

STATE OF ART AND CHALLENGES FOR THE CALCULATION OF RADIATIVE AND TRANSPORT PROPERTIES OF THERMAL PLASMAS IN HVCB

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Abstract. This paper is focused on the state-of-the-art and challenges concerning the thermophysical properties of thermal plasmas used in numerical modelling devoted to high voltage circuit breakers. For Local Thermodynamic Equilibrium (LTE) and Non-Local Thermodynamic (NLTE) and/or Chemical Equilibrium (NLCE) plasmas, the methods used to calculate the composition, thermodynamic, transport and radiative properties are presented. A review of these last data is proposed and some comparisons are given for illustrations.

Keywords: HVCB, chemical composition, transport coefficients, radiative properties, NLTE.

1. Introduction

Due to the cost of the experimental tests for HVCB, more and more numerical modelling is developed in order to compare the interruption capability of gases or mixtures with lower ODP than SF₆ (CF₃I, CO₂, C₄F₇N with Air, N₂ or O₂), to optimize the efficiency of the process by using new designs, new electrodes or new walls, or simply to better understand the physical and chemical processes in the plasma.

To develop the simulation of a high voltage circuit breaker using magneto-hydro-dynamic models (MHD), it is necessary to elaborate databanks of fundamental properties: the thermodynamic properties, the transport coefficients, and the radiative properties, these data being used in the different equations of conservation: mass conservation, momentum conservation and energy conservation. Depending on the region of the plasma (near the electrodes, near the walls, in the surrounding gases, in the core of the plasma), depending on the pressure and the temperature, depending on the intensity or power (ignition of the arc or its cooling and extinction), presence of turbulence or not, the Local Thermodynamic Equilibrium (LTE) hypothesis can be assumed or not when non-equilibrium effects appears. Consequently, the numerical simulations have to be performed considering nonequilibrium phenomena, which are done using the two-temperature assumption. In this we usually distinguish the electron kinetic temperature T_e which characterizes the high temperatures area of the plasma, while the heavy particles kinetic temperature T_h describes low temperature regions.

This paper tries to make a state of the art and to present the challenges on the calculation of the properties of plasmas used in HVCB, in the case of LTE

and non-LTE assumptions. The first part is devoted to properties of plasmas in LTE conditions with a summarize of the gases studied as alternative gases or mixtures to SF₆ and of the methods used to estimate the composition, the thermodynamic, transport and radiative properties. As the expressions available in the literature are accepted by the community in the case of LTE assumption, we will remember the recent works done on new mixtures and radiative properties. The second part is focused on the 2T-properties of plasmas in non-local thermodynamic equilibrium (nLTE) and/or non-local chemical equilibrium (nLCE). Few works have been done on this topic and lot of questions remain. We make a brief overview of the recent papers dealing with the 2T-properties applied to HVCB and finish the paper by indicating some challenges to solve.

2. Plasma in LTE conditions

For the HVCB applications, the first studies were devoted to the description of the heat zone, corresponding to the ignition of the arc in the first instants after the opening of the contacts. The plasma is assumed to be in Local Thermodynamic Equilibrium, the thermal properties are calculated with or without metallic vapors coming from the erosion of the electrodes (Cu, W, CuW), with or without thermoplastic vapors coming from the erosion of the nozzle (C₂F₄). The LTE properties have been calculated for lots of pure gases or mixtures (SF₆, N₂, Air, CO₂, CF₃I [1, 2], CF₄, C₃F₈, C₄F₈, c-C₄F₈, C₄F₇N, C₄F₈N₂, C₅F₁₀, C₆F₁₂O, CF₃NSF₂, SF₆-N₂, SF₆-CO₂, SF₆-CF₄, C₂F₄-CO₂, C₄F₈-CO₂ [3–5], CO₂-N₂-Cu [6], SF₆-Cu [7–9], C₅F₁₀O-CO₂-O₂ [10]). The plasma composition is the first and unavoidable step allowing

the computation of these properties, and is obtained from the law of mass action and the chemical base concept defined by Godin and Trépanier [11]. This technique is derived from the laws of thermodynamics and condensed phases are sometimes considered in the calculation of the population densities. Virial [12] and Debye [13] pressure corrections are often considered at high pressure ($P > 10$ bars) where the perfect gas law has to be replaced by a real gas law.

