

# Numerical Simulation of Multi-Component Inductive Plasma Flows under Chemical Non-Equilibrium

DAVID VANDEN ABEELE,<sup>a,c,d</sup> PAOLO BARBANTE,<sup>b,c</sup> GÉRARD DEGREZ,<sup>c</sup>

JUAN-PEDRO MELLADO GONZÁLEZ,<sup>c,e</sup>

<sup>c</sup> von Karman Institute for Fluid Dynamics, Brussels, Belgium

<sup>d</sup> Center for Plasma-Astrophysics, Catholic University of Leuven, Heverlee, Belgium

<sup>e</sup> Universidad Politécnica de Madrid, Spain

**ABSTRACT:** This contribution presents an elegant and efficient numerical model of a multi-component inductive plasma under chemical non-equilibrium. The plasma is modeled by a finite number of species. Chemical reactions are taken into account by Arrhenius-type source terms. The plasma thermodynamic properties are computed from statistical mechanics. The plasma transport properties are computed with the method of Chapman and Enskog. Multi-component species diffusive fluxes are obtained from the full Stefan-Maxwell equations by means of a straightforward iterative procedure, in which no distinction needs to be made between electrons and heavy particles. The governing equations are discretized in a second order accurate cell-centered finite volume manner on a collocated mesh. The discretized system of equations is solved with a damped Newton method. Powerful Krylov subspace methods are used to efficiently solve the linear systems arising from the Newton linearization. Results are presented for a five-species nitrogen plasma. The effect of chemical non-equilibrium is investigated at various pressures. Though some problems still need to be addressed, good convergence is already found. Calculations of eleven-species air plasmas under thermochemical non-equilibrium seem feasible for the near future.

## INTRODUCTION

In an inductive plasma torch, a gas is heated in an electrodeless manner to a plasma state with peak temperatures of about 10 000 K. The non-polluted plasma thus obtained is well-suited to a wide variety of industrial and scientific applications.<sup>1</sup> In the aerospace industry, air inductive plasmas are used to test thermal protection systems for re-entry space vehicles. To properly simulate re-entry flight conditions, inductive plasmas need to be operated at pressures below 0.1 atm, where thermal and chemical non-equilibrium effects are known to be important.<sup>2</sup>

To the authors' best knowledge, continuum models of inductive plasmas under thermal and chemical non equilibrium have already been presented by Mostaghimi et al.<sup>3</sup> (1987), by Semin<sup>4</sup> (1991) and by Benoy<sup>5</sup> (1993). However, these models only dealt with the relatively straightforward case of a three-component plasma (argon) and can not be easily extended to more general multi-component mixtures such as air. Whereas argon plasmas can be modeled with a straightforward three-component formalism, a full multi-component formalism (e.g. multi-component diffusion model) is required to simulate more complex mixtures.

Moreover, the evaluation of the thermodynamic and transport properties of a multi-component plasma mixture is usually very costly, due to the large number of chemical components involved. The traditional models are no longer feasible, as they converge through a large number of semi-implicit iterations, where the cost of a single iteration

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<sup>a</sup>Ph.D. candidate at the von Karman Institute for Fluid Dynamics, Stwg.-op-Waterloo 72, 1640 St. Genesius-Rode, Belgium. Tel.: 0032/2/359/96/11—Fax: 0032/2/359/96/00—E-mail: vdabeele@vki.ac.be. The research of the first author was financed by a grant of the Flemish Institute for the Promotion of Scientific-Technological Research in the Industry (IWT).

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can be of the order of several minutes on a modern workstation. Instead, a fully implicit model should be used to limit the number of iterations to a minimum.

As a step in the development of a model of the 1.2 MW 'Plasmatron' wind tunnel at the von Karman institute,<sup>6</sup> this contribution presents a numerical model of multi-component inductive plasmas (e.g. air) under chemical non-equilibrium. From a physical point of view, one could remark that considering chemical non-equilibrium alone is not enough—in fact, thermal non-equilibrium effects may well be more important. From a numerical point of view, however, difficulties are encountered mainly in the modeling of chemical non-equilibrium. The present contribution focuses entirely on these numerical problems, while setting aside aspects connected to thermal non-equilibrium.

