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Determination of equilibrium composition of $C_xH_yO_zN_t$ plasmas out of thermodynamic equilibrium

Z. Koalaga[a]

Department of Physic, FAST - University of Ouagadougou, 03 B.P. 7021, Ouagadougou, Burkina Faso

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Abstract. In this paper, we determine the composition of $C_xH_yO_zN_t$ plasmas out of thermodynamic equilibrium using a more rigorous thermodynamic derivation of the Saha equation modified to two-temperature plasma system proposed by Chen *et al.* The calculation is made for these plasmas in pressure range 0.1–1 MPa and for electron temperature range 5 000–30 000 K. A great attention is given to the evolution of the major species such as hydrogen components (H, H_2), carbon monoxide (CO) and electrons. We compared the results obtained with our previous experimental and theoretical studies; the latter one has been carried out in equilibrium situation. We also compared our results with those given by the method based on the minimization of the Gibbs free energy widely used in the literature.

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1 Introduction

Some polymeric materials such as PMMA and nylon are commonly used in electrical apparatus for their potential of arc breaking [1–3]. The created plasma is essentially formed by insulator vapors [2,4–6] by means of complex physical processes. In previously published papers [7–10], the calculation of the composition, thermodynamic and transport properties for such plasmas in local thermodynamic equilibrium (LTE) condition have been shown. In these cases, the plasma is characterized by one temperature. But some studies have shown that plasma of polymeric materials seems to be out of thermodynamic equilibrium [11]. In order to take into account the thermodynamic disequilibrium, we introduce two temperatures into the plasma as an assumption: one temperature (T_e) for the electrons and the other (T_h) for the heavier species. The two-temperature ($2T$) composition of specific insulating substances (polyethylene and polyoxymethylene) has already been carried out by André [12] for heavy species in temperature ranging from 1 000 K to 6 000 K. The method used by this author is the Gibbs free energy minimization. In the literature [13–17], two quite different modified forms of the Saha equation were proposed and employed to take into account the thermodynamic disequilibrium. In recently published papers [18], Chen *et al.* have made a short synthesis of this problem and proposed a more rigorous thermodynamic derivation of the Saha equation modified to a two-temperature plasma system. In this paper, we use the formulation proposed by Chen *et al.* and apply it to the case of $C_xH_yO_zN_t$ plasmas; where x, y, z and t are the initial proportions of carbon, hydrogen, oxygen and nitrogen atoms. These plasmas gave good results in previous experimental study [2,3]. The work is divided in two main parts. In the first part, the physical model and the complete set of model equations are shown and solved. In the second part, the results obtained are discussed to show the influence of non-equilibrium state on the plasma composition. These results are also compared with those given by the minimization of Gibbs free energy.

2 Physical model

Due to the temperature range here considered (5 000–30 000 K), we take into account 32 chemical species, *i.e.*:

(i) electron: e^-;
(ii) 11 monatomic species: C, C^+, C^{++}, H, H^+, O, O^+, O^{++}, N, N^+, N^{++};
(iii) 20 diatomic species: C_2, H_2, O_2, N_2, CH, CO, CN, OH, HN, NO, CH^+, CO^+, CN^+, OH^+, HN^+, NO^+, C_2^+, H_2^+, O_2^+, $N_2^\pm$.

Following the experimental and theoretical works by Hettwer [19] about common polymeric materials, the polyatomic species can be neglect at high temperature (above 2 000 °C).

To determine the evolution of the 32 chemical species, we need to establish and to solve 32 equations *versus* T_e

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[a] e-mail: `koalaga@univ-ouaga.bf`

and T_{h} for a given pressure. The set of equations constitutes a nonlinear system of equations.

2.1 Nonlinear system of equations

To get the nonlinear system of equations, we consider the following equations and hypothesis:

(i) the Dalton's law given by:

$$P = kT_{\mathrm{e}}n_{\mathrm{e}} + kT_{\mathrm{h}} \sum_{i=1, i\neq \mathrm{e}}^{\nu} n_i - \Delta P \qquad (1)$$

where n_{e} and n_i are the species densities, k is the Boltzmann's constant, ν is the total number of species and ΔP is the pressure correction due to Colombian interaction given by:

$$\Delta P = \frac{1}{24\pi\varepsilon_0\lambda_{\mathrm{d}}} \sum_{i=1}^{\nu} q_i^2 n_i \qquad (2)$$

where q_i is the charge of species i and λ_{d} the Debye length given by [20]:

$$\lambda_{\mathrm{d}}^{-2} = \frac{e^2}{\varepsilon_0 k}\left[\frac{n_{\mathrm{e}}}{T_{\mathrm{e}}} + \sum_{i=1,i\neq \mathrm{e}}^{\nu} \frac{q_i^2}{T_{\mathrm{h}}}\right]; \qquad (3)$$

(ii) the electrical neutrality of the plasmas; this relation remains identical to that used in the thermodynamic equilibrium calculation case [2,8];

(iii) the relative stoichiometric concentration between carbon, hydrogen, oxygen and nitrogen atoms; because x, y, z, and t are variables and can take any value comprising zero, we use 3 main equations that are not function of T_{e} and T_{h};

(iv) the $2T$ laws for ionization and dissociation: following the paper of Chen *et al.* [18] and previous works using kinetic theory [15,20], the ionization equilibrium laws are:
for monatomic species ionization: $\mathrm{a} \leftrightarrow \mathrm{a}^{Z+} + e^-$

$$\frac{n_{\mathrm{e}}n_{\mathrm{a}^{Z+}}}{n_{\mathrm{a}}} = 2\frac{Q_{\mathrm{a}^{Z+}}(T_{\mathrm{e}})}{Q_{\mathrm{a}}(T_{\mathrm{e}})}\left(\frac{2\pi m_{\mathrm{e}}kT_{\mathrm{e}}}{h^2}\right)^{3/2} \exp\left(-\frac{E_{\mathrm{I}}}{kT_{\mathrm{e}}}\right) \qquad (4)$$

for diatomic species (ab or a_2) ionization: $\mathrm{ab} \leftrightarrow \mathrm{ab}^{Z+} + e^-$

$$\frac{n_{\mathrm{e}}n_{\mathrm{ab}^{Z+}}}{n_{\mathrm{ab}}} = 2\frac{Q_{\mathrm{ab}^{Z+}}(T_{\mathrm{e}})}{Q_{\mathrm{ab}}^{\mathrm{ex}}(T_{\mathrm{e}})Q_{\mathrm{ab}}^{\mathrm{rot}}(T_{\mathrm{h}})Q_{\mathrm{ab}}^{\mathrm{vib}}(T_{\mathrm{e}})} \times \left(\frac{2\pi m_{\mathrm{e}}kT_{\mathrm{e}}}{h^2}\right)^{3/2} \exp\left(-\frac{E_{\mathrm{I}}}{kT_{\mathrm{e}}}\right); \qquad (5)$$

while for the dissociation equilibrium laws: $\mathrm{ab} \leftrightarrow \mathrm{a} + \mathrm{b}$, they give

$$\frac{n_{\mathrm{a}}n_{\mathrm{b}}}{n_{\mathrm{ab}}} = \frac{Q_{\mathrm{a}}(T_{\mathrm{e}})Q_{\mathrm{b}}(T_{\mathrm{e}})}{Q_{\mathrm{ab}}^{\mathrm{ex}}(T_{\mathrm{e}})Q_{\mathrm{ab}}^{\mathrm{rot}}(T_{\mathrm{h}})Q_{\mathrm{ab}}^{\mathrm{vib}}(T_{\mathrm{e}})}\left(\frac{2\pi kT_{\mathrm{h}}}{h^2}\right)^{3/2} \times \left(\frac{m_{\mathrm{a}}m_{\mathrm{b}}}{m_{\mathrm{a}}+m_{\mathrm{b}}}\right)^{3/2} \exp\left(-\frac{E_{\mathrm{d}}}{kT_{\mathrm{e}}}\right) \cdot \qquad (6)$$

In the three relations (4–6), E_{d} is the dissociation energy of the ab molecules, E_{I} is the ionization energy of the atomic or molecular species, $Q_i(T_{\mathrm{e}})$ is the internal partition function of monatomic or ionized diatomic particle i and $Q_{\mathrm{ab}}^{\mathrm{ex}}(T_{\mathrm{e}})$, $Q_{\mathrm{ab}}^{\mathrm{vib}}(T_{\mathrm{e}})$, $Q_{\mathrm{ab}}^{\mathrm{rot}}(T_{\mathrm{h}})$ are respectively the diatomic species partition function for excitation energy, vibration energy and rotational energy states. We have used 16 ionization and 10 dissociation reactions in addition to equations (i) to (iii).

2.2 Complementary equations

Because one or more parameters x, y, z and t can take the value zero, it is then possible that some relations defined in (ii) become invalid. If this happens, we must replace these equations with other equations called "Complementary Equations" to be able to compute any $\mathrm{C}_x\mathrm{H}_y\mathrm{O}_z\mathrm{N}_t$ plasma composition. These complementary equations are also independent of T_{e} and T_{h}, like the 3 principal equations.

