calculated or measured the radiation losses in electric arcs and compare them to the global power balance. In fact the comparison should be done with the column power balance, subtracting the electrode voltage fall (rather independent on the metal vapour presence) from the total arc voltage. For current intensity of the order of 100–200 A the radiation losses may represent 50% of the input column energy [9, 58], whereas the same authors have indicated that without metal this part is of the order of 25%. The strongest emission of radiation is created by long and powerful arcs seeded by metallic vapour. Strachan [59] measured radiation power in air–Cu arc plasmas of the order of 50% of the input power. For very long (2 m) and high current (up to 40 kA) air arcs with strong electrode ablation, Cressault et al. [60] measured the total radiation received 10 m from the arc. The main results relative to the radiated power are presented in Fig. 13 as a function of the AC intensity and of the nature of the electrode (copper, steel or aluminum). For copper electrodes the results are consistent with the previous ones of Strachan [59]. With Al and

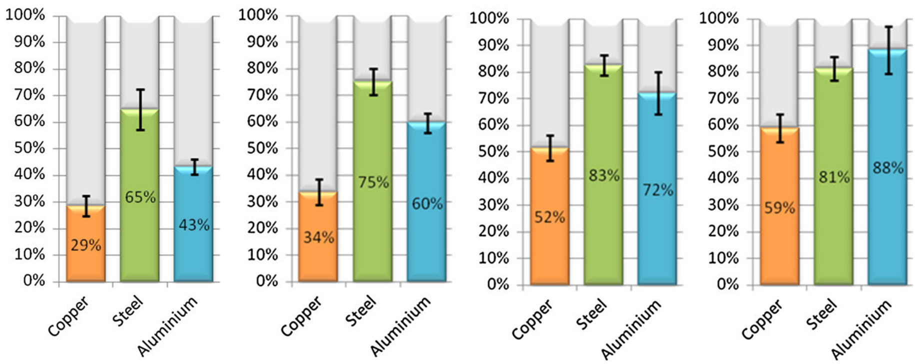

Fig. 13 Proportion of radiative energy relative to the total arc energy. The 4 diagrams correspond respectively (from *left* to *right*) to 4 values of mean rms current intensity: 4, 10, 20 and 40 kA (from [60]) (© IOP Publishing. Reproduced with permission. All rights reserved)

Fe vapors the radiated power may reach 80% of the total input arc energy; this result is quite surprising for Al but the authors indicate that a chemical energy (combustion of Al) is added to the electrical energy.

Heating of Particles in Plasma

In classical plasma spraying metallic or ceramics particulates are injected in a thermal plasma jet and are heated by various mechanisms. Radiation takes part in the global energy balance due to the interaction of the particulate with the plasma. In a lot of studies, this radiation process is not taken into account because it does seem to be an important term of the balance. Furthermore there is some compensation between the radiation from the plasma to the particle and the radiation emitted by the heated particle (grey body). In pioneer works [6, 7] Pfender and co-authors have shown that the role of radiation emitted by the particulate depends on various parameters.

But in certain conditions the presence of a cloud of metal vapour around the particle may increase greatly the radiation from the plasma to the particle. For example using some simplifications on the radiative transfer, Essoltani et al. [61, 62] have shown that for iron particles in an argon plasma, the properties of the particles and of the surrounding plasma jet can be strongly influenced by the radiation of the mixture plasma whereas radiation of pure argon plasma has no influence. Of course this case of Ar–Fe is extreme because the thermal conductivity of this mixture is rather low whereas the radiation is strongly enhanced by the iron vapour.

Conclusion

In this paper we have first presented and explained the well-known effect of metallic vapours in thermal plasmas, consisting in increasing the radiation emission at low and intermediate temperature ranges for a lot of gas–metal mixtures. Let us note that the calculation of radiation properties for a new metal would need a specific work because the NEC depends on the electronic structure of the metal. For a new mixture of gas and metal and when the NEC of the pure species are known, one can use mixture laws to obtain a first estimation of the mixture radiation [63]. We have also described the influence of metal radiation on global properties showing in particular its influence on the total energy balance and comparing its role with that of the electrical conductivity on the plasma cooling.

All the presented results assume that the plasma is in LTE. This hypothesis allowing the use of the Kirchhoff's law is largely used and validated in thermal plasmas studies. Departures from equilibrium may occur in the external regions of the plasmas where the electron number density n_e is lower than a few 10^{21} m^{-3}. The presence of metallic vapour increases the validity of LTE for two reasons:

- This vapour tends to enhance the value of n_e in these regions, because the ionisation potential of the metals is lower than that of the usual gases (see Table 1);
- At very low temperature, metal radiation does not play any role in the plasma because the metal (pure metal or its by-products such as oxides, nitrides, …) condensates.

