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AN APPROXIMATE QUANTITATIVE ANALYSIS OF NON-EQUILIBRIUM PLASMA TRANSPORT FOR HIGH DENSITY PLASMAS

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For the evaluation of transport in high density plasmas numerical models have been developed in which simultaneously the conservation laws for mass, momentum and energy are solved. For high density plasmas, which are not too far from equilibrium the commonly used thermodynamic quantities are, electron temperature T_e, electron density n_e, heavy particle temperature and neutral density (or pressure). In this contribution an alternative formulation is described in which the plasma state is described by electron density n_e and total pressure p and two non-equilibrium parameters: the deviation from Saha equilibrium of the neutral ground state ($\delta b_1 = n_1/n_1^{Saha}-1$) and the deviation from thermal equilibrium between electrons and heavy particles $\delta\Theta = 1-T_h/T_e$. The latter two parameters are zero in local thermodynamic equilibrium.
The advantage of this formulation is, that the transport coefficients and radiative properties can be reformulated as function of mainly n_e (at constant pressure), as the influences of non zero δb_1 and $\delta\Theta$ are small or can be explicitly given. As a result a simpler approximate formulation of the transport problem can be obtained. As an example the procedure is illustrated for atmospheric argon plasmas and for one aspect a comparison is made with work from e.g. E. Pfender.

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1. INTRODUCTION

Thermal plasmas with high electron density and relatively high pressure gain more and more importance as a highly reactive medium for plasma processing [1]. The high power density and thus high electron densities lead to high radical and ion densities. The high specific reactivity and the large flow capacity bring with it the possibility to treat relatively large amounts of gas and allows for fast and thus economic processing. In many applications use is made of thermal plasmas: plasma spraying [2], chemical conversion [3], deposition of crystalline [4, 5, 6] and amorphous materials [6, 7,] etching [8], surface modification [9], electric power switching and, more recently, also waste destruction and effluent gas treatment [10].

It is evident that for optimization modeling with experimental verification is needed. Several models have been developed in the past, first models based on LTE assumptions of the plasma state, later taking also non-equilibrium effects into account [1, 11, 12, 13, 14, 15]. In these numerical models basically the momentum conservation balance is solved with, depending on assumptions on LTE, simultaneously solution of the mass and energy balances. Emphasis in studies of non-equilibrium effects has been on atomic plasmas in which two deviations from LTE can be considered. First the electron temperature T_e may deviate from the heavy particle temperature T_h (the ion and neutral temperature) and the ionization composition may deviate from Saha-equilibrium. For the latter usually the pLSE (partial local Saha equilibrium) assumption can be used, in which only the ground state of the neutrals deviate from Saha-Boltzmann equilibrium with the continuum at the prevailing temperature. Similarly, in molecular plasmas the density of molecules, radicals and atoms may deviate with respect to equilibrium composition.

As said, in the past commonly use was made of LTE composition for the calculation of the various transport coefficients, required to solve the coupled set of conservation laws. More recently investigations are taken chemical kinetics into account. This however is a cumbersome task, as many kinetic reactions are possible and an elaborate set of kinetic equations are required.
In this contribution it is investigated in how far such a detail can be avoided and still to take non-equilibrium effect into account in an approximate way. We will do this for the most simple case: an atomic plasma in a noble gas for which two types of non-equilibrium exist: the electron temperature may deviate from the heavy particle temperature and

the ion (and electron) density is not in equilibrium with the neutral ground state. We will however assume that the excited states are in equilibrium with the continuum i.e. the pLSE state is assumed. It should be noted however, that for atomic plasmas in excitation saturation [16] (i.e. electron (de)excitation between all excited states is more important than radiative transitions) the description is justified with only minor adaptations.

The principle route to arrive at such an approximate solution is the following: First we will use the electron density as the primary thermodynamic variable in stead of T_e. The reasoning for this is simple; as can be seen in fig. 1 for a LTE plasma in argon at 1 bar, n_e varies over more than two orders of magnitude, whereas the corresponding T_e variation in LTE varies only a factor of 3. At specified pressure the electron density is the best thermodynamic quantity to compare models with experiments. Hence the LTE state will be defined by pressure p and electron density n_e; T_e^{LTE} is then a dependent plasma parameter which can be derived from n_e and p [17-19].
Second, the deviation from LTE will be defined by two non-equilibirum parameters

$$\delta b_1 = b_1 - 1 \equiv \frac{n_1}{n_1^{Saha}} - 1 \qquad , \delta\Theta = 1 - \frac{T_h}{T_e} \qquad (1)$$