2.3 Internal partition function (IPF)

The computation of the internal partition function can have an influence on the composition of plasma. To compute the IPF appearing in the $2T$ laws for ionization and dissociation, the following assumptions are commonly made [20]:

- the atomic IPF depends only on T_{e} because the excited levels of atomic species are mainly populated by electron collisions;
- for diatomic molecules, the IPF used is a function of T_{e} and T_{h}. The IPF expression is given by [18]:

$$Q_{\mathrm{ab}}^{\mathrm{int}} = Q_{\mathrm{ab}}^{\mathrm{ex}}(T_{\mathrm{e}})Q_{\mathrm{ab}}^{\mathrm{vib}}(T_{\mathrm{e}})Q_{\mathrm{ab}}^{\mathrm{rot}}(T_{\mathrm{h}}) \cdot \qquad (7)$$

Now, we have all the equations necessary to the calculation of the composition of the plasma. The calculation method is derived from our previous works under LTE conditions [8,9]. It is important to say that the system of equation is solved numerically using the Newton-Raphson method; for more details see references [8,9].

3 Results and discussion

3.1 Testing the computer program

First of all, we consider the thermodynamic equilibrium state in order to test our computer program. The case of Perspex (PMMA) is shown in Figure 1. These results are in good agreement with published results [9]. For single gases such as N_2, H_2 and O_2 [17–23] and some mixtures of gases [17,24], calculations of $2T$ plasma composition were performed. With our program, we also determined the composition for single gases N_2, H_2 and O_2 out of thermodynamic equilibrium (OTE). The comparison with André's results [17] shows some differences for

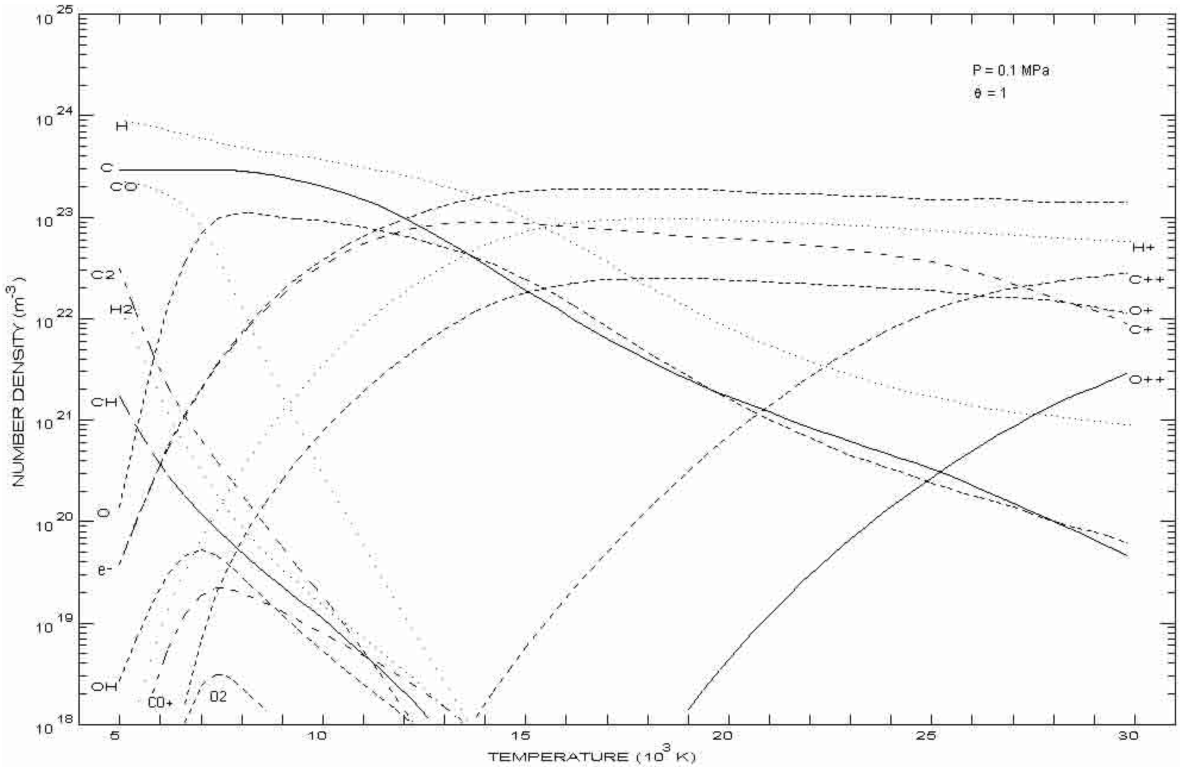

Fig. 1. Species densities *versus* the electrons temperature of PMMA plasma at atmospheric pressure under local thermodynamic equilibrium ($\theta = 1$).

all species at low temperatures below 10 000 K. For $T_e \approx$ 15 000 K, the deviations are less significant. Because hydrogen species play a great role on the performance of the plasma, we give in Figure 2 the composition of H_2 plasma for $\theta = T_e/T_h = 1$, 1.5 and 2. In conclusion, we see that the concentration of the various species grows with the thermodynamic disequilibrium.

3.2 $C_xH_yO_zN_t$ plasmas composition

After testing the program, we consider some polymeric materials such as Perspex or PMMA ($C_5H_8O_2$), melamine ($C_2H_6N_6$), polyoxymethylene (POM:CH_2O) and nylon6-6 ($C_{12}H_{22}O_2N_2$). Experimentally, these materials gave good results for arc breaking [6] and are also been studied theoretically in LTE condition [2,8,9]. For each plasma, we determine the concentration of the species for $\theta = 1.5$ and 2; the pressure is set to one atmosphere (0.1 MPa) and the electron temperature is in the range 5 000–30 000 K. Figures 3 to 10 show the results obtained for Perspex, melamine, POM and nylon6-6. From theses figures, we can deduce the following observations.

(a) In all cases, the monatomic specie that has the highest concentration value is the hydrogen atom in all the temperature range. By comparing with the thermodynamic equilibrium composition, we note that monatomic species concentration increases at a given temperature. As we showed in the LTE condition, the increase of hydrogen particles will contribute to reduce the time of the quenching of the arc. So we can obtain a quick breaking of the arc comparing to classical technique using arc chutes with steel plates. An other monatomic specie that has an influence on the plasma characteristics is the carbon. Indeed, as we will see it further, the carbon provides the major part of the electrons of which the density determines the value of electrical conductivity. We remark that for this particle, the density increases with the increase of the thermal disequilibrium.

(b) Diatomic particles appear at temperature bellow 15 000 K. Highest values for these kinds of species are observed for CO, C_2, H_2, N_2 and CH. For plasmas containing oxygen in their formula, we observe that oxygen species are less important. The analysis of the evolution of the densities of ionized diatomic particles let's think that these species can have a great influence on the electronic density at very low temperatures for high disequilibrium.

(c) When θ increase from 1 to 2, we observe that all species densities increase except for oxygen. For this particle the density decreases initially for temperature lower than 6 500 K, and increases then for the higher temperatures (see Fig. 11). This shows that the reactions of dissociation take place at temperatures relatively higher compared to LTE condition. This shift to high temperature is not so significant, but this phenomena can have an influence on the thermal conductivity. As a matter of fact, Hertz [25] have shown that thermal conductivity of reactions play a major role on the value