### PHYSICO-CHEMICAL MODELING OF MULTI-COMPONENT PLASMAS

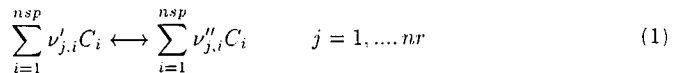
A finite mixture of  $nsp$  species is used to represent the plasma. For instance, an air inductive plasma may be represented by an eleven-component mixture:<sup>7</sup>

- Neutral species:  $O_2$ ,  $N_2$ ,  $O$ ,  $N$ ,  $NO$
- Charged species:  $NO^+$ ,  $O^+$ ,  $N^+$ ,  $O_2^+$ ,  $N_2^+$ ,  $e^-$

The plasma thermodynamic properties are computed from statistical mechanics and the plasma transport properties are computed with the method of Chapman and Enskog.<sup>8</sup>

#### Chemical kinetics model

A model of  $nr$  reactions is adopted to represent the most important chemical processes taking place in the plasma. Each reaction has a corresponding stoichiometric relation:



where  $\nu'$  and  $\nu''$  are the forward and backward stoichiometric coefficients and the molar density of a species  $i$  is written  $C_i$ . For the case of eleven-species air, the reaction model proposed by Dunn and Kang<sup>9</sup> is used here. As an alternative, the well-known model of Park can be found in the same reference.

The net mass production  $\omega_i$  of a species  $i$  is given by:

$$\omega_i = M_i \sum_{j=1}^{nr} (\nu'_{j,i} - \nu''_{j,i}) \left\{ k_{f,j} \prod_{k=1}^{nsp} C_k^{\nu'_{j,k}} - k_{b,j} \prod_{k=1}^{nsp} C_k^{\nu''_{j,k}} \right\} \quad (2)$$

where  $M_i$  stands for the molar mass of the species  $i$ . The forward reaction rate constants  $k_{f,j}$  are modeled by Arrhenius-type source terms.<sup>9</sup> The backward reaction rate constants  $k_{b,j}$  are taken such that the equilibrium composition is found back when chemical reactions are fully equilibrated:

$$k_{b,j} = k_{f,j} / K_{c,j} \quad (3)$$

Here, the equilibrium constant  $K_{c,j}$  is computed from statistical mechanics.<sup>8</sup>

#### Multi-Component Diffusion Model

The species mass diffusion fluxes  $\vec{J}_i$  are governed by the Stefan-Maxwell relations. For inductive plasma flows, contributions of thermal and pressure diffusion may be neglected.<sup>8</sup> Under conditions of thermal equilibrium, the following formalism is then obtained:<sup>10,11</sup>

$$\sum_{j=1}^{nsp} \frac{x_i M_i \vec{J}_j - x_j M_j \vec{J}_i}{C M_i M_j D_{ij}} = \nabla x_i - \frac{e_i x_i \vec{E}_A}{k_B T} \quad i = 1, \dots, nsp \quad (4)$$

where  $x_i$  represents the mole fraction of the species  $i$  and  $C$  represents the mixture molar density.  $D_{ij}$  stands for the binary diffusion coefficient between species  $i$  and  $j$ , for now still in the first Chapman-Enskog approximation.<sup>†</sup> The electrical charge of a single particle  $i$  is written  $e_i$  and  $\vec{E}_A$  stands for the ambipolar electric field.<sup>8, 12</sup>

Only  $nsp - 1$  of the above relations are linearly independent. To obtain a fully determined system, the constraint that mass diffusion fluxes sum up to zero needs to be added:

$$\sum_{i=1}^{nsp} J_i = 0 \tag{5}$$

An additional relation is needed to specify  $\vec{E}_A$ . It is assumed that ions and electrons diffuse together such that the net electric current in the radial and axial direction is zero, a situation commonly referred to as ambipolar diffusion:<sup>12</sup>

$$N_A \sum_{i=1}^{nsp} e_i J_i / M_i = 0 \tag{6}$$

where  $N_A$  is Avogadro's number.