in which b_1 is the so-called overpopulation of the neutral ground state at the actual values of n_e and T_e. We mention, that the assumption of pLSE implies that all excited states are in Saha equilibrium with the continuum, i.e. the overpopulation factors b_p=1 for main quantum number p≥2.
The other non-equilibrium parameter is $\delta\Theta = 1 - T_h/T_e$. It describes the deviation from thermal equilibrium between electron and heavy particles.
Hence, we will use p, n_e, δb_1, $\delta\theta$ to describe the plasma state. The advantages of this set instead of the usual parameter set p, T_e, n_e, T_h are obvious: the parameters p, and n_e describe the pLSE state already to a large extent and the last two parameters are equal to zero in equilibrium. We will show that most transport terms depend mainly on the first two parameters and, if at all, only weakly on the non-equilibrium parameters [15, 21]. Only the ionization terms in mass and energy balances depend linearly on δb_1. Further $\delta\theta$ is only non-zero at lower electron densities. We note
also that the electron density is a good measurable quantity by various unambiguous methods which facilitates comparison with experiment.

The third aspect of the proposed approach is to distinguish active (ionizing) plasmas and passive recombining plasmas. In the active part of the plasma the power is dissipated; because of the high ionization ratio: n_e/n_1 the electric conductivity is Coulomb collision dominated. The passive part of the plasma is governed by particle and heat diffusion and usually the contributions of ionization and Ohmic dissipation are small in this region. Recombination plays a rôle there only in the presence of molecules.

From various detailed experimental observations it has been established for 1 atm. argon plasmas that the pLSE state can be approximately described by a (n_e, T_e) relation, slightly different from the LTE one, as will be described in the next section.

2. THE DEVIATION FROM EQUILIBRIUM IN ARGON PLASMAS AT ATMOSPHERIC PRESSURE

Several investigations have been performed to measure the deviations from equilibrium. In this section we will concentrate on δb_1 which describes the deviation from Saha-Boltzmann equilibrium. Very precise measurements are needed to verify the small deviations from equilibrium. Here we will use the findings by Rosado [18] and Timmermans [19] obtained from end on measurements of stationary, non flowing atmospheric plasmas in wall stabilized arcs. End on wavelength resolved measurements of emission and absorption of various argon lines and confirmed by interferometry at two wavelengths yielded values for n_e and T_e. The result has also been given in fig.1[a] as the pLTE (pLSE and $\delta\Theta$=0) labeled curve. The source function method [19, 20] was used to obtain T_e, whereas n_e followed from the absolute emission strength of the upper level of the observed atomic argon line. It has been shown [19] that small errors in e.g. the calibration procedure changed both T_e and n_e in such a manner that the resulting pLTE relation was hardly affected. Moreover the electron density measured at axis with this method was verified by model independent methods such as Stark broadening, continuum emission and by two wavelength interferometry. For the current range used (40 A- 200 A in 5 mm cascade arc) the plasma is in the ionizing phase up to close to the wall. Hence the result relates to this phase of the plasma. From the resulting n_e and T_e the actual ground state density n_1 can be calculated assuming $T_h/T_e = 1$, ($\delta\Theta$=0). The result, together with the parameter δb_1,

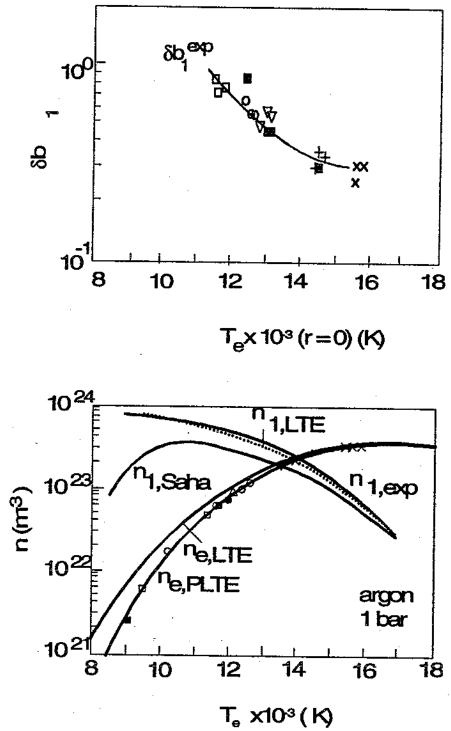

Figure 1[a] Electron density and ground state neutral density as a function of temperature(T_e=T_h) and non-equilibrium parameter δb_1 for stationary argon wall stabilized arc at 1 bar. Also shown are n_1^{LTE} (at 1 bar) and n_1^{Saha}; from [19]

which describes the deviation from equilibrium is also shown in fig. 1[a]. Further are shown n_1^{LTE}, the value of the ground state density in full LTE at 1 bar and n_1^{Saha}, the value of n_1 in Saha equilibrium with measured n_e and T_e. In fig. 1b the same quantities are shown as function of n_e, again compared with LTE and Saha values for n_1.