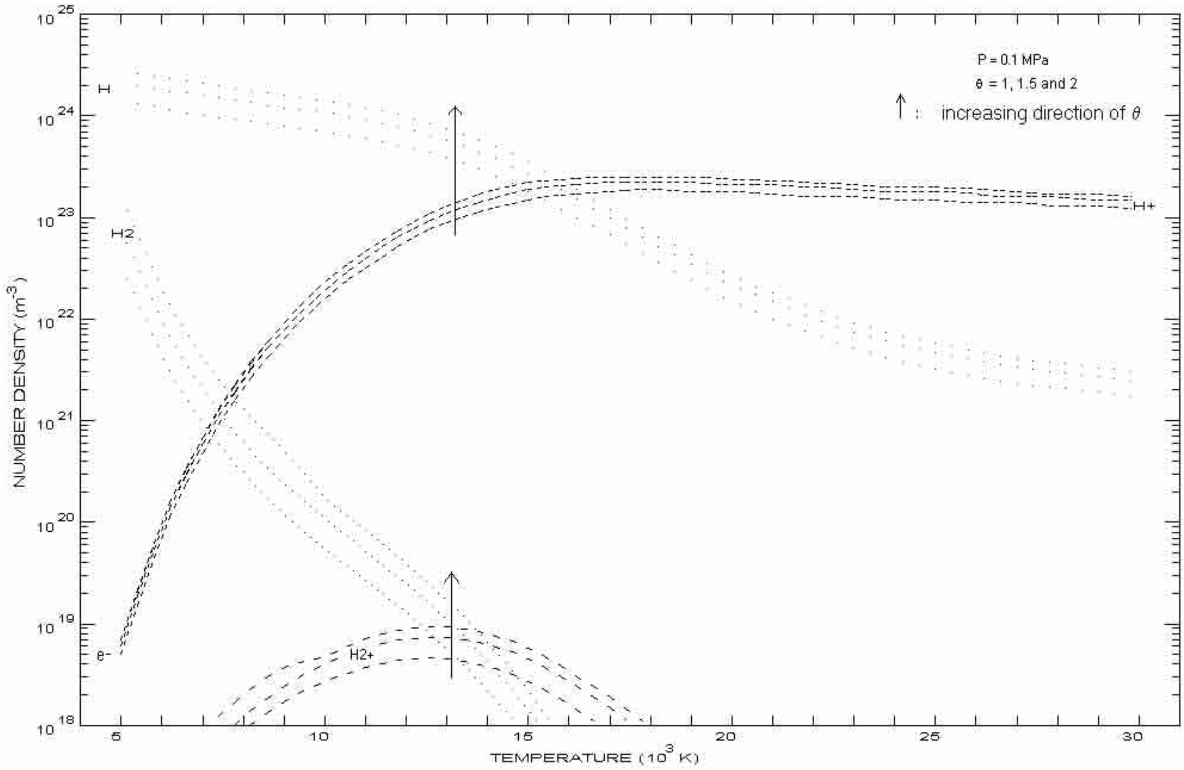

Fig. 2. Species densities *versus* the electrons temperature of hydrogen plasma at atmospheric pressure for three value of thermodynamic disequilibrium ($\theta = 1$, 1.5 and 2).

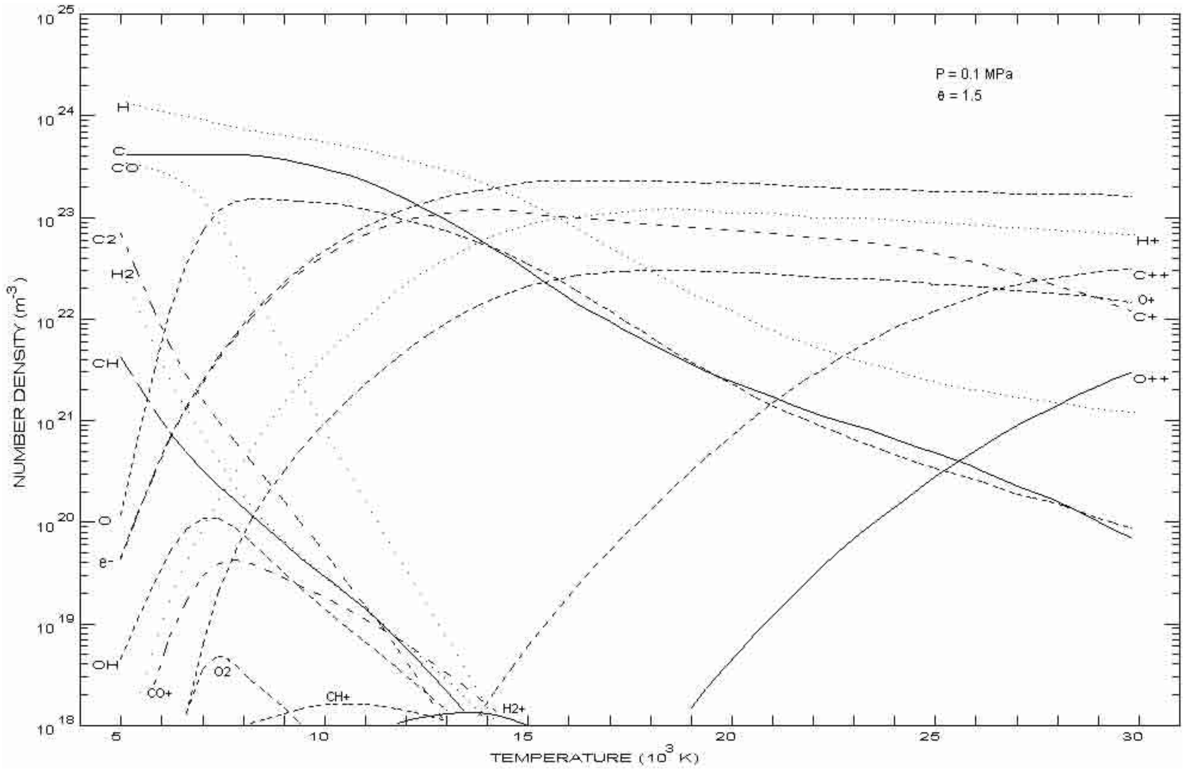

Fig. 3. Species densities *versus* the electrons temperature of PMMA plasma at atmospheric pressure and out of thermodynamic equilibrium ($\theta = 1.5$).

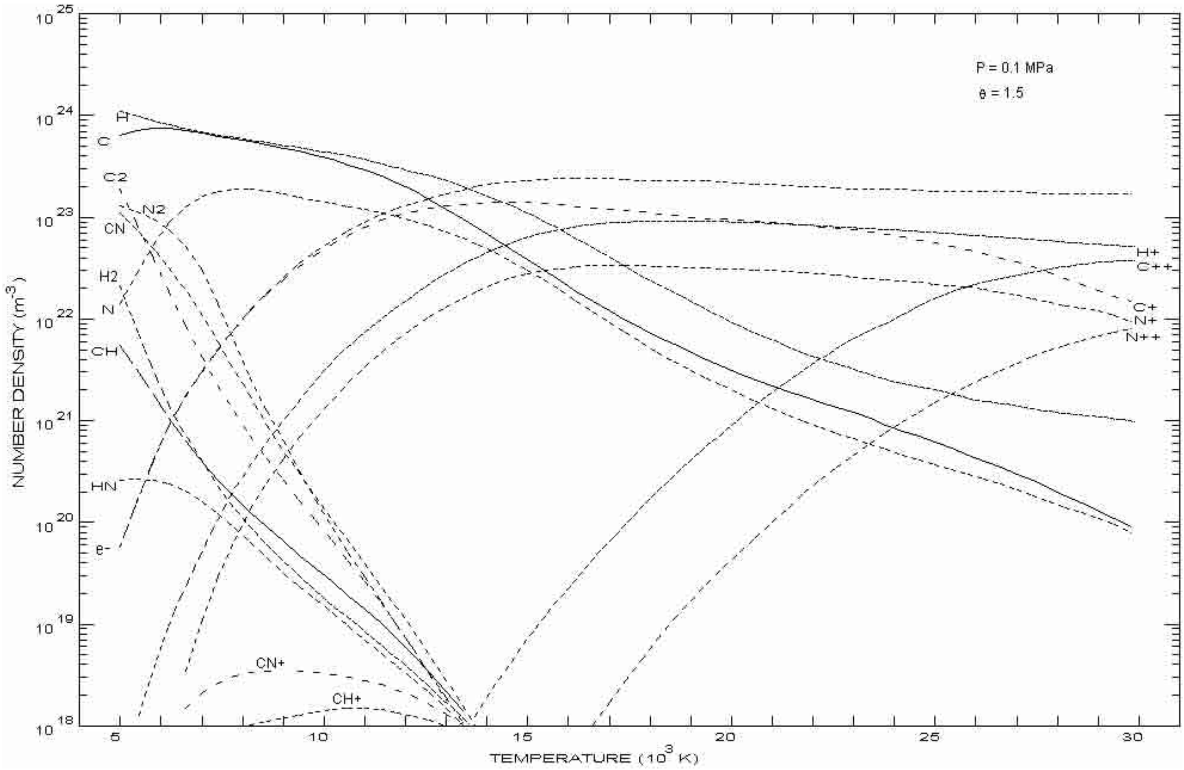

Fig. 4. Species densities *versus* the electrons temperature of melamine plasma at atmospheric pressure and out of thermodynamic equilibrium ($\theta = 1.5$).