Finally the NEC is largely used in presence of metals but the real radiative transfer is in general not computed. Is it necessary to treat it using for example mean absorption coefficients and then a classical method such as P1 or Discrete Ordinate methods? In general the spectral region where radiation is strongly absorbed corresponds roughly to the

VUV region ($\lambda < 200$ nm) and the most important part of the neutral metal radiation is emitted and reabsorbed at $\lambda > 200$ nm, even the resonance lines. Hence as a first approximation, a good solution could be a hybrid method coupling the radiative transfer in the VUV region if necessary (for air or SF_6 for example) and the use of a NEC (without the VUV part) in the other spectral ranges. Of course the reality is more complex in particular if the temperature in the central region is higher than 12,000 K, the most emissive metal species being the ions, with resonance lines in the VUV range. But even in this case the use of P1 or DOM method is not recommended because they are enable to correctly treat the real line radiative transfer. If such treatment is needed, in particular to interpret some experimental spectra showing a very marked absorption of resonance lines, with non-homogeneous distribution of metal vapour (see for example [64]) a special calculation should be developed.

References

1. Boulos M, Fauchais P, Pfender E (1994) Thermal plasmas—fundamentals and applications. Plenum Press, New York
2. Cressault Y, Gleizes A (2013) Thermal plasma properties for Ar–Al, Ar–Fe and Ar–Cu mixtures used in welding plasmas processes. Part I: data tables of net emission coefficients. J Phys D Appl Phys 46:415206
3. Cressault Y, Gleizes A, Riquel G (2012) Properties of air–aluminium thermal plasmas. J Phys D Appl Phys 45:265202
4. Eckert ERG, Pfender E (1967) Advances in plasmas heat transfer. Adv Heat Transf 4:229–316
5. Pfender E (1976) Heat transfer from thermal plasmas to neighboring walls or electrodes. Pure Appl Chem 48:199–213
6. Chen X, Pfender E (1982) Unsteady heating and radiation effect of small particles in a thermal plasma. Plasma Chem Plasma Proc 2:293
7. Chen X, Chyou YP, Lee YC, Pfender E (1985) Heat transfer to a particle under plasma conditions with vapor contamination from the particle. Plasma Chem Plasma Proc 5:119
8. Menart J, Heberlein J, Pfender E (1996) E line-by-line method of calculating emission coefficients for thermal plasmas consisting of monoatomic species. J Quant Spectrosc Radiat Transf 56:377–398
9. Menart J, Heberlein J, Pfender E (1999) Theoretical radiative transport results for a free-burning arc using a line-by-line technique. J Phys D Appl Phys 3:55–63
10. Modest MF (1993) Radiative heat transfer. Mc-Graw-Hill Int. Ed., New York. ISBN 0-07-112742-9
11. Siegel R, Howell J (2002) Thermal radiation heat transfer, 4th edn. Taylor and Francis, London. ISBN 1-56032-839-8
12. Cabannes F, Chapelle J (1971) Spectroscopic Plasma Diagnostic. In: Venugopalan M (ed) Reactions under plasma conditions (Chap. 7), vol 1. Wiley-Interscience, New York
13. Gleizes A, Gongassian M, Rahmani B (1989) Continuum absorption coefficient in SF_6 and SF_6–N_2 mixtures plasmas. J Phys D Appl Phys 22:83–89
14. Biberman LM, Norman GE (1960) Continuous spectra of atomic gases and plasma. Opt Spectrosc 8:230
15. Hofsaess D (1977) Photoabsorption in alkali and alkaline earth elements calculated by the scaled Thomas Fermi method. Z Phys A 281:1–13
16. Hofsaess D (1979) Photoionization cross sections calculated by the scaled Thomas-Fermi method (hv $\leq$ 50 eV). Atomic Data Nuclear Data Tables 24:285–321
17. TOPbase data table. http://cdsweb.u-strasbg.fr/topbase/xsections.html
18. Cressault Y, Hannachi R, Teulet P, Gleizes A, Gonnet J-P, Battandier J-Y (2008) Influence of metallic vapours on the properties of air thermal plasmas. Plasma Sour Sci Technol 17:035016
19. Naghizadeh-Kashani Y, Cressault Y, Gleizes A (2002) Net emission coefficient of air thermal plasmas. J Phys D Appl Phys 35:2925–2934
20. Chauveau S, Deron C, Perrin M-Y, Riviere P, Soufiani A (2003) Radiative transfer in LTE air plasmas for temperatures up to 15,000 K. J Quant Spectrosc Radiat Transf 77:113–130
21. Jan C, Cressault Y, Gleizes A, Bousoltane K (2014) Calculation of radiative properties of SF_6–C_2F_4 thermal plasmas-application to radiative transfer in high-voltage circuit breakers modelling. J Phys D Appl Phys 47:015204