It appears from these results that approximately a (non-equilibrium) pLTE state can be defined for which the following approximate relation holds:

$$\delta b_1 \cong C_1 + C_2 / n_e \simeq 0.3 + 10^{22} / n_e \qquad (2)$$

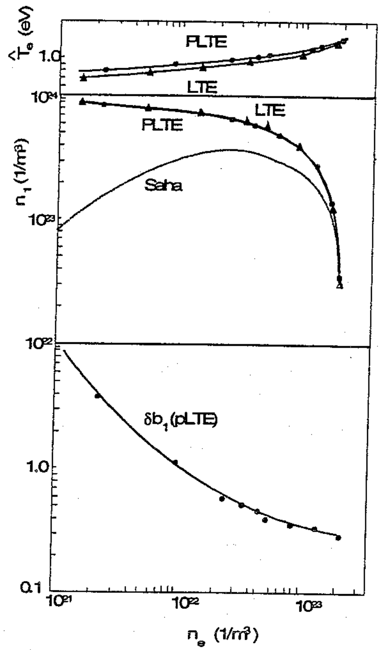

Figure 1^b As function of n_e

The general form of this relation can be explained by the two loss contributions to the mass balance: one from (radiative) recombination (C_1) and one from diffusive losses (C_2/n_e). It appears that the numerical expression holds approximately for 1 atm. pressure plasmas rather independent on radial position and on arc conditions. This result was somewhat unexpected as contributions from diffusion and radiative recombination depend on actual position in the plasma and on plasma conditions. Of course the relation should be seen as a first approximation. Still as the sensitivity to the precise value of δb_1 is not large it proves to be usefull as a first order non-equilibrium approach for argon plasmas at 1 bar.

For lower densities and temperatures (and thus power density) as in ICP's it is more difficult to obtain results with the accuracy obtained in the above described experiments. Usually radial densities have to be obtained from lateral observations through the use of Abel inversion. Still the results from these experiments [22, 23] confirm roughly the findings described above. These experiments feature also larger volumes where the plasma is recombining and where also the passive state can be observed.

Here usually T_h deviates from T_e and $\delta\Theta \neq 0$ has to be taken into account. However, in this case we can use the combined factor $b_1 \cdot T_h / T_e$ as one non-equilibrium parameter and still obtain a simplification of the non-equilibrium treatment. Such a first order simplification is based on the following observation. In the equations to be described and in the transport properties the heavy particle density $n_e + n_1 = n_h$ and the product $n_1 n_e$ play a role; n_1 is the neutral ground state density. At the low electron densities, where $\delta\Theta \neq 0$, the electron density is small compared to the (ground state) neutral density. Then Dalton's law reads

$$p = (n_1 + n_e)kT_h + n_e kT_e \simeq n_1 kT_h \simeq n_1^s kT_e b_1 T_h / T_e \qquad (3)$$

The Saha density times n_1s the electron temperature is an expression in n_e and T_e only. In the present formulation T_e depends formally on n_e, p as main parameters and on δb_1 and $\delta\Theta$ as non-equilibrium parameters. However, the latter dependences are weak and usually T_e is not too far from the equilibrium value, and the small deviation can again be expressed in the product b_1 T_h/T_e. Commonly the deviation from excitation equilibrium is more significant (δb_1) than the deviation from thermal equilibrium ($\delta\Theta$). In this explorative contribution we will hence focus on the deviation from excitation/ionization equilibrium and thus treat the higher electron density case for which $\delta\Theta \simeq 0$.

3. THE CONSERVATION LAWS

In this contribution we will limit ourselves to a summary of the conservation laws for mass, momentum and energy and only indicate where specific assumptions are made. We will use the intrinsic formulation of the momentum and energy balances, i.e. formulations in which the mass and momentum balances are respectively substracted from the extrinsic formulations. Further, we will distinguish between electrons and heavy particles (i.e. ions and atoms) and we will assume that the mass density in the excited states is small compared to the mass in the ground states.