The thermodynamic properties (mass density, enthalpy, and specific heat at constant pressure) are directly deduced from the plasma compositions. Some results are given in [2–9] and behaviors are explained versus temperature, mixtures and pressure. The transport coefficients (viscosity, thermal and electrical conductivities) are obtained according to the Chapman-Enskog method and a previous calculation of collision integrals depending on the interaction potentials chosen to characterize the different collisions between particles (neutral-neutral, ion-neutral, electron-neutral and charged-charged collisions). The study of the collision integrals constitutes the most important part of the calculation of the transport coefficients and are often responsible for the differences observed between the authors [2–13].

The radiative losses play an important role in the HVCB arc plasmas: this is the main term of energy losses in the central region of the arc, the absorption in the surrounding plasma and cold gas leads to an increase of the pressure, it is responsible of the PTFE ablation influencing the behavior and the extinction of the plasma. The divergence of the flux is obtained by solving the radiative transfer equation which depends on the wavelengths, directions, and the geometry of the plasma. Due to the complexity of this equation, several simplifying approaches have been developed during the last decades such as the method of the net emission coefficient (NEC) initially proposed by Lowke and first calculated for SF_6 by Liebermann and Lowke [14, 15], then by Aubrecht and Barlova or Gleizes et al. [16, 17]. In order to treat efficiently the radiative transfer in the middle or cold regions, the P1-Model or discrete ordinates methods are often preferred using grey body approximation or Mean Absorption Coefficients (MACs) [18–22]. Then, due to rise of the computer's capacity, some works have been developed on pure SF_6 in order to perform an exact calculation of the radiative transfer equation with a very fine description of the spectrum, evaluating the accuracy and the validity of the simplified methods by comparisons. A quick review has been presented during the FSO conference in 2011 by A. Gleizes et al. [23]. Since, more studies on the radiative transfer equation have been done for pure gases (SF_6 , N_2 , Air, CO_2), binary mixtures (SF_6 - C_2F_4 , C_4F_8 - CO_2 , CO_2 -Cu [3, 24]) and ternary mixtures (SF_6 - C_2F_4 -Cu/W) in 1D-2D-3D geometry with imposed temperatures profiles [25–28]. An example is given in Figure 1 for the divergence of the flux for a SF_6 pure plasma at

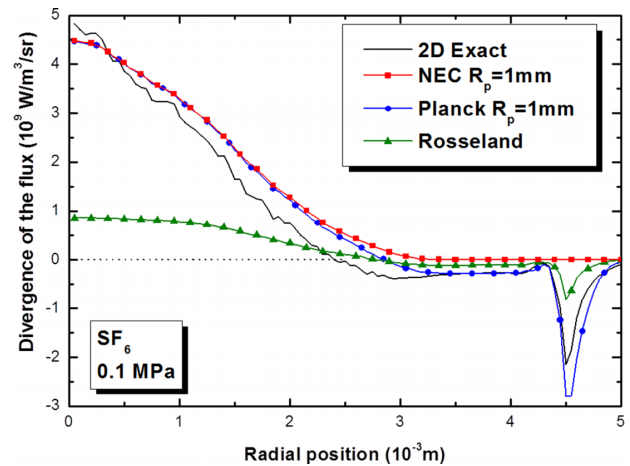


Figure 1. Divergence of the flux for a SF_6 pure plasma at 0.1 MPa, obtained with different approached methods [1].

1bar, obtained with different approached methods [1].

From these studies, some conclusions can be done: (1) for the equilibrium composition and thermodynamic functions, the methods are well-known and valid, maybe questionable for plasmas at very high pressure and low temperature; (2) for transport coefficients, the interaction potentials have a significant influence on the values obtained and validation is done through other parameters such as the pressure for the thermal conductivity. A very small concentration of metallic vapors ($>1\%$) strongly influences the electrical conductivity whereas viscosity and thermal conductivity are modified with concentration of metals or PTFE higher than 20–25%. Consequently, it is important to consider those coefficients in the conservation equations if we want to study the plasma regions where the ablation/erosion of walls and electrodes is important; (3) for the radiative transfer, some differences can be observed on the NEC when estimated using the escape factor approximation instead of the line by line method (more than 40% of difference can be observed at very high pressure > 100 bar [25]).