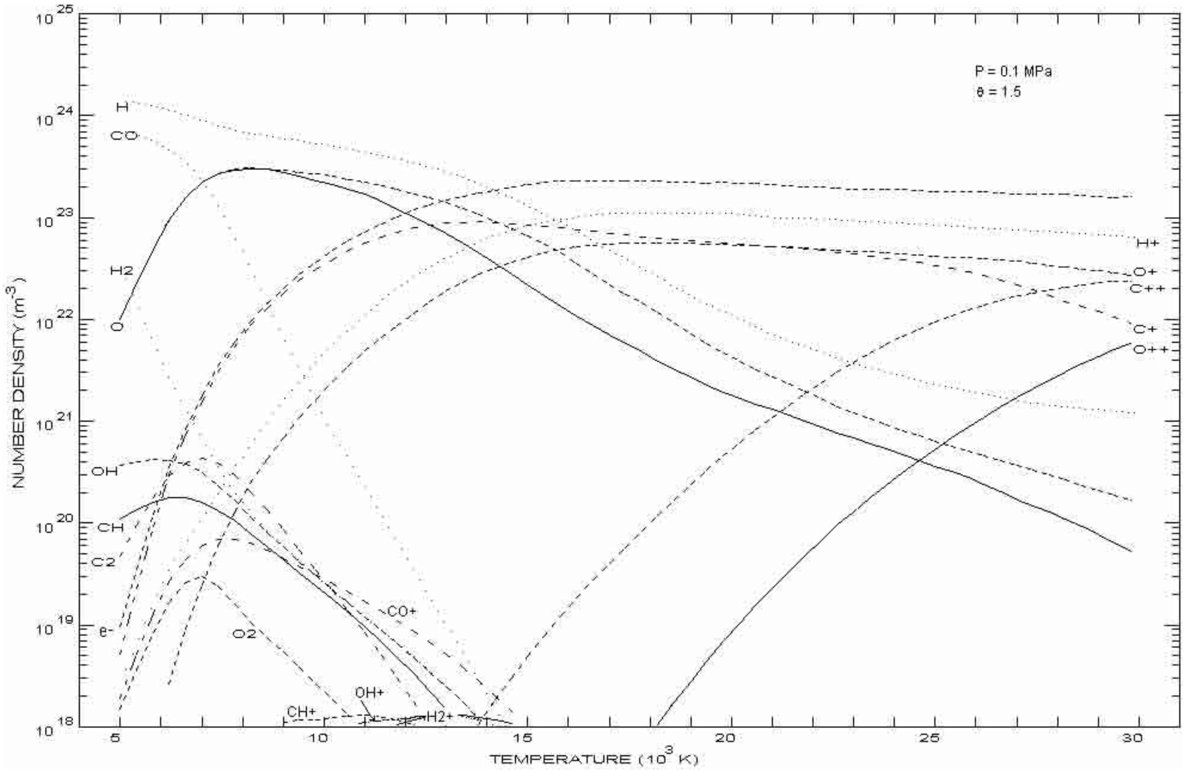

Fig. 5. Species densities *versus* the electrons temperature of POM plasma at atmospheric pressure and out of thermodynamic equilibrium ($\theta = 1.5$).

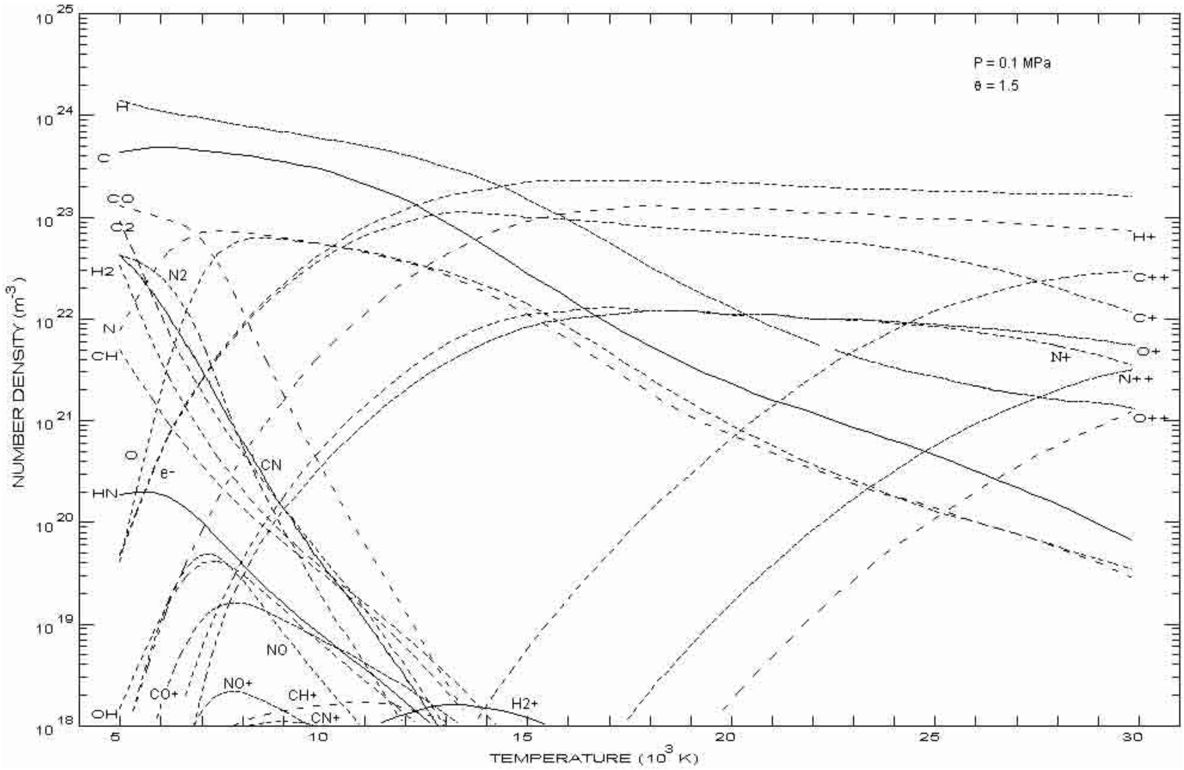

Fig. 6. Species densities *versus* the electrons temperature of nylon plasma at atmospheric pressure and out of thermodynamic equilibrium ($\theta = 1.5$).

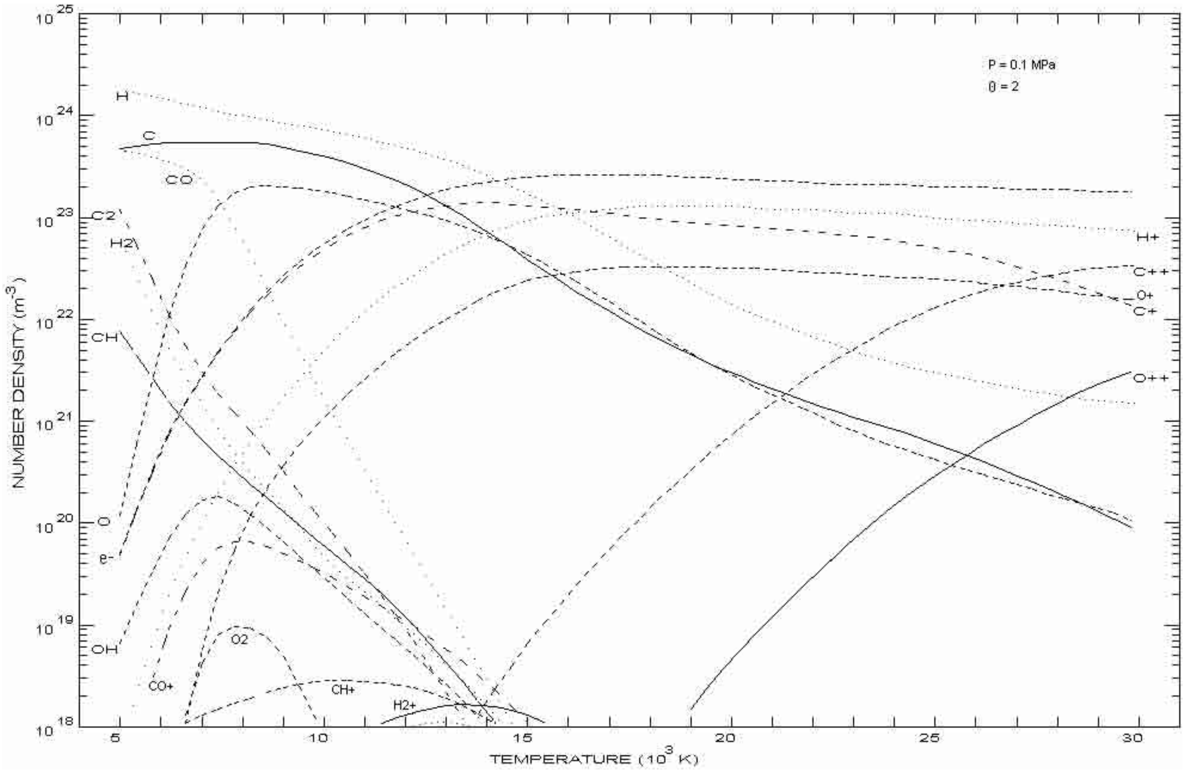

Fig. 7. Species densities *versus* the electrons temperature of PMMA plasma at atmospheric pressure and out of thermodynamic equilibrium ($\theta = 2$).