Then it follows for a stationary mono-atomic plasma like argon, with -h- for heavy particles and -e- for electrons (**u** is the averaged flow velocity):

mass -h- $\nabla . n_h \mathbf{u} = 0$ (4)

Navier Stokes total mom. $n_h m_h(\mathbf{u}.\nabla)\,\mathbf{u} + \nabla(p_h + p_e) + \nabla{:}\pi_h = \mathbf{j} \times \mathbf{B}$ (5)

energy-h- $\frac{5}{2} n_h\, \mathbf{u}.\nabla kT_h - \mathbf{u}.\nabla p_h = Q_{eh} - \pi_h : \nabla \mathbf{u} - \nabla.\mathbf{q}_h$ (6)

These equations describe the heavy particle dynamics, i.e. the mass, motion and energetics. It can be observed that eqs. (4) to (6) are very similar to those of normal hydrodynamics for the heavy particles, apart from the following:

– the pressure term in the momentum balance includes the electron pressure besides the heavy particle (h) pressure:

Dalton $p = p_h + p_e = p_h(1 + n_e T_e / n_h T_h)$ (7)

in which n_e/n_h is the ionization degree. For the usually relevant situation of $10^{-3} << n_e/n_h << 1$ for the active part of the plasma, this addition is not large.

– for the indicated range of n_e/n_h the viscosity tensor π_h is strongly affected by the presence of ions (through i-o, o-i and i-i collisions); the electron viscosity contribution can be neglected in all cases, compared to ∇p_e. For the low ionization degree recombining plasma, e.g. at the wall, the changes in viscosity compared with normal hydrodynamics are small.

– the energy equation for heavy particles contains a source term Q_{eh}, which for the active part is mainly e-i collisions and for the passive part mainly e-o collisions dominated. Note, that this term is important in the active part, where the difference between T_e and T_h is small! There this equation can often be replaced by $T_e = T_h$. In the cold passive part the difference can be large, but the importance of this term is small.

– the viscous heating term (which we will not work out in this paper).

The conservation laws for electrons read:

mass -e- $\delta b_1 K_1 n_1^{Saha} n_e - k_{+1}\Lambda_{+1} n_e^2 + \nabla D^{amb} \nabla n_e - \nabla.n_e \mathbf{u} = 0$ (8)

mom. -e-, Ohm $\mathbf{j} = \sigma(\mathbf{E} + \frac{\nabla p_e}{n_e e} - 0.71 \frac{\nabla T_e}{n_e e})$ (9)

(total) energy $-\frac{5}{2}\left[n_h \mathbf{u} \cdot \nabla kT_h + n_e \mathbf{u} \cdot \nabla kT_e\right] + \mathbf{u} \cdot \nabla p$

$$+\frac{3}{2}\mathbf{j}\cdot\nabla\frac{kT_e}{e}-\frac{kT_e}{n_e e}\mathbf{j}\cdot\nabla n_e+\sigma E^2=$$
$$=K_1 n_1^{Saha}\delta b_1 n_e(E_1^+ +\frac{5}{2}kT_e)-n_e^2 k_{+1}\Lambda_{+1}kT_e+Q_{rad}+Q_{visch}$$
$$-\nabla\cdot\kappa_e\nabla T_e-\nabla\cdot\kappa_h\nabla T \qquad (10)$$

The electron mass and energy balances contain the inelastic production terms in which K_1 is the total excitation and ionization rate [19-24]. Here the deviation from Saha equilibrium is taken into account by the factor b_1-1= δb_1. In the pLSE approximation the three particle recombination is contained in the production term. Therefore only radiative recombination to the ground state is found in the mass balance; $k_{+1}\Lambda_{+1}$ is the rate for these processes corrected for the radiation trapping by the escape factor Λ_{+1}. In the energy balance the energy loss by resonant and recombination radiation to the neutral ground state is hidden in the inelastic term, as the electrons loose energy in the excitation. Only a small negative rest term results from substraction of the mass balance times $^5/_2$ kT_e from the energy balance as this substraction overcompensates with regard to the heat loss associated with diffusion (also called the expansion term).
The radiation term in the energy balance Q_{rad} = Q_{cont} + Q_{line} contains the radiative recombination to the excited states (fb) and the Bremmsstrahlung (ff) (Q_{cont}) and the line radiation to excited states (Q_{line}). The reasoning behind this formulation is, that the electrons loose energy in excitation and ionization. For the ground state this is formulated explicitly in the term with δb_1=b_1-1, where b_1 stands for upward processes and -1 comes from reverse processes from excited states and continuum. In pLSE the excited states are in equilibrium with the continuum. Hence any radiation ending in excited states has to be equilibrated by electron collisions. The energy loss in that process is equal to the energy loss in the radiative process, which can thus replace the loss in the electron energy balance. The non-equilibrium influence on Q_{rad} has been investigated by Wilbers [21] and De Regt [25].
The balance equations have to be complemented by Maxwell equations and boundary conditions.
In table I the various plasma quantities and the required transport properties are summarized. In pLSE the plasma state is determined by 4 of the 5 parameters p, n_e, T_h, T_e, and n_1. For the active part of the plasma the best parameter set is p, n_e and two non-equilibrium parameters, δb_1 and $\delta\Theta$ (of which $\delta\Theta$ can usually be set equal to 0 ($T_e \equiv T_h$).