In LTE conditions, the challenges on thermal properties are mainly “driven” by the use of new alternative gases to SF_6 (CO_2 , CF_3I , $c\text{-C}_4\text{F}_8$, $\text{C}_4\text{F}_7\text{N}$, $\text{C}_5\text{F}_{10}\text{O}$, $\text{C}_6\text{F}_{12}\text{O}$, CF_3NSF_2 , pure gases or mixed with CO_2 , CF_4 , C_2F_4 , O_2 , N_2 , air) or by the use of new materials for the manufacturing of electrodes or nozzles (Cu, W, CuW which have an important impact on the electrical conductivity and the radiation even if in few concentration). Some numerical modelling have been developed to study the influence of the erosion of electrodes or ablation of nozzle on the arc behavior from its ignition to its extinction [29–34]. The radiative transfer calculation with simplified methods can be improved following four ways: a better treatment of the absorption coefficients for molecules [26]; a careful definition of the spectral intervals for the calculation

of the Mean Absorption Coefficients as in their works of Jan et al., Norborg and Iordanidis or Kloc et al. [26, 27, 35]; the elaboration of MACs tables from a mix of the different mean functions (Planck averaging at high temperature considering the absorption of the lines, and mean natural value at low temperature with only continuum contribution) [26]; the use of an optimized function based on the minimization of the difference between the exact calculation of the radiative flux or the divergence of the flux and the results obtained with simplified methods [36–38].

3. Plasma in nLTE and/or nLCE conditions

In many plasma processes, departures from thermal or chemical equilibrium exist on the edges of the plasma and close to the walls and the electrodes. For example, in the case of breaking devices and during the current zero phase, the electron number density is not very significant, the energy transfer between the electrons and the heavy particles is not enough efficient to preserve thermal equilibrium. As a consequence, the electron kinetic temperature T_e is higher than the heavy particles temperature T_h , and two-temperature models must be developed. The recent papers propose two equations of energy conservation: one for the electrons and the other for the heavy species [39]. These models require the computation of thermodynamic, transport and radiative properties as a function of T_e and $\theta = T_e/T_h$ and the first step to obtain these data is, as in the case of LTE, the calculation of the 2T plasma composition.

There are several available theories in the literature to calculate the densities of species for non-equilibrium plasmas: (i) the minimization of a thermodynamic function (Gibbs' free enthalpy for plasma at constant pressure or Helmholtz's free energy at constant volume), (ii) the method based upon the 2T mass action law (LMA) assuming the chemical equilibrium and coupled with a set of equations for the conservation of pressure, electrical charge and mixture proportions and (iii) collisional radiative models (CR) or a chemical kinetic model (CK) coupled with an equation for the conservation of pressure. These methods give the same results in LTE but present strong differences if the plasma is not in equilibrium, depending on the temperatures affected the processes: the translational kinetic temperatures of electrons and heavy particles T_e and T_h , the temperatures characteristic of the population of the various internal energy modes i.e. excitation temperature T_{ex} associated to the population of excited electronic states (atomic and molecular), the vibration temperature T_v linked to the distribution of vibrational levels and the rotation temperature T_r related to the population of rotational states. The LMA method is relatively easy to implement in numerical codes, and requires few data (partition functions, enthalpies of formation at 0 K of each chemical species and mass of species). The collisional-radiative (CR)

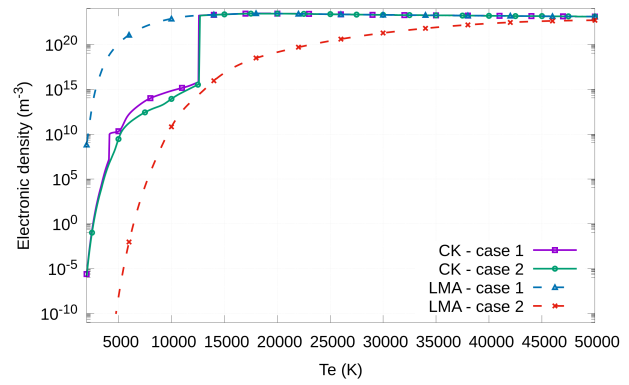


Figure 2. Electron number densities versus electronic temperature T_e (CK: chemical kinetic model, LMA: law of mass action). Pure SF_6 , $\theta = 5$, $P = 1$ bar.