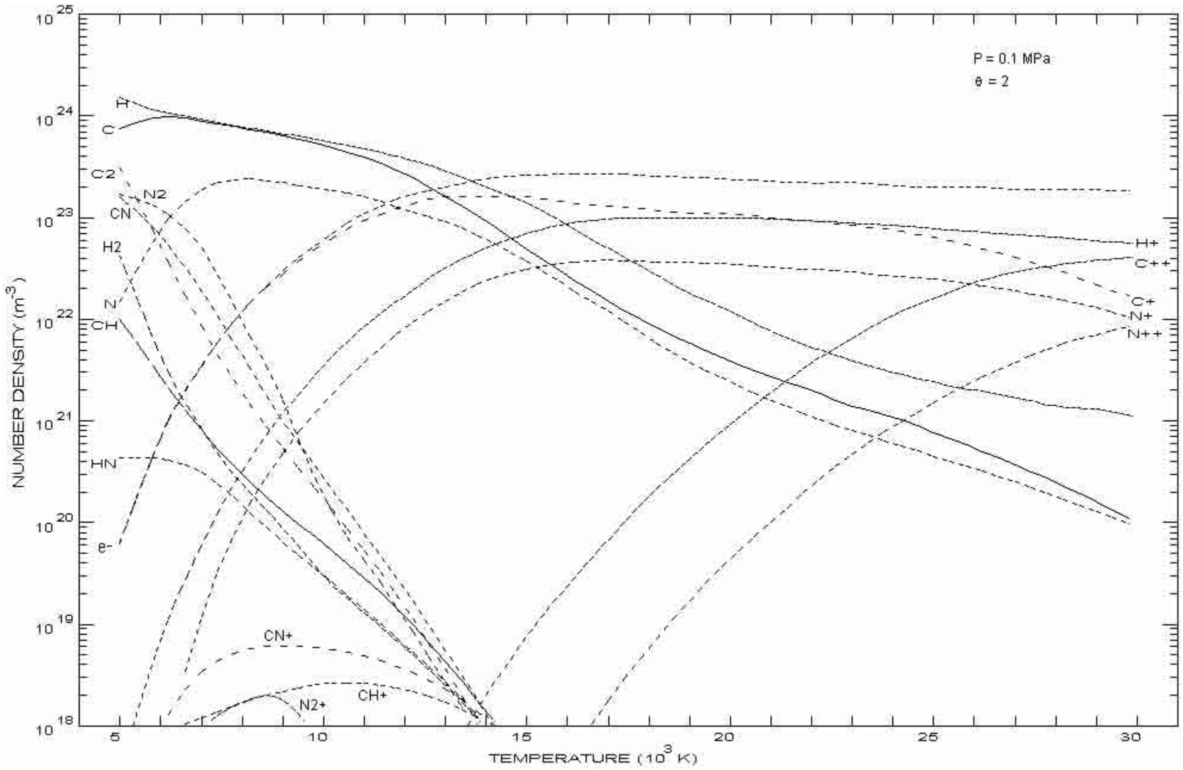

Fig. 8. Species densities *versus* the electrons temperature of melamine plasma at atmospheric pressure and out of thermodynamic equilibrium ($\theta = 2$).

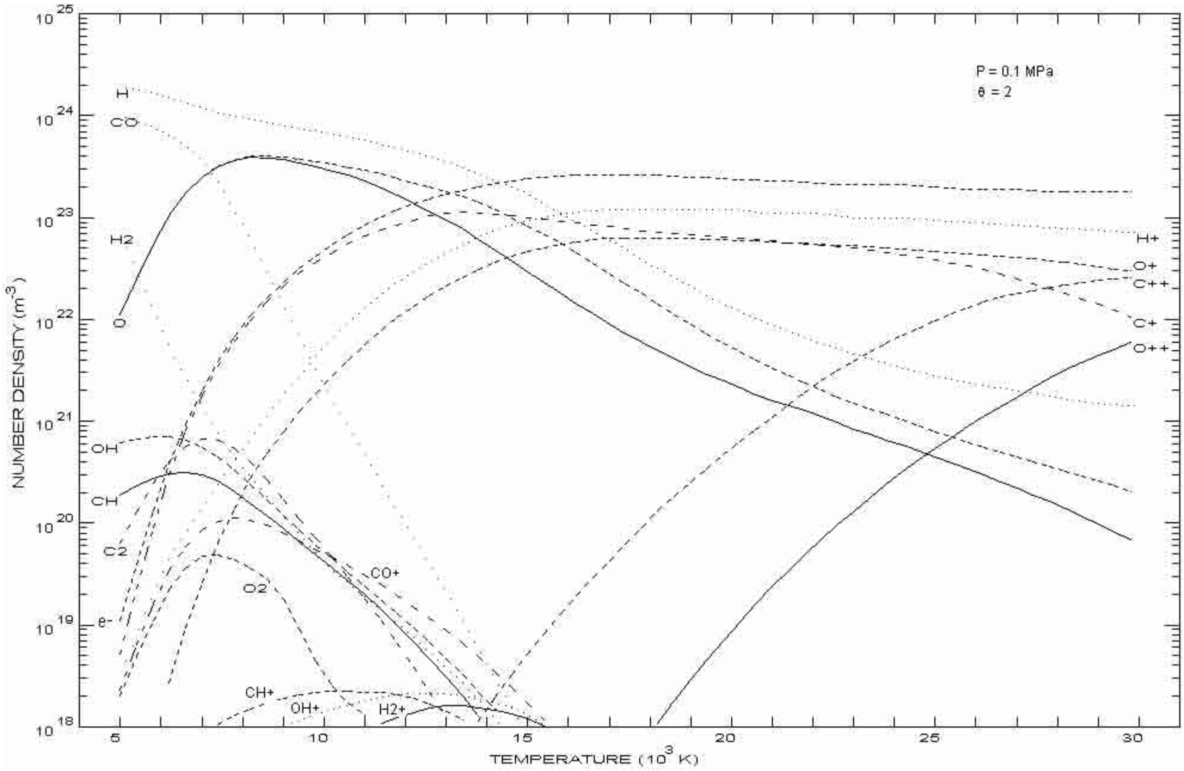

Fig. 9. Species densities *versus* the electrons temperature of POM plasma at atmospheric pressure and out of thermodynamic equilibrium ($\theta = 2$).

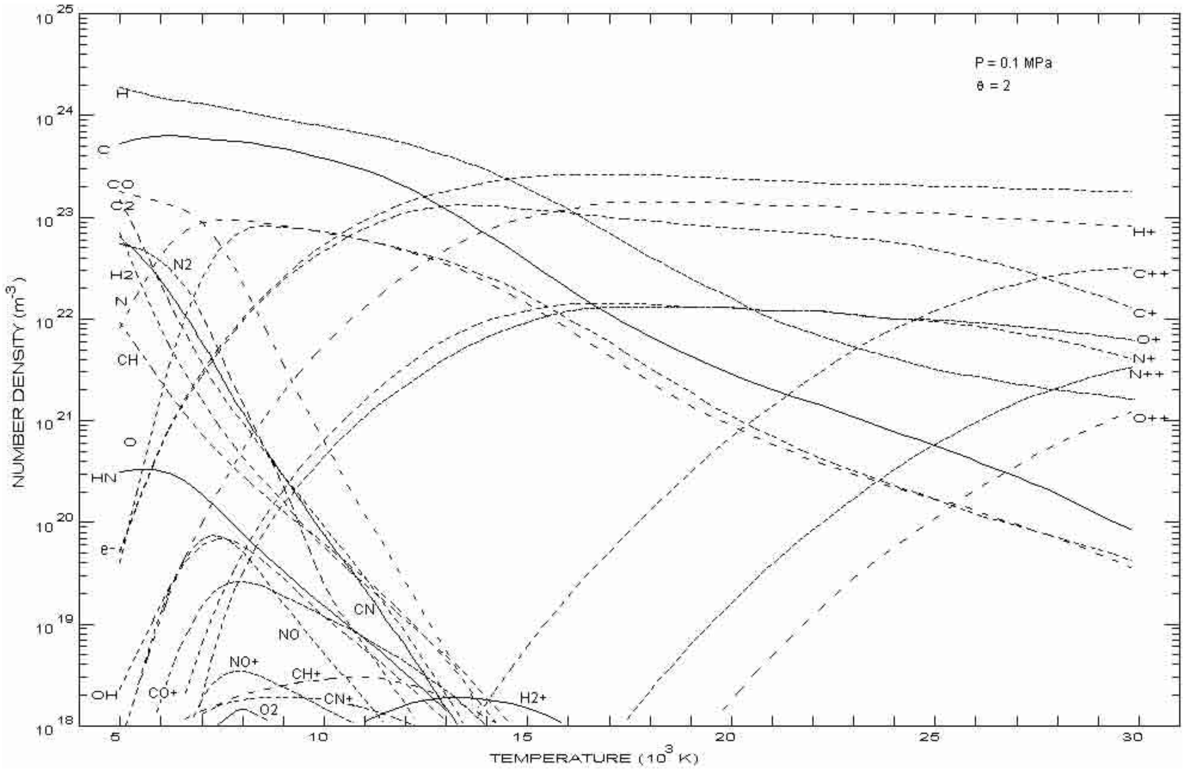

Fig. 10. Species densities *versus* the electrons temperature of nylon plasma at atmospheric pressure and out of thermodynamic equilibrium ($\theta = 2$).