Independent plasma variables		Dependent plasma quantities	
flow velocity	$\mathbf{u}$	electron temperature	$T_e(n_e, p; \delta b_1, \delta\Theta)$
total pressure	p	heavy particle temperature	$T_h(n_e, p; \delta b_1, \delta\Theta)$
electron density	n_e	neutral groundstate density	$n_1(n_e, p; \delta b_1, \delta\Theta)$
deviation from Saha	δb_1	viscosity	$\pi(\mathbf{u}, n_e, p; \delta b_1, \delta\Theta)$
deviation thermal	$\delta\Theta$	electron pressure	$p_e = n_e k T_e$
		heavy particle pressure	$p_h = n_h k T_h$

K_1	Total excitation and ionization rate from the ground state.
k_{+1}	Rate for recombination to the ground state and resonance radiation including radiation trapping.
Q_{eh}	Energy transfer from electrons to heavy particles by elastic (e-i and e-o) collisions
Q_{visc}	Viscous heating (not treated in this paper).
κ_h	Heavy particle heat conductivity
κ_e	Electron heat conductivity
E_1^+	Ionization energy
σ	Electrical conductivity
$Q_{rad} = Q_{cont} + Q_{line}$	radiative losses due to continuüm radiation including recombination and line radiation to excited states.
$\mathbf{q}$	$\kappa\nabla T$, heat flux
π	viscosity tensor

Hence we have to eliminate the T_e dependence by solving the implicit relation, which follows from Dalton's law and the Saha value for the groundstate.

The balance equations are structured in coupled subsystems of equations (cf. fig.2):

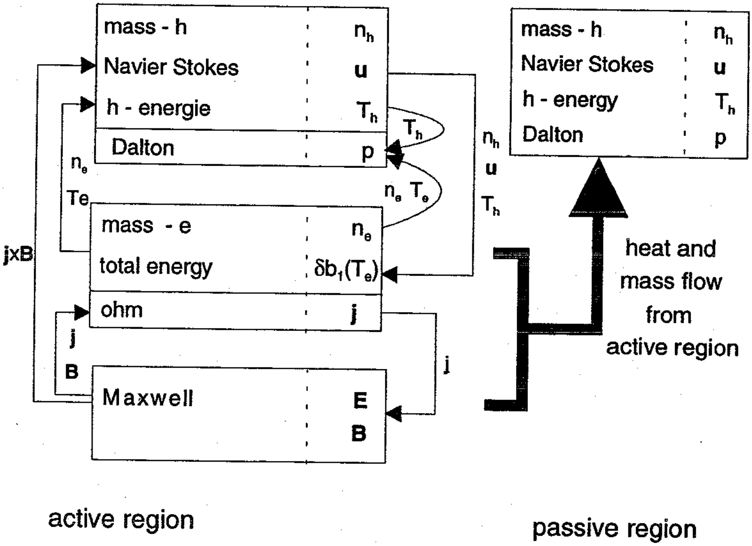

Figure 2[a] Schematic structure of balance equations problem for active and passive parts of the plasma.

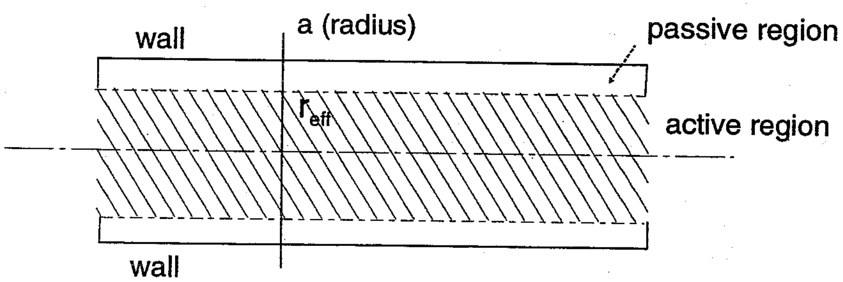

Figure 2[b] Sketch of the active and passive zones in a cylindrical plasma.