For a three-species plasma (e.g. argon), one easily derives a Fick-type law for the ion-diffusion flux from (4, 5, 6).<sup>12</sup> For a general multi-component plasma, the full formalism needs to be solved numerically. Ramshaw and Chang<sup>11</sup> give an approximate expression for the ambipolar field. The electron diffusion flux no longer follows from its corresponding Stefan-Maxwell relation but is obtained from the ambipolar constraint instead. An approximate technique may be used to solve the remaining multi-component system of equations.<sup>13</sup>

As an alternative, it is suggested here to generalize a technique proposed by Sutton and Gnoffo<sup>14</sup> for neutral mixtures. From an intermediate value of the diffusion fluxes  $\vec{J}_i^n$ , updated fluxes  $\vec{J}_i^{n*}$  are obtained by a convenient rewriting of the relations (4):<sup>14</sup>

$$\vec{J}_i^{n*} = -\frac{\rho M_i}{M} D_i \left\{ \nabla x_i - \frac{e_i x_i \vec{E}_A^n}{k_B T} \right\} + x_i M_i D_i \sum_{j=1}^{nsp} \frac{\vec{J}_j^n}{M_j D_{ij}} \tag{7}$$

where  $M$  stands for the mixture molar mass  $\sum_{i=1}^{nsp} x_i M_i$  and the 'effective' diffusion coefficient  $D_i$  is defined:

$$D_i = \left( \sum_{j=1}^{nsp} x_j / D_{ij} \right)^{-1} \tag{8}$$

$\vec{E}_A^n$  is set such that the updated diffusion fluxes  $\vec{J}_i^{n*}$  satisfy the ambipolar constraint (6). The complete update is obtained upon adding a small correction to guarantee that (5) is satisfied as well.<sup>13, 14</sup>

$$\vec{J}_i^{n+1} = \vec{J}_i^{n*} - y_i \sum_{j=1}^{nsp} \vec{J}_j^{n*} \tag{9}$$

For slightly ionised mixtures, the iterative process (7-9) needs to be repeated about ten times to converge.<sup>14</sup> For partially ionized mixtures, it was found that up to forty iterations were sometimes needed. However, performing just three iterations already yields solutions correct up to a few percents; the mixture rules used for the modeling of other transport properties (viscosity, heavy particle thermal conductivity) are of similar accuracy. Though approximate, the obtained solution is fully consistent with the constraints (5) and (6).

<sup>†</sup>NOTE: It should be stressed that a first approximation works well only for neutral mixtures. For ionized mixtures, higher order corrections should be applied to all collisions involving one or two charged particles.<sup>10</sup>

GOVERNING EQUATIONS

*Modeling Assumptions*

The torch is modeled by an axisymmetric configuration (figure 1). To this end, the inductor surrounding the torch is replaced by a set of parallel current-carrying rings; for simplicity, these rings are supposed to be infinitely thin. The physics of the problem may be described by the axisymmetric equations of magnetohydrodynamics, where the flow variables are averaged out in time and the electromagnetic variables are pure Fourier modes at the torch operating frequency  $f$ .<sup>15,16</sup>

As a working assumption, the plasma is supposed to be in thermal equilibrium. Given its low Reynolds number ( $\sim 500$ ), the flow is considered laminar. An investigation of experimental results obtained in atmospheric arc plasmas<sup>17</sup> suggests that below 10 000 K, radiative losses are small in air plasmas. Moreover, radiative losses are known to decrease strongly with pressure.<sup>18</sup> As the final aim of this work is a simulation of air inductive plasmas at subatmospheric pressures, radiative effects are entirely neglected.