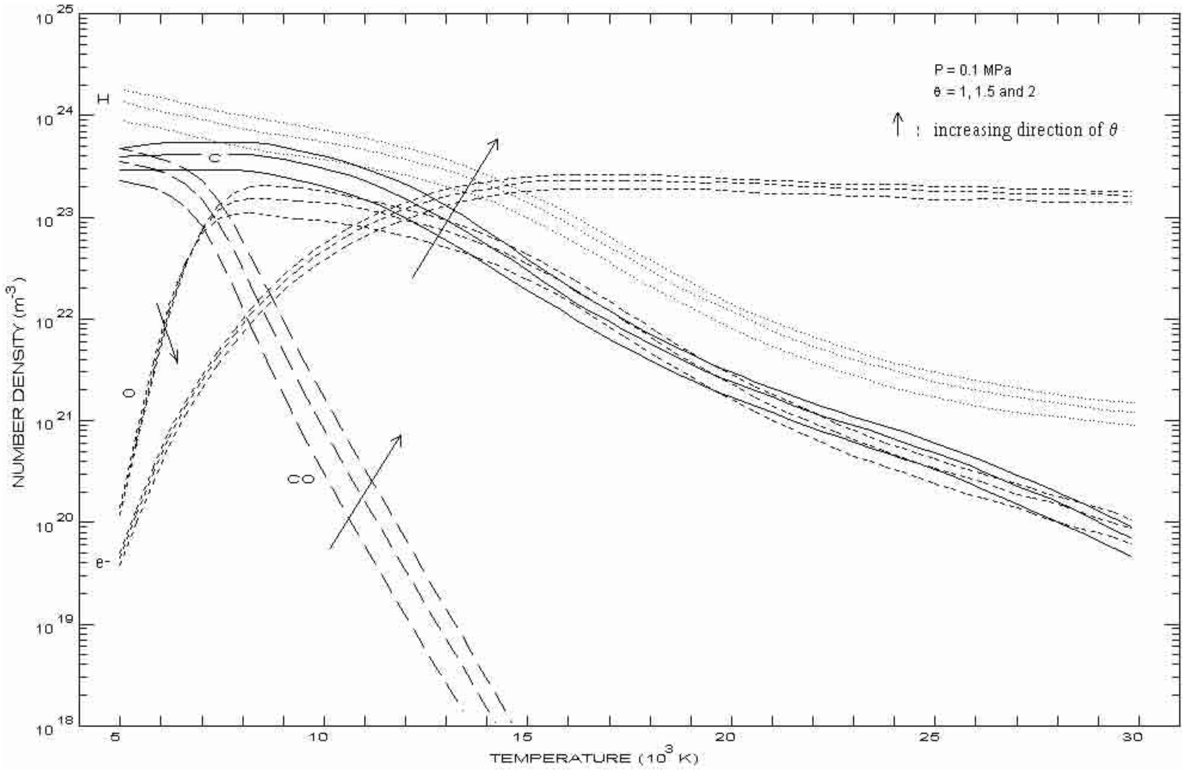

Fig. 11. Species densities *versus* the electrons temperature of PMMA plasma at atmospheric pressure for three values of thermodynamic disequilibrium ($\theta = 1$, 1.5 and 2).

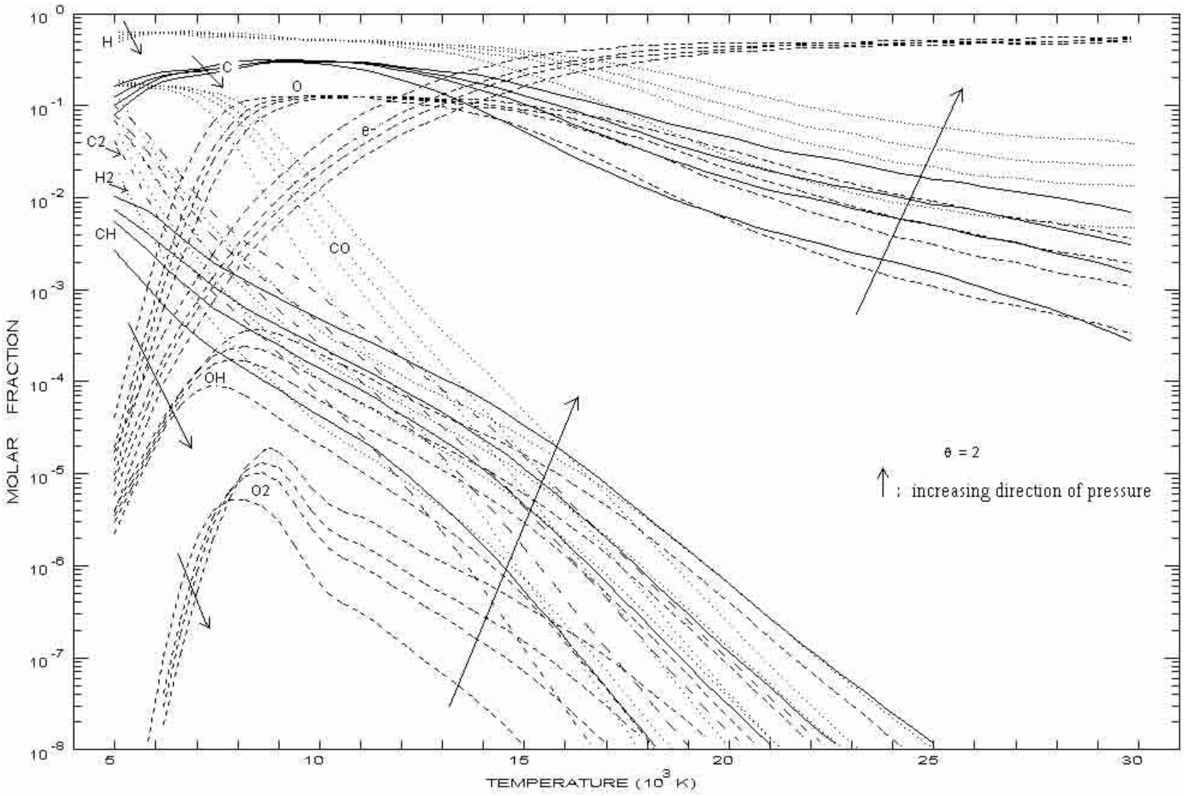

Fig. 12. Electrons and non-ionized species densities *versus* the electrons temperature of PMMA plasma out of thermodynamic equilibrium ($\theta = 2$) for four values of the pressure ($P = 0.1$, 0.3, 0.5 and 1 MPa).

of time-constant of the arc. To determine how θ values influence the thermal conductivity, it is necessary to compute the transport properties of $C_xH_yO_zN_t$ plasmas out of thermodynamic equilibrium (OTE). This will be the topic of our next paper.

At a given temperature, electron density and ionized monatomic species density increase with the increasing value of θ and for temperature above 8 000 K. To show clearly the influence of θ on the electron concentration, we choose the case of Perspex plasma and compute it's $2T$ composition for different values of θ ($\theta = 1$, 1.5 and 2). In Figure 11, the results obtained are reported for electrons, H, C, O and CO. The ionization of carbon atoms is the major provider of electron density and hydrogen's ionization become significant at temperature above 16 000 K. The same observations are also been made for other polymeric materials. But for melamine, the hydrogen's ionization occurs after 20 000 K. The contribution of ionization of oxygen and nitrogen is weak for all temperatures and for all plasmas. However, the observation of the evolution of O^+ density according to the disequilibrium shows that ionization reactions are slightly shifted towards the high temperatures.

(d) The influence of pressure in OTE is remarkable, since the ablation plasmas are characterized by high pressure during the arc live. The pressure can vary up to 5 atmospheres and even more. In Figures 12 and 13, the concentration evolution of species is reported with pressure $p = 1$, 3, 5 and 10 atmospheres and for one value of the disequilibrium parameter $\theta = 2$. In these figures, we have represented the evolution of the molar fractions of the species to show the influence of the pressure on the plasma composition. In each figure, the direction of the arrow on the curves indicates the increasing direction of the pressure. At low temperature, the molar fractions of electron and ionized species decrease when the pressure increases for a given temperature. For high temperatures, the molar fractions of electrons, H^+ and O^+ species tend towards values independent of the pressure. For C^+ atom, this value is observed for temperature ranging in 18 000–24 000 K. At temperature above 24 000 K, the molar fraction of C^+ increases as the pressure increases. We can predict a similar evolution for electrons, H^+ and O^+ species but outside of our range of temperature.