I. Navier Stokes, heavy particle mass and energy balances and Dalton's Law.
II. electron mass and total energy equations and Ohms' law.
III. Maxwell equations

These sets of equations can be solved separately if the spatial dependences of some other quantities are given and hence this system can be solved in an iterative scheme.

The first subsystem (I) deals with the flow problem. It is quite similar to a hydrodynamic system. However in addition it needs n_e and T_e from system II and if the Lorentz force is important also **j** and **B** from system III. The second subsystem (II) describes the non-equilibrium n_e and T_e evolution under influence of fields (system III) and flow (system I).

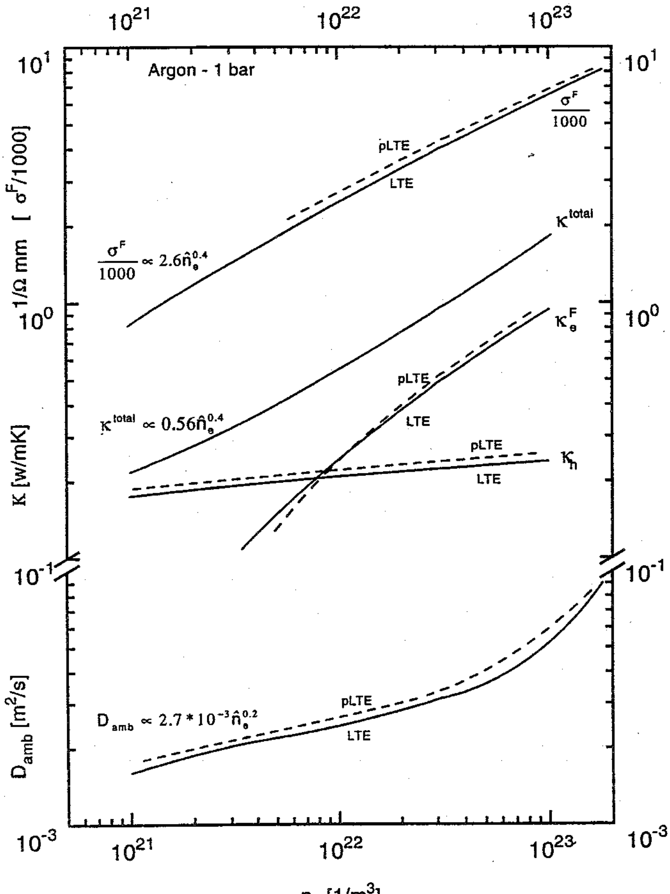

Figure 3[a] (Normalized) Transport coefficients for 1 at argon plasma as function of n_e with approximate power law representation: Frost electrical conductivity σ^F, heat conductivities, κ^{total}, κ_e^F, κ_h and ambipolar diffusion coefficient D^{amb}.