22. Ralchenko Y, Kramida AE, Reader J and NIST ASD team (2008), NIST Atomic spectra database (version 3.1.5). National Institute of Standards and Technology, Gaithersburg, MD. http://physics.nist.gov/asd3. Accessed May 2016
23. Kurucz RL, Bell (1995) Atomic line data. Kurucz CD-ROM No. 23. Smithsonian Astrophysical Observatory, Cambridge, MA
24. Cressault Y, Rouffet M-E, Gleizes A, Meillot E (2010) Net emission coefficient of Ar–H_2–He thermal plasmas at atmospheric pressure. J Phys D Appl Phys 43:335204
25. Herzberg G (1989) Molecular spectra and molecular structure: I. Spectra of diatomic molecules. Krieger, ISBN 0-89464-268-5; II. Infrared and Raman spectra of polyatomic molecules. Krieger, ISBN 0-89464-269-3; III. Electronic spectra and electronic structure of polyatomic molecules. Krieger, ISBN 0-89464-270-7
26. Luque J, Crosley DR (1999) LIFBASE database and spectral simulation program (version 1.5). Presented at the SRI international report MP 99-009
27. Laux C, Spence TG, Kruger CH, Zare RN (2003) Optical diagnostics of atmospheric pressure air plasmas. Plasma Sour Sci Technol 12(2):25. http://www.specair-radiation.net/. Accessed June 2016
28. Aubrecht V, Bartlova M (2009) Net emission coefficients of radiation in air and SF_6 thermal plasmas. Plasma Chem Plasma Proc 29:131–147. doi:10.1007/s11090-008-9163-x
29. Kloc P, Aubrecht V, Coufal O, Bartlova M (2014) Influence of copper vapour on the radiative transfer in thermal air plasma. In: International conference on gas discharges applications, GD2014, Orleans, France
30. Aubrecht V, Bartlova M, Coufal O (2010) Radiative emission from air thermal plasmas with vapour of Cu or W. J Phys D Appl Phys 43:434007. doi:10.1088/0022-3727/43/43/434007
31. Lowke JJ (1974) Predictions of arc temperature profiles using approximate emission coefficients for radiation losses. J Quant Spectrosc Radiat Transf 14:111–122
32. Randrianandraina HZ, Cressault Y, Gleizes A (2011) Improvements of radiative transfer for SF_6 thermal plasmas. J Phys D Appl Phys 44:194012
33. Essoltani A, Proulx P, Boulos MI, Gleizes A (1990) Radiation and self-absorption in argon-iron plasmas at atmospheric pressure. JAAS 5:543–547
34. Gleizes A, Gonzalez J-J, Liani B, Raynal G (1993) Calculation of net emission coefficient of thermal plasmas in mixtures of gas with metallic vapour. J Phys D Appl Phys 26:1921–1927
35. Essoltani A, Proulx P, Boulos MI, Gleizes A (1994) Volumetric emission of argon plasma in the presence of vapors of Fe, Si and Al. Plasma Chem Plasma Proc 14:437–450
36. Essoltani A, Proulx P, Boulos MI, Gleizes A (1994) Effects of the presence of iron vapors of Ar/Fe, Ar/Fe/H2 plasmas. Plasma Chem Plasma Proc 14:301–315
37. Raynal G, Vergne P-J, Gleizes A (1995) Radiative transfer in SF_6 and SF_6–Cu arcs. J Phys D Appl Phys 28:508–515
38. Cressault Y, Gleizes A (2001) Radiative and transport properties in Ar–H_2–Cu mixtures at atmospheric pressure. Progress in plasma processing of materials. Begell House, New York, pp 433–438
39. Paul K, Takemura T, Matsuno H, Hiramoto T, Dawson F, Gonzalez JJ, Gleizes A, Zissis G, Erraki A, Lavers J (2004) Predicted results of a HID DC current lamp considering a P − 1 radiation model. IEEE Trans Plasma Sci 32:118–126
40. Hannachi R, Cressault Y, Teulet P, Ben Lakhdar Z, Gleizes A (2008) Net emission of H_2O–air–$MgCl_2$/$CaCl_2$/NaCl thermal plasmas. J Phys D Appl Phys 41:205212
41. Teulet P, Gonzalez J-J, Mercado-Cabrera A, Cressault Y, Gleizes A (2009) 1-D hydro-kinetic modelling of the decaying arc in air–PA66–copper mixtures. Part I. Chemical kinetics, thermodynamics, transport and radiative properties. J Phys D Appl Phys 42N:17-175201
42. Billoux T, Cressault Y, Boretskij VF, Veklich AN, Gleizes A (2012) Net emission coefficient of CO_2–Cu thermal plasmas: role of copper and molecules. J Phys: Conf Ser 406:012027
43. Cressault Y, Teulet P, Zissis G (2016) Radiative properties of ceramic metal-halide high intensity discharge lamps containing additives in argon plasma. Jpn J Appl Phys 55(72):705
44. Menart J, Heberlein J, Pfender E (1995) Theoretical radiative emission results for argon/copper thermal plasmas. Plasma Chem Plasma Proc 16:245–265
45. Moscicki T, Hoffman J, Szymanski Z (2004) Emission coefficients of low temperature thermal iron plasma. Czech J Phys 54:C677–C682
46. Moscicki T, Hoffman J, Szymanski Z (2008) Net emission coefficients of low temperature thermal iron–helium plasma. Opt Appl 38:365–373
47. Menart J, Malik S (2002) Net emission coefficients for argon–iron thermal plasmas. J Phys D Appl Phys 35:867
48. Wendt M (2011) Net emission coefficients of argon iron plasmas with electron Stark widths scaled to experiments. J Phys D Appl Phys 44:125201