For most of neutral diatomic species, the molar fraction increases when pressure increases. But when we consider the case of OH molecule, we remark that for a given temperature lower than 7 500 K, the molar fraction decreases with the pressure, but increases for temperatures higher than this value of 7 500 K. Theses observations are valid for oxygen and even for carbon. The conclusion of all this is that the reactions of dissociation and ionization take place at temperature all the more high since the pressure increases. For the LTE, we arrive at identical conclusions, but the reactions take

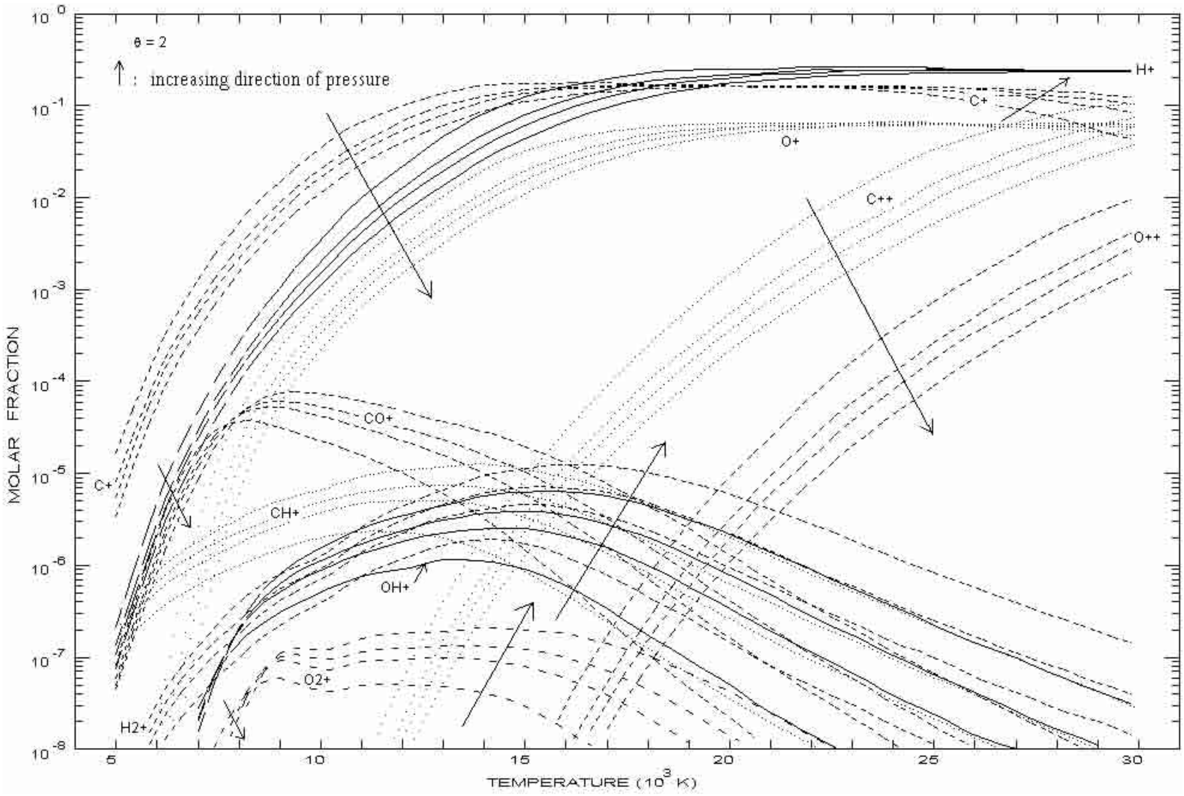

Fig. 13. Ionized species densities *versus* the electrons temperature of PMMA plasma out of thermodynamic equilibrium ($\theta = 2$) for three values of the pressure ($P = 0.1$, 0.3, 0.5 and 1 MPa).

place at temperature less higher than the temperature reached with the OTE.

(e) Influence of IPF calculation: as said by André [12], one of the important difficulties in the determination of species concentration is the choice of the correct hypothesis for the vibration temperature. For our study, we have supposed the following hypothesis called hypothesis 1:

$$\begin{cases} T_{\mathrm{rot}} = T_{\mathrm{tr}} = T_{\mathrm{h}} \\ T_{\mathrm{vib}} = T_{\mathrm{ex}} = T_{\mathrm{e}} = \theta\, T_{\mathrm{h}} \end{cases}$$

where T_{ex} is the excitation temperature for monatomic and diatomic species and T_{tr} the translational temperature. To show the influence of this choice, we have also determined the $2T$ composition of Perspex plasma for two others hypothesizes:

hypothesis 2:

$$\begin{cases} T_{\mathrm{vib}} = T_{\mathrm{rot}} = T_{\mathrm{h}} \\ T_{\mathrm{ex}} \;= T_{\mathrm{e}} \;\;= \theta\, T_{\mathrm{h}} \end{cases},$$

hypothesis 3:

$$\begin{cases} T_{\mathrm{ex\,diatom}} = T_{\mathrm{vib}} = T_{\mathrm{rot}} = T_{\mathrm{h}} \\ T_{\mathrm{ex\,atom}} \;\;= T_{\mathrm{e}} \;\;= \theta\, T_{\mathrm{h}} \end{cases}.$$

The last assumption supposes in fact that the IPF of the diatomic molecules is only a function of T_{h}. We choose $\theta = 2$ and perform the calculation of species concentrations that we reported in Figures 14 and 15. The results show that for temperatures higher than 15 000 K, the choice of various temperatures does not have any influence on the densities of particles present in the plasma in both cases. On the other hand, at low temperatures we see that this choice can modify the values of species concentrations. The influence of the choice of T_{vib} and T_{ex} is more clear on the densities of the neutral diatomic species. When T_{vib} tends towards T_{e}, their concentration increases for a given temperature (see Fig. 15). For the monatomic species, this influence is appreciable only for the oxygen particle at temperature below 8 000 K. Comparing the two figures, we can see that uncoupling the excitation temperatures of simple atoms and diatomic species does not involve notable changes in the composition of plasma. Due to this fact, the two figures seem to be identical.

3.3 Comparison with the method of the Gibbs free energy minimization

The present work is based on the recent investigation made by Chen *et al.* [18] on the two-temperature plasmas. But in the literature, the greater part of the results of two-temperature plasmas is obtained by using the minimization of the Gibbs free energy [13,17,22–24]. However, the use of this method seems to be an invalid procedure

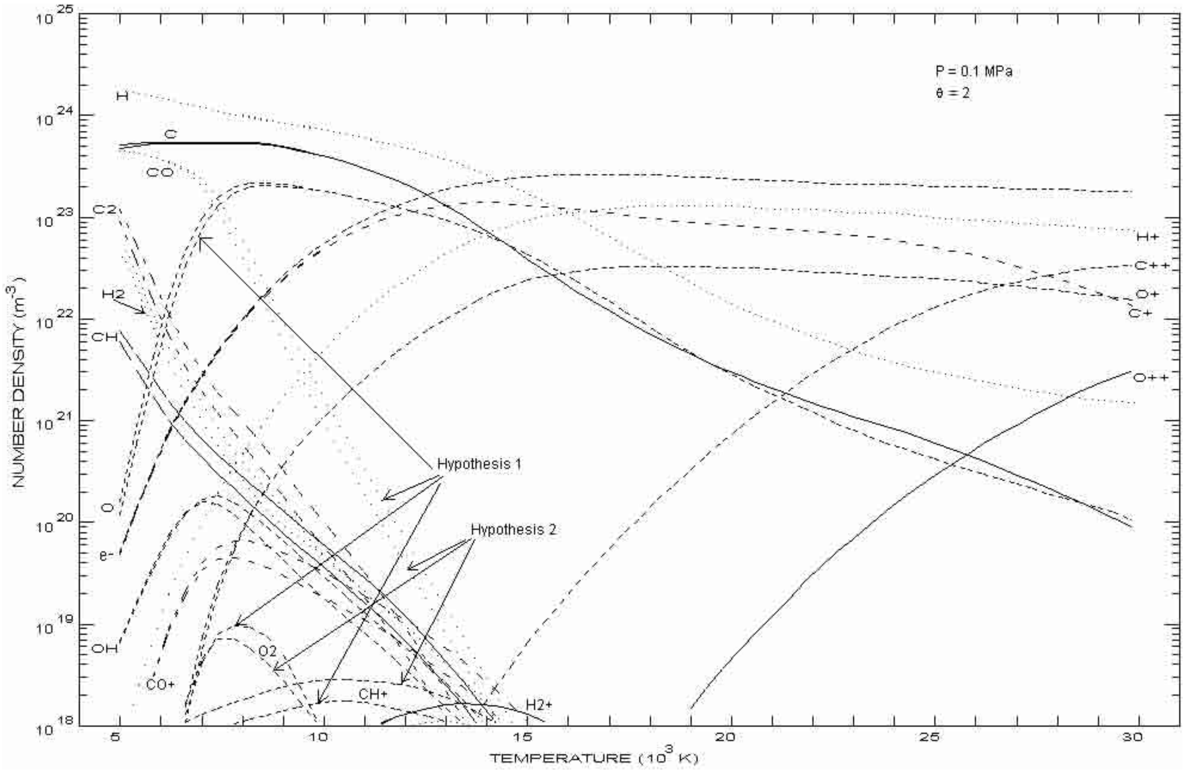

Fig. 14. The influence of the choice of the vibration temperature on species densities of PMMA plasma at atmospheric pressure and out of thermodynamic equilibrium ($\theta = 2$).