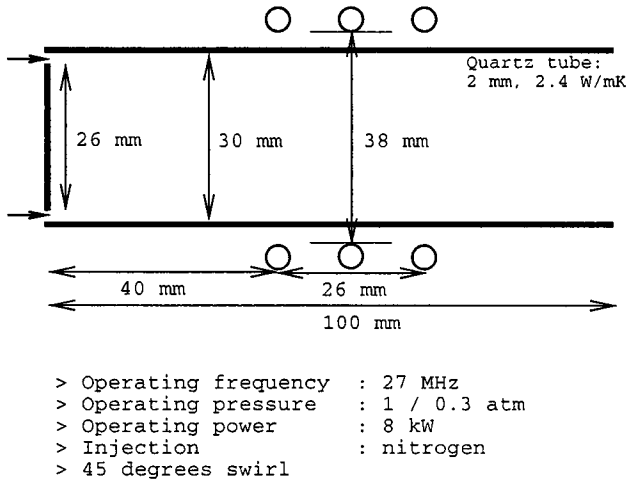


FIGURE 1 Torch geometry and operating conditions used in the numerical simulations (VKI mini-torch)

*MHD Induction Equation*

All electromagnetic field phenomena may be expressed in terms of the electric field, which is directed purely in the azimuthal direction:

$$\vec{E} = E \exp(i 2\pi f t) \vec{I}_\theta \tag{10}$$

where the electric field amplitude  $E$  needs to be a complex variable to take into account phase differences inside the plasma. The electric field is split in a part  $E_V$  induced by the outer coil rings in the absence of the plasma and a part  $E_P$  induced by the plasma:

$$E = E_V + E_P \tag{11}$$

The induced electric field is obtained by solving the following fully resistive induction equation on a domain which extends into the space surrounding the torch:<sup>19</sup>

$$\frac{\partial^2 E_P}{\partial z^2} + \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial E_P}{\partial r} \right) - \frac{1}{r^2} E_P - i 2\pi \mu_0 \sigma f (E_P + E_V) = 0 \tag{12}$$

where  $\mu_0$  stands for the vacuum magnetic permeability and  $\sigma$  represents the plasma electrical conductivity. By solving for the induced electric field  $E_P$  rather than for  $E$ , singularities near the infinitely thin coil rings are avoided.

*Time-Averaged Flow Field Equations*

The time-averaged flow field equations read:<sup>19</sup>

$$\nabla \cdot (\rho \bar{u}) = 0 \quad (13)$$

$$\nabla \cdot (\rho \bar{u} y_i + \bar{J}_i) = \omega_i \quad (14)$$

$$\nabla \cdot (\rho \bar{u} \bar{u} + p \bar{I} - \bar{\tau}) = \bar{F}_L \quad (15)$$

$$\nabla \cdot \left( \rho \bar{u} h + \sum_{i=1}^{nsp} h_i \bar{J}_i \right) = \nabla \cdot (\Lambda \nabla T) + P_J \quad (16)$$

where  $\bar{I}$  is the unit tensor and  $\bar{\tau}$  stands for the tensor of viscous stresses. A precise expression for the time-averaged Lorentz force  $\bar{F}_L$  and the Joule heating source term  $P_J$  in terms of the electric field amplitude  $E$  has been given by e.g. Chen and Pfender.<sup>20</sup> The plasma enthalpy per unit mass is indicated by  $h$ .  $\Lambda$  represents the *frozen* thermal conductivity; the species internal enthalpies per unit mass are written  $h_i$ . The effect of viscous dissipation may be neglected in the energy equation.