49. Adineh VR, Coufal O, Zivny O (2012) Thermodynamic and radiative properties of plasma excited in EDM process through N_2 taking into account Fe. IEEE Trans Plasma Sci 40:2723–2735. http://ieeexplore.ieee.org/document/6279489/?reload=true&arnumber=6279489
50. Adineh VR, Coufal O, Bartlova M (2015) Calculation of net emission coefficient of electrical discharge machining arc plasmas in mixtures of nitrogen with graphite, copper and tungsten. J Phys D Appl Phys 48(40):405202. doi:10.1088/0022-3727/48/40/405202
51. Nordborg H, Iordanidis A (2008) Self-consistent radiation based modelling of electric arcs: I. Efficient radiation approximations. J Phys D Appl Phys 41:135205. doi:10.1088/0022-3727/41/13/135205
52. Kloc P, Aubrecht V, Bartlova M, Coufal O, Ru C (2015) On the selection of integration intervals for the calculation of mean absorption coefficients. Plasma Chem Plasma Proc 35:1097–1110. doi:10.1007/s11090-015-9648-3
53. Bartlova M, Bogatyreva N, Aubrecht V (2014) Radiation heat transfer in thermal argon plasma with iron vapors. Plasma Phys Technol 1(1):8–10
54. Kloc P, Aubrecht V, Coufal O (2015) Radiative transfer in air and air–Cu plasmas for two temperature profiles. J Phys D Appl Phys 48:5. doi:10.1088/0022-3727/48/5/055208
55. Hannachi R, Cressault Y, Salem D, Teulet P, Bejo L, Ben Lakhdar Z (2012) Mean absorption coefficient of H_2O–air–$MgCl_2$/$CaCl_2$/NaCl thermal plasmas. J Phys D Appl Phys 45:485206
56. Murphy AB (2010) The effects of metal vapor in arc welding. J Phys D Appl Phys 43:434001
57. Gleizes A, Gonzalez JJ, Liani B (1992) Net emission coefficient of mixtures of gas with metallic vapour and its influence on arc modelling. In: 10th international conference on gas discharges and applications, GD1992, Swansea, vol 1, pp 208–211
58. Gonzalez J-J, Gleizes A, Proulx P, Boulos M (1993) Mathematical modeling of a free-burning arc in the presence of metal vapor. J Appl Phys 74:3065–3070
59. Strachan DC (1973) Radiation losses from high-current free-burning arcs between copper electrodes. J Phys D Appl Phys 6:1712
60. Cressault Y, Bauchire J-M, Hong D, Rabat H, Riquel G, Gleizes A (2015) Radiation of long and high power arcs. J Phys D Appl Phys 48:415201
61. Essoltani A, Proulx P, Boulos MI, Gleizes A (1991) Radiative effects on plasma-particle heat transfer in the presence of metallic vapors. In: 10th international symposium on plasma chemistry, ISPC-10, Bochum, vol 1, pp 1–7
62. Essoltani A, Proulx P, Boulos MI, Gleizes A (1993) A combined convective, conductive and radiative heat transfer study for an evaporating metallic particle under plasma conditions. J High Temp Chem Proc 2:37–46
63. Gleizes A, Cressault Y, Teulet P (2010) Mixing rules for thermal plasma properties in mixtures of argon, air and metallic vapours. Plasma Sour Sci Technol 19:055013
64. Hong D, Sandolache G, Bauchire JM, Gentils F, Fleurier C (2005) A new optical technique for investigations of low-voltage circuit breakers. IEEE Trans Plasma Sci 33:976–981
65. Salem D, Hannachi R, Cressault Y, Teulet P, Béji L (2015) Radiative properties of argon–helium–nitrogen–carbon–cobalt–nickel plasmas used in CNT synthesis. J Phys D Appl Phys 48:065202