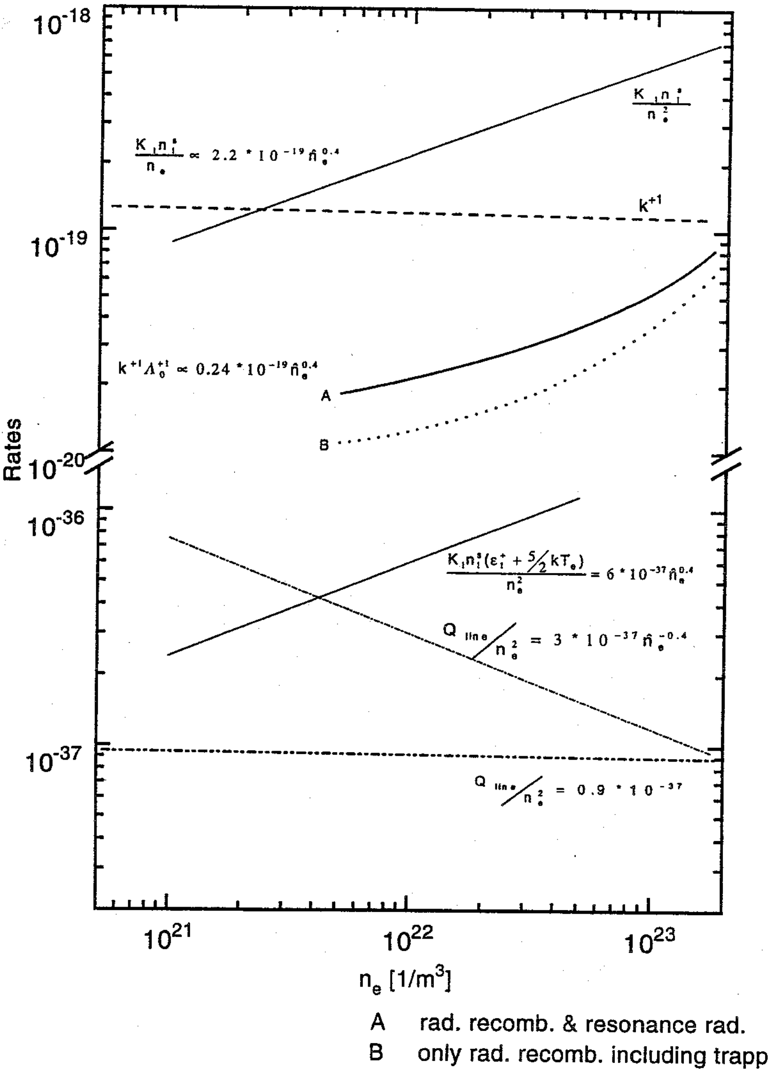

Figure 3b Similar for mass source rate $K_1 n_1^{Saha}/n_e$, radiative recombination rate k^{+1}, idem. including trapping (curve B) and also including resonance lines with trapping (curve A). Energy source rate

$$\frac{K_1 n_1^{Saha}(\varepsilon_1^+ + \frac{5}{2}kT_e)}{n_e}$$

and radiative losses of lines Q_{line}/n_e^2 and continuum Q_{cont}/n_e^2.

The above applies to the active part of the system, where the plasma i maintained and heated by the applied fields.
For the passive part of the system, surrounding the active part, one need usually only to consider the first subsystem I. However the heat an particle flows from the active part to the passive part have to be known.

For an argon plasma at 1 bar the resulting transport properties are given a function of n_e in the figure 3 for LTE ($\delta b_1=0$) and for values deviatin from LTE. For the latter the approximate expression for $\delta b_1 \simeq 0.3 \cdot 10^{22}/n_e$ is used. For given pressure (1 bar) and electron density the electroı temperature T_e and neutral density can be calculated. Then using Frost lik rules for the combined effect of Coulomb collisions and charged particl neutral collisions the transport properties can be calculated [13]. For th radiative terms a normalization to n_e^2 is appropriate and here results fron Wilbers et al.[20] can be used. Note that Wilbers has also investigated th influence of $\delta\Theta\neq 0$, which appeared to be negligeable for the highe densities range. Other transport properties as electron heat conductivit and electric conductivity are dominated by Coulomb collisions over most

Table II. Numerical expressions for the transport coefficients. Approximate $\hat{n}_e$ in units of $10^{22}/m^3$; expressions valid for $0.1 < \hat{n}_e <$ 10.

Electric conductivity:	$\sigma^F \simeq 2.6\ 10^3\ \hat{n}_e^{0.4}$	$\left(\frac{1}{Ohm\, m}\right)$
Heat conductivity:	$\kappa^{total} \simeq 0.56\, \hat{n}_e^{0.4}$	(W/mK)
Diffusion coefficient	$D_{amb} \simeq 2.7 10^{-3}\ \hat{n}_e^{0.2}$	[m^2/s]
Mass production rate-factor:	$\frac{K_1 n_1^s}{n_e} \simeq 2.2 10^{-19}\, \hat{n}_e^{0.4}$	[m^3/s]
Radiative recombination rate: (including resonant lines and trapping; curve A)	$k^{+1}\Lambda_0^{+1} \simeq 0.24 10^{-19}\, \hat{n}_e^{0.4}$	[m^3/s]
Energy loss rate factor:	$\frac{k_1 n_1^s\left(\varepsilon_1^+ + \frac{5}{2}kT_e\right)}{n_e} = 6.10^{-37}\, \hat{n}_e^{0.4}$	[Wm^3]
Line radiation loss:	$Q_{line} / n_e^2 = 3.10^{-37}\, \hat{n}_e^{-0.4}$	[Wm^3]
Continuum radiation loss:	$Q_{cont} / n_e^2 = .9 10^{-37}$	[Wm^3]

of the range. It appears that the transport coefficients as a function of n_e are not, or only weakly dependent on deviations from equilibrium and thus we can use the LTE values, which is a significant simplification, with not much loss of accuracy. We will illustrate the procedure for the calculation of a stationary non-flowing argon arc, for which detailed radially resolved measurements and full non-equilibrium modelling are available. In exploring searching the functional dependence on n_e we will emphasize first the high n_e part, as for stationary argon plasmas the active region extends almost to the wall of the arc.