and chemical kinetic (CK) models have advantages to reduce simplifying assumptions in the calculation. Moreover, the CK model allows studying the temporal chemical dynamic of the system. Nevertheless, a large databank of reaction rates or cross sections for each chemical process are needed. Relations between cross sections and reaction rates are well known for plasmas in thermal equilibrium conditions but are more complex for non-LTE plasmas. Indeed, electrons and heavy particles involved in chemical reactions have not the same temperature (T_e and T_h) and are not described statistically by the same distribution functions. The temporal evolution of densities for each chemical species is given by a set of master equations:

$$\frac{\partial n_i}{\partial t} = C_i - n_i D_i \quad (1)$$

where C_i and D_i are respectively the creation and destruction factors for the specie i . They depend on the densities of species participating at the creation/destruction of specie i through the chemical reaction and on the reaction rates of chemical reactions involved. Using relations between forward and reverse reaction rates, we used the library DVODE to solve the system formed (Eq. 1). The recent works of the team compared these methods in the case of pure SF_6 [40–42] and mixture $SF_6-C_2F_4$. We used two different assumptions concerning the population of internal energy modes: case (1) with $T_{ex} = T_e$, $T_v = T_e$ and $T_r = T_h$ more adapted to ionized gases (high temperatures), and case (2) $T_{ex} = T_h$, $T_v = T_h$ and $T_r = T_h$ more adapted to molecular gases (low temperatures). The figure 2 compares the electron number densities versus the electronic temperature T_e ($\theta = T_e/T_h = 5$), for pure SF_6 at 1 bar, and obtained from the CK and LMA methods.

The thermodynamic properties of 2T plasma are deduced from relations given Colombo et al. [43, 44] which propose to calculate a unique 2T specific heat C_p as the derivative of the total enthalpy $H_{tot} = H_e + H_h$ compared to the electronic temperature T_e :

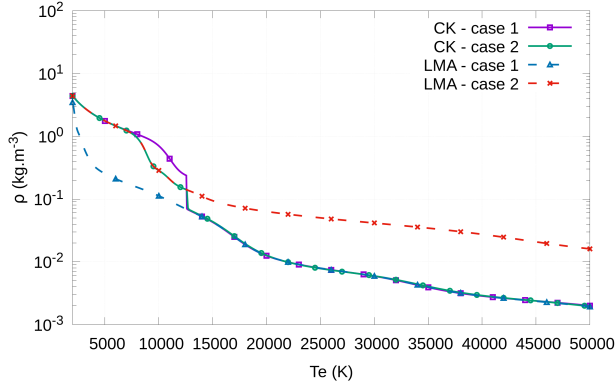


Figure 3. Mass density ρ versus electronic temperature T_e for pure SF_6 plasma, for $\theta = 5$ and pressure of 1 bar.

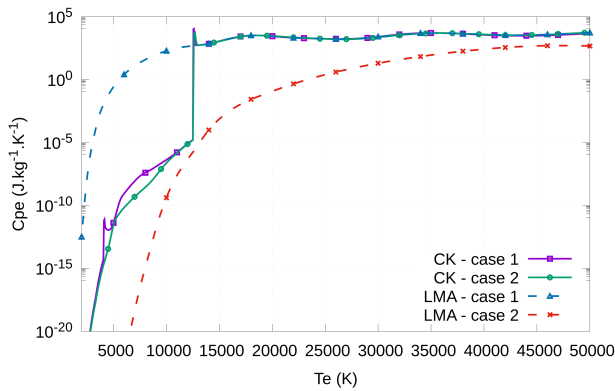


Figure 4. Specific heat at constant pressure of electrons C_{Pe} versus electronic temperature T_e for pure SF_6 , for $\theta = 5$ and pressure of 1 bar.

$$C_P(\theta, T_e) = \left(\frac{\partial H_{\text{tot}}}{\partial T_e} \right)_P \quad (2)$$