Provided that the global and species continuity equations (13-14) are discretized with care, the following property holds exactly on the discrete level:

$$\rho \bar{u} \cdot \nabla \left\{ \sum_{i=1}^{nsp} y_i - 1 \right\} = 0 \quad (17)$$

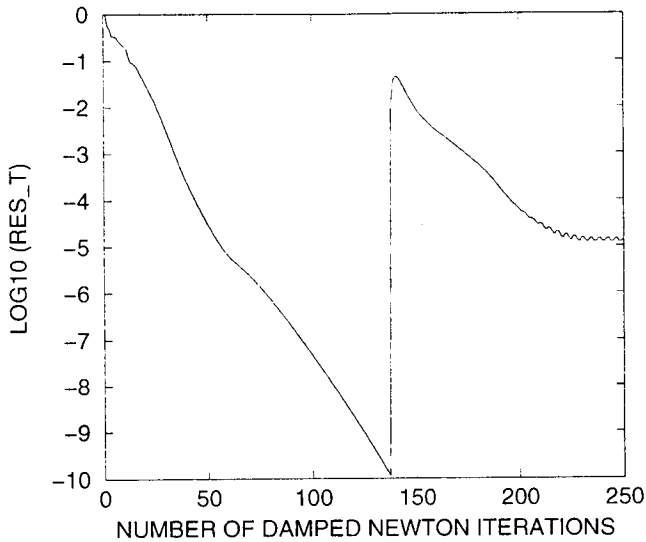
If all mass fractions sum properly to one at the inlet, it follows that their sum remains equal to one throughout the entire domain. Similarly, making use of (6), one finds that

$$\rho \bar{u} \cdot \nabla \left\{ N_A \sum_{i=1}^{nsp} e_i y_i / M_i \right\} = 0 \quad (18)$$

If the plasma is quasi-neutral at the inlet, it will remain so throughout the entire domain.

*Discretization and iterative solution procedure*

The governing equations (12-16) are discretized in a second order accurate 'TVD' manner<sup>21</sup> on a structured mesh. The low Mach number flow field is solved with a pressure-stabilized technique on a collocated mesh. The induced electric field is discretized on a far field mesh, which extends into the space beyond the torch but coincides with the interior mesh inside the torch.<sup>22</sup> Whereas almost all other models use a computationally expensive integral boundary condition for the electric field at the edges of the interior domain (see e.g. McKelliget<sup>16</sup>), the far field approach has the advantage of leading to sparse linear systems, to which modern convergence acceleration techniques are applicable. The discretized system of equations is solved with a fully coupled damped Newton method. Krylov subspace methods are used to efficiently solve the linear systems arising from the Newton linearization.<sup>19</sup>



**FIGURE 2** Convergence history found for the calculation (0.3 atm). The equilibrium solution is computed first and then used as an initial guess for the non-equilibrium model

## RESULTS

At the time of writing, some stability problems still tend to occur when simulating eleven-species air inductive plasmas. As the behaviour of air and nitrogen plasmas is known to be very similar,<sup>23</sup> a preliminary evaluation of the model is given by calculating a five-species nitrogen plasma instead. The torch geometry and operating conditions used for the simulations are given on figure 1. Simulations were performed on a fine inner mesh of 86 by 40 cells and a far field mesh of 100 by 70 cells. Results are presented for operating pressures of 1 and 0.3 atm.

Figure 2 shows the convergence history found for the calculations. The equilibrium solution is first computed and then used as an initial guess for the non-equilibrium model. The equilibrium model converges 10 orders of magnitude in about 150 iterations. Half an hour of CPU time is needed for this on a modern (235 MFlops) workstation. The non-equilibrium model converges with roughly the same speed and with similar CPU times per iteration: 65 MByte of memory is needed. It is seen that the non-equilibrium code stagnates after converging about five orders of magnitude. Though this problem still needs to be addressed, the convergence of the non-equilibrium model is already very satisfactory.