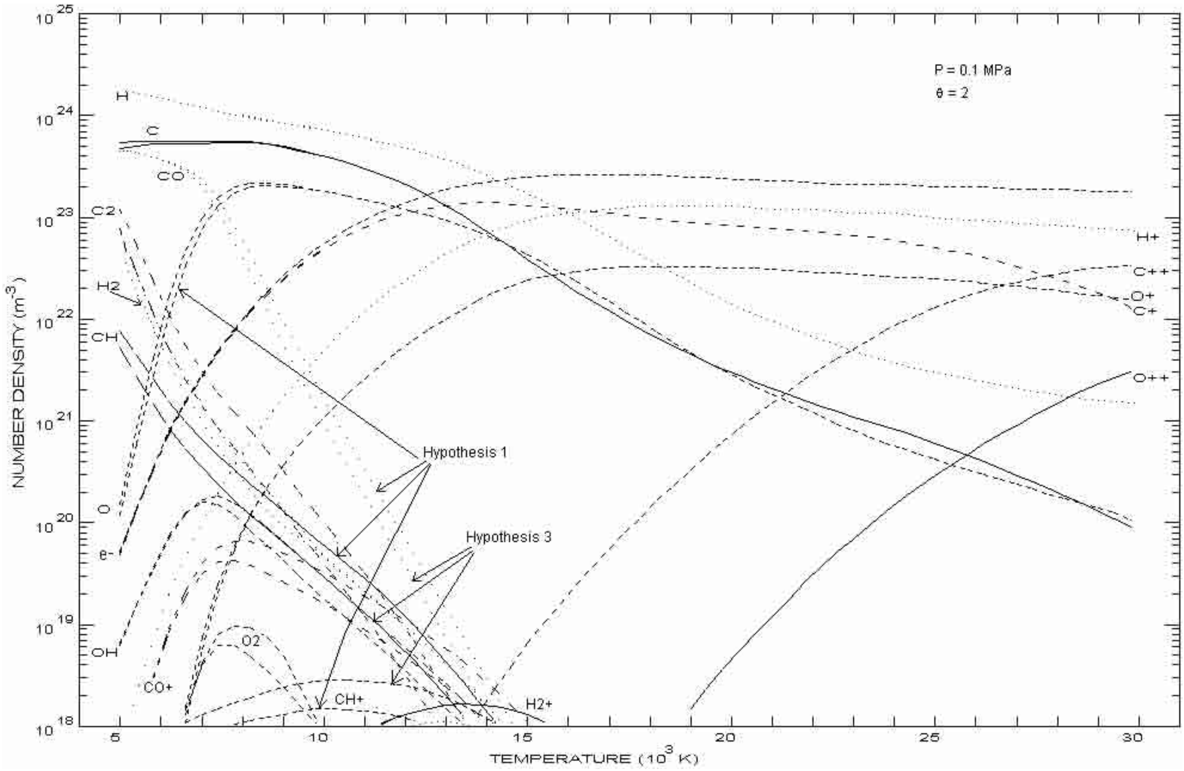

Fig. 15. The influence of the choice of the vibration temperature and the uncoupling of the excitation temperature on species densities of PMMA plasma at atmospheric pressure and out of thermodynamic equilibrium ($\theta = 2$).

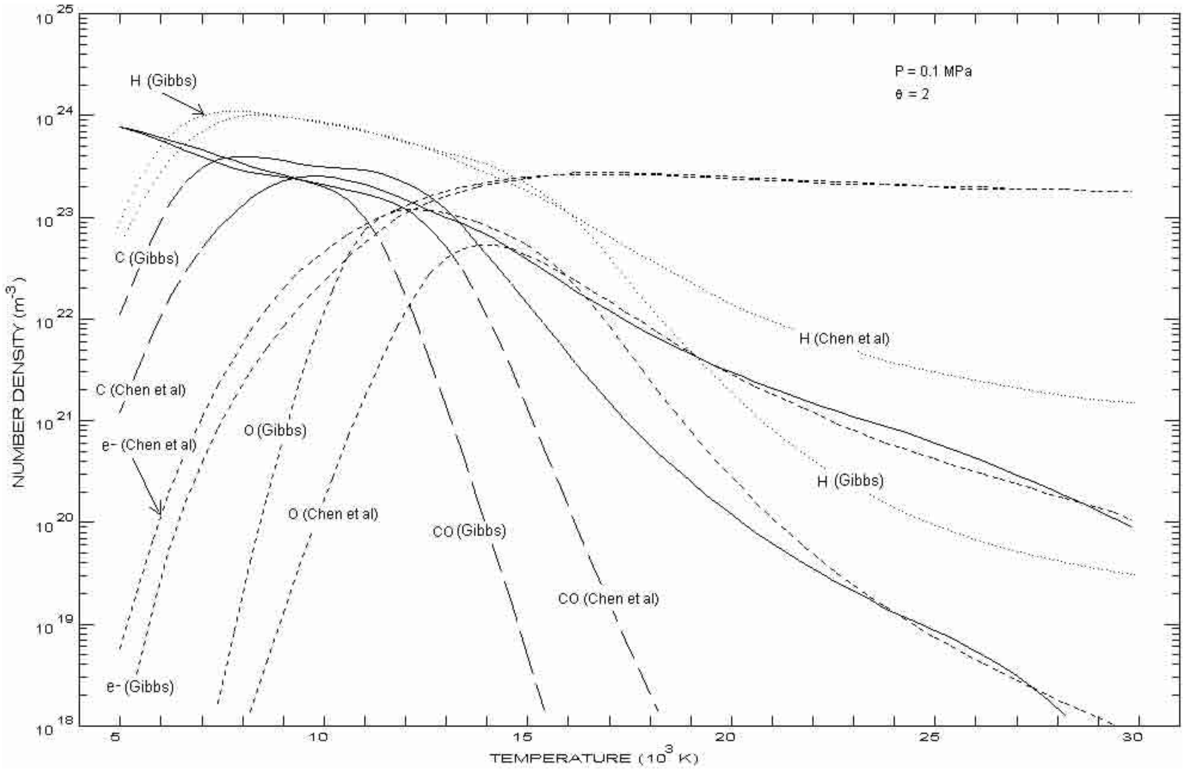

Fig. 16. The influence of entropy maximization and free energy minimization methods on species densities of PMMA plasma at atmospheric pressure and out of thermodynamic equilibrium ($\theta = 2$).

for Richley *et al.* [14], van der Saden *et al.* [15] and Chen *et al.* [18]. Even though Gordano and Capitelli [26] claim that both methods (maximization of entropy and minimization of free energy) are still valid depending on the constraint exerted on the two-temperature plasma system, Gleizes *et al.* [20] in a recent work recommend a method similar to the one used in our study.

To show the influence of both methods on the plasma composition, we have reported the evolution of species concentration for Perspex plasma ($C_5H_8O_2$) in Figure 16. We have taken into account only the hydrogen (H), carbon (C), oxygen (O), carbon monoxide (CO) and electrons. The words (Gibbs) and (Chen *et al.*) following the species symbol indicate the results obtained respectively with the minimization of Gibbs free energy and the maximization of entropy.

A comparison of both methods shows significant differences in the species concentration for all temperatures. The main results from minimization of free energy method are:

- a lowest concentration of electron for temperature below 11 000 K,
- a highest significant shift of the reaction of dissociation and ionization towards the high temperatures,
- under 8 500 K and upper to 17 000 K, the values of hydrogen density are lower than those obtained in our study. In intermediate range, differences between values given by both methods are weak.

The same observations can be made for other polymeric plasmas. They may lead to think that the method of minimization of Gibbs free energy will give a plasma having not only a lower electric conductivity, but also a lower thermal conductivity. Meanwhile, a detailed study of the transport coefficients is necessary for a confirmation of these tendencies.

4 Conclusion

The plasmas $C_xH_yO_zN_t$ formed from ablated vapors are in a non-equilibrium thermodynamic state. Therefore the chemical equilibrium composition is calculated using a more rigorous thermodynamic derivation of the Saha equation modified according to a two-temperature plasma system. The calculations not only have shown the evolution of the chemical species in plasmas out of thermodynamic equilibrium, but have shown also the importance of the determination of transport and thermodynamic properties of plasmas. From this study, we can make the following conclusions.

- The thermodynamic disequilibrium has a clear influence on the major species concentrations such as H, H_2, CO, C and electrons. At a given temperature, species density increases with the increase of the non-equilibrium state.
- At a given thermodynamic disequilibrium, dissociation and ionization reactions take place at temperatures

relatively higher compared to LTE condition. The shift to high temperature increases with increasing thermodynamic disequilibrium. This can have a great influence on the time-constant of the arc.

- In the range of temperatures utilized, the choice of various internal temperatures for the calculation of the IPF modifies slightly the densities of diatomic species at low temperature. But for temperature below 15 000 K, there is no influence on the densities of all particles.
- The pressure has an influence on the composition of the plasma out of thermodynamic equilibrium. Indeed, the reactions of dissociation and ionization take place at temperatures higher when the pressure increases. The displacement of the reactions towards the high temperatures is all the more marked when the disequilibrium becomes significant.
- In comparison, maximization of entropy and minimization of free energy methods present significant differences in the species concentration for all temperatures. The suggested tendencies about electric and thermal conductivity have to be confirmed by further detailed study.

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