As energetic contributions of electrons and heavies are separated in numerical codes through two coupled energy conservation equations, it is necessary to calculate independently the enthalpy of electron H_e and heavy particles H_h , the specific heats of electrons C_{Pe} and heavy species C_{Ph} :

$$C_{Pe}(\theta, T_e) = \left(\frac{\partial H_e}{\partial T_e} \right)_P \quad (3)$$

$$C_{Ph}(\theta, T_e) = \left(\frac{\partial H_h}{\partial T_h} \right)_P \quad (4)$$

This separation of the specific heat in two distinct contributions for electrons and heavy species is also the approach retained by Ghorui et al. [45, 46] or Zhang et al. [47]. The figure 3 shows the mass density of pure SF_6 obtained from the LMA and CK for the two cases, $\theta = 5$ and 1 bar. The figures 4 and 5 present the two specific heats in the same conditions. The law of mass action is in dashed lines and the chemical kinetic method in full lines.

With the CK model, there is a transition between both cases of the LMA. We can conclude that the

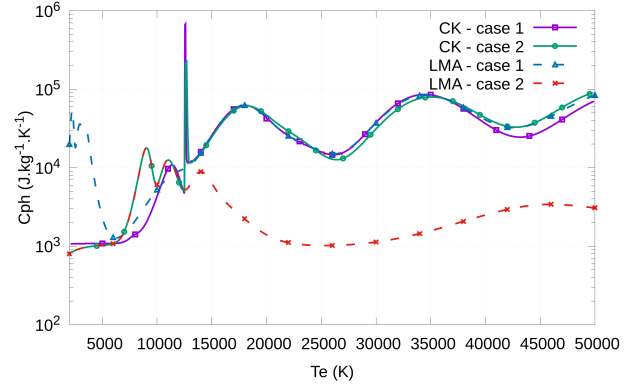


Figure 5. Specific heat at constant pressure of heavy particles C_{Ph} versus electronic temperature T_e for pure SF_6 , for $\theta = 5$ and pressure of 1 bar.

LMA taken in the case 1 is accurate for the high temperatures and in the case 2 for the low temperature range. In high temperatures, there is lot of electrons and the inelastic processes are dominated by electron collisions and have to be calculated at T_e (case 1). In low temperatures, the heavy particles chemical processes are dominant and have to be calculated at T_h (case 2). There is a range of temperature where the LMA is not exact and the CK is necessary to calculate the composition.

Whereas the community agrees the calculation of the thermodynamic properties, translation thermal conductivities of electrons and heavy particles, viscosity and electrical conductivity, there is no consensus for the calculation of the reactive thermal conductivity. The theory of Hirschfelder et al. [12] or Bonnefoi et al. [48, 49] consider separately the electrons and the heavy particles whereas the theory of Rat showed that these two theories did not consider correctly not only the diffusion in 2T plasmas but also the coupling between electrons and heavy particles. The Rat's theory leads to new definitions of collision integrals, transport coefficients and consequently new terms in energy fluxes. Ghorui et al. [45, 46] in 2008 and Colombo et al. in 2009 [43, 44] showed in their works that the method of Devoto is sufficient to determine the transport properties (electrical conductivity, thermal conductivity and viscosity) in 2T assumption only if diffusion coefficients defined by Ramshaw were used. From the last works published on 2T-properties for several gases applied in various applications (Bonnefoi et al. [48, 49] for Ar-H, Mexmain [50] for Ar-O, McCourt [51] for polyatomic mixtures, Aubreton et al. [52–54] for Ar-O2, Ar-He or pure N2, Capitelli et al. [55, 56] with a "State-to-State" model applied for pure N2 and H2 plasmas, Guo et al. [57], Zhang et al. [58] and Yang et al. [59] on helium, argon and SF_6 -N2 mixtures) we can consider as accepted that the translation and internal thermal conductivities of heavy particles and the viscosity can be calculated at T_h , the translation thermal conductivity of electrons and the electrical conductivity at T_e . The figure 6 compare

these coefficients obtained for the cases 1 and 2, the LMA and CR/CK methods, in the case of pure SF₆ at 1 bar and $\theta = 5$. The computation of the 2T reactive thermal conductivity at high pressure for complex gases such as SF₆ using multi-temperature expressions from the literature (such as the one provided by Ghorui et al. [45, 46]) gave poor results. The use of the Ghorui's expressions seems to be accurate for pressure lower than 16 bar and pure gases (O₂, N₂) or mixtures with few chemical reactions (O₂-N₂). For pure SF₆ and SF₆-C₂F₄ mixtures, we extended the Butler and Brokaw theory to 2T assumption. We divided the total reactive thermal conductivity into two contributions, one for the heavy particles (considering neutral atoms and molecules) and one for the electron part (considering electrons and ions):