On figure 3, computed equilibrium and non-equilibrium temperature fields are compared for an operating pressure of 1 atm. The solution is seen to be close to equilibrium. Surprisingly, differences are seen in the high temperature regions, where the non-equilibrium model predicts a peak temperature of about 1000 K lower than found with the equilibrium model. On figure 4, computed equilibrium and non-equilibrium temperature fields are compared for an operating pressure of 0.3 atm. Temperatures predicted by the non-equilibrium model are significantly lower than indicated by the equilibrium result. In particular, it is noticed that peak temperatures shift away from the torch axis. In general, it is observed that non-equilibrium effects become more and more pronounced as pressure decreases, a conclusion drawn earlier by Mostaghimi *et al.* in their simulations of argon inductive plasmas under non-equilibrium.<sup>3</sup>

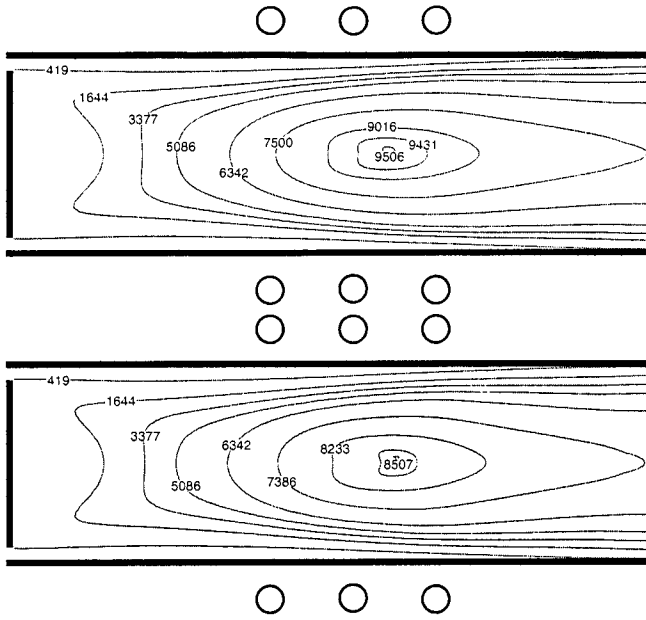


FIGURE 3 Computed temperature fields at an operating pressure of 1 atm. Upper: equilibrium solution; lower: non-equilibrium solution (2<sup>nd</sup> order accurate, 5 orders of magnitude converged)

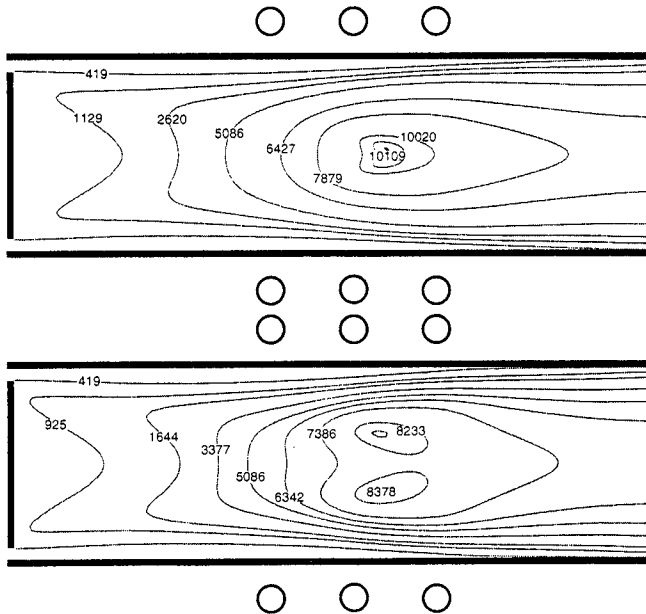


FIGURE 4 Computed temperature fields at an operating pressure of 0.3 atm. Upper: equilibrium solution; lower: non-equilibrium solution (2<sup>nd</sup> order accurate, 5 orders of magnitude converged)

## CONCLUSION

By combining an elegant physical model with a powerful iterative solution strategy, it has been shown that multi-component inductive plasma flows under chemical non-equilibrium may be computed in reasonable CPU times on fine meshes. Once the remaining convergence and stability problems (for air plasmas) are solved, thermal non-equilibrium effects will be brought into the code. A three temperature model (heavy particle, vibrational and electron temperature) will be considered as a first step. The physico-chemical modeling of air under thermal non-equilibrium will undoubtedly be a challenging task. Fortunately however, no substantial numerical difficulties are foreseen and CPU times should remain reasonable.

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