$$\kappa_{\text{reac}}^{\text{h}} = -\frac{1}{RT_{\text{h}}^2} \frac{\begin{vmatrix} 0 & \Delta h_1 & \cdots & \Delta h_{N_e} \\ \Delta h_1 & A_{11} & \cdots & A_{N_e 1} \\ \vdots & \vdots & \ddots & \vdots \\ \Delta h_{N_e} & A_{1N_e} & \cdots & A_{N_e N_e} \end{vmatrix}}{\begin{vmatrix} A_{11} & \cdots & A_{N_e 1} \\ \vdots & \ddots & \vdots \\ A_{1N_e} & \cdots & A_{N_e N_e} \end{vmatrix}} \quad (5)$$

$$\kappa_{\text{reac}}^{\text{e}} = -\frac{1}{RT_{\text{e}}^2} \frac{\begin{vmatrix} 0 & \Delta h_1 & \cdots & \Delta h_{N_e} \\ \Delta h_1 & A_{11} & \cdots & A_{N_e 1} \\ \vdots & \vdots & \ddots & \vdots \\ \Delta h_{N_e} & A_{1N_e} & \cdots & A_{N_e N_e} \end{vmatrix}}{\begin{vmatrix} A_{11} & \cdots & A_{N_e 1} \\ \vdots & \ddots & \vdots \\ A_{1N_e} & \cdots & A_{N_e N_e} \end{vmatrix}} \quad (6)$$

with

$$A_{ij} = \frac{RT_{ij}}{P} \sum_{k=2}^{N_e-1} \sum_{l=k+1}^{N_e} \frac{x_k x_l}{D_{kl}^b} \left(\frac{\alpha(i, k)}{x_k} - \frac{\alpha(i, l)}{x_l} \right) \times \left(\frac{\alpha(j, k)}{x_k} - \frac{\alpha(j, l)}{x_l} \right) \quad (7)$$

T_{ij} is equivalent to T_e if one of the two particles in collision is an electron, T_h in the other cases. From the original expression of Butler and Brokaw, the temperature T was replaced by T_e , T_h or T_{ij} in relations (4). This choice has been validated by comparisons of shape of the reaction thermal conductivities versus temperature, θ and pressure. As examples, the results of the electrical conductivity, viscosity, translational, reactive and total thermal conductivities of electron and heavy particles are presented in figure 6 for 50%SF₆-50%C₂F₄ at 1 bar and $\theta = 5$, obtained from CK and LMA methods. For the case 1, the reaction term for heavy particles increases with θ , the contribution of electrons decreases. For the case 2, the values of $\kappa_{\text{reac}}^{\text{h}}$ are shifted toward high temperatures and the peaks of $\kappa_{\text{reac}}^{\text{e}}$ are diminished and shifted to higher temperatures [60].

Finally, the last part concerns the calculation of the 2T radiative properties. For radiation in nLTE, some works have been done on atmospheric reentry by Soucasse et al. [61], or Scoggings [62] but few are available in the literature for plasmas used in HVCB. Contrarily to studies in LTE, the specialita emission and absorption coefficients cannot be linked in the Kirshhoff law through the Planck function. Recent papers propose to write two equations of conservation of energy: one for the electron with divergence of the flux obtained with the net emission coefficient approximation considering only the continuum part, another one for the heavy species with divergence of the flux obtained with the same approximation considering only the contribution of the lines. Consequently, the radiation has to be separated into two parts: one coming from atomic (radiative recombination, radiative attachment, Bremsstrahlung) and molecular continua and calculated at the temperature T_e , and the other one coming from the atomic lines and calculated at T_{ex} (consequently T_e or T_h depending on the case 1 or 2), the molecular lines being neglected at this step. The broadenings have been calculated assuming T_h for the FWHM due to the Doppler effect, T_h for the van der Waals effect, and T_e for the Stark perturbation. The correction proposed by Griem has been taken into consideration by calculating the parameter R with T_h . Then, two net emission coefficients can be estimated:

$$\varepsilon_N^{\text{cont}} = \int_0^{+\infty} \varepsilon_{\lambda}^{\text{cont}}(\lambda, T_e) e^{-R_p \kappa'_{\text{tot}}(\lambda, T_e)} d\lambda \quad (8)$$

$$\varepsilon_N^{\text{lines}} = \int_0^{+\infty} \varepsilon_{\lambda}^{\text{lines}}(\lambda, T_{\text{ex}}) e^{-R_p \kappa'_{\text{tot}}(\lambda, T_{\text{ex}})} d\lambda \quad (9)$$

Figure 7 shows the influence of θ on the NECs for the two cases. It means in our case that NEC for continuum is always dependent on T_e but NEC for atomic lines depend on T_e (case 1) or T_h (case 2). As the NEC for the continuum only depends on T_e , the factor θ has no influence. This is not the same conclusion for the NEC of the atomic lines since T_{ex} is equal to T_e for the case 1 ($T_{\text{ex}} = T_e$, $T_v = T_e$ and $T_r = T_h$) or T_h for the case 2 ($T_{\text{ex}} = T_h$, $T_v = T_h$ and $T_r = T_h$).

4. Conclusion

We have presented the methods commonly used to determine the thermophysical properties (composition, thermodynamic, transport and radiative properties) for LTE and NLTE/NLCE plasmas existing in HVCB. For LTE plasmas, the methods are well-known, as the influence of the metallic vapors on the properties: a small proportion of metallic vapors (inferior to 1%) strongly impacts the electrical conductivity and the radiation, higher proportions is required (more than 20%) to modify significantly the thermal conductivity and the viscosity. The challenges will be not only the calculation of these properties for new materials

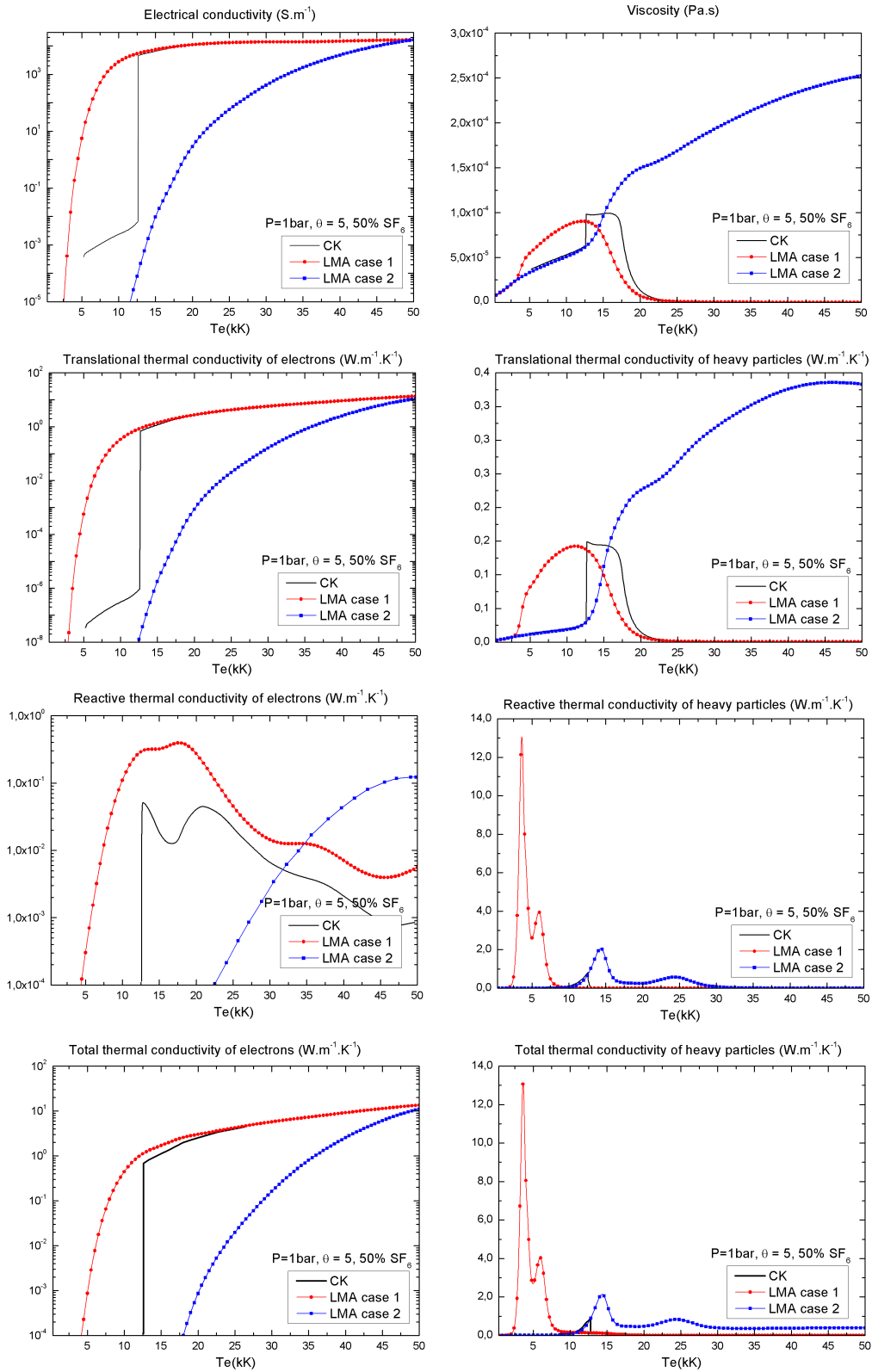


Figure 6. 2T transport coefficients of 50%SF₆-50%C₂F₄ (mass proportions) obtained with LMA, and CK methods, for $\theta = 5$ and 1 bar.

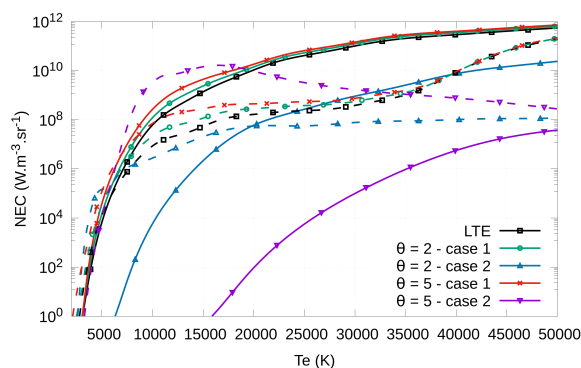


Figure 7. Influence of θ on the two NECs versus electron temperature for a pure SF_6 plasma at 1 bar and for $R_p = 1$ mm : full lines for the atomic lines, dashed lines for the continuum.

(used for electrodes or nozzles) but also the treatment of the RTE solving based on fine discretization and better approached methods. For NLTE/NLCE plasmas in HVCB, lots of challenges persist and must be solved rapidly in order to contribute to the improvement of the MHD modelling. For the plasmas compositions, it will be necessary to develop more precise models such as collisional radiative models or chemical kinetic models which are (for the moment) complex to achieve due to the lack of the reaction rates or collisional cross-sections. For the transport properties, the community must consider the thermal reactional conductivity and answer to the questions: should we consider only one coefficient (only for heavy particles for example), two contributions, a total thermal conductivity and how to calculate them under these conditions of non-thermal/chemical equilibrium? For the radiative properties, some works have been developed on CO_2 or air for re-entry atmosphere phenomena, syngas or combustion researches but few data are available for HVCB with or without metallic vapors due to the numerous chemical reactions. As the Planck and Kirchhoff laws are not accurate in non-equilibrium conditions, it is essential to treat differently the emission and absorption phenomena. The greatest difficulty lies in estimating correctly the populations energy levels, the emission and the absorption being proportional to these populations. The treatment of the atomic lines will be one of the main locks to raised (population of levels and broadenings being governed by different temperatures). New models will need to be developed to consider radiative phenomena separately: Hybrid Statistical Narrow Band model (HSNB) combining Statistical Narrow-Band model (SNB) for the optically thick molecular systems and Narrow Band means for the emission and absorption coefficients of the optically thin molecular systems and continua, a line by line (LBL) calculation for the bound-bound atomic lines [62]. Finally, these new methods and databank have to be validated with the comparison of MHD modeling results and experimental works.

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