4. NON-FLOWING STATIONARY WALL STABILIZED ARC IN ARGON

For non-flowing systems the heavy particle balance equations reduce to:

$$\mathbf{u} = \mathbf{0}; \quad \nabla p = \mathbf{j x B}; \qquad Q_{eh} + \nabla . \kappa_h \nabla T_h = 0 \tag{11}$$

Usually the Lorentz force can be ignored in the momentum balance and for the passive part of the plasma also the ∇p_e contribution is small. For the active part we obtain:

$$p_h + p_e = p = \text{constant} \tag{12}$$

$$Q_{eh} = \frac{3}{2} . \frac{2m_e}{m_h} n_e^2 (kT_e - kT_h) \left(k_{ei} + \frac{n_1}{n_e} k_{eo} \right) = -\nabla \kappa_h \nabla kT_h \tag{13}$$

Here k_{ei} and k_{eo} are the rates for momentum transfer from electron to ions and neutrals respectively. For argon the first (Coulomb) term overweights the second term for ionization ratios $\frac{n_e}{n_1} > 10^{-3}$. From the heavy particle heat balance we can conclude that for atmospheric argon plasma for $n_e a > 10^{20}$ /m^2 (a = plasma radius) the difference between T_e and T_h is small and that thus the non equilibrium parameter $\delta\Theta \cong 0$.
The electron balances, which are relevant for the high n_e, active part of the arc, reduce to the following eqs.:

$$\delta b_1 K_1 n_1^{Saha} n_e - k_{+1} n_e^2 + \nabla . D^{amb} \nabla n_e = 0 \tag{14}$$

$$\sigma E^2 = \delta b_1 K_1 n_1^{Saha} n_e \left(E_1^+ + \frac{5}{2} kT_e\right) - n_e^2 k_{+1} kT_e + Q_{rad} - \nabla . K_{total} \nabla T_e \quad (15)$$

$$\mathbf{j} = \sigma \mathbf{E}; \qquad j_z = \sigma E_z; \quad (16)$$

In this case the mass and energy balances describe the spatial dependences of n_e and δb_1 under influence of the electric field **E**(**r**), which is linked to the current density **j**(**r**) through eq. (16). For a cylindrical arc commonly the total current I is specified, which is linked to E by the integral of eq. (16),

$$I = E_z \int_0^a 2\pi r \sigma(r) dr \quad (16^a)$$

if it is assumed that E_z is constant over the current carrying part of the arc channel, for $r < r_{eff}$. Here the effective radius r_{eff} is defined such that there Coulomb collisions are equal to electron neutral collisions ($k_{ei} = k_{eo}$ at $r = r_{eff}$). Note, that outside this radius, which for argon plasmas is close to the channel radius a, the radial dependence of E_z is important and thus E_r is finite. For our problem this needs not to be considered, as in the outside region the dissipation contribution is small compared to the conductive terms. Of course in the active part of the discharge Ohmic dissipation is important, as it drives the system.
As mentioned the transport coefficients appearing in the mass and energy balances can be reformulated in forms which depend only on n_e at specified pressure. From the elimination of T_e, only weak dependences on δb_1 result and these dependences can be ignored without loosing much accuracy. Actually the inaccuracy which with several transport coefficients are known may be larger than the errors introduced by this procedure. By inserting the approximate simple n_e^x power laws as given in fig. 3 for the various transport coeficients the following equations result (valid for $p = 10^5$ Pa and $0.1 < \hat{n}_e < 10$; $\hat{n}_e$ in units of $10^{22}/m^3$):

$$0 = \hat{n}_e^{2.4} \delta b_1 - 1.1 \hat{n}_e^{2.4} + \frac{1}{\hat{r}} \frac{\partial}{\partial \hat{r}} \hat{r} \frac{\partial}{\partial \hat{r}} \hat{n}_e^{1.2} \quad (17)$$

$$41 \hat{n}_e^{0.4} \hat{E}^2 = \hat{n}_e^{2.4} \delta b_1 + 0.14 \hat{n}_e^2 (1 + 3.4 \hat{n}_e^{-0.4}) - \frac{15.4}{\hat{r}} \frac{\partial}{\partial \hat{r}} \hat{r} \frac{\partial \hat{n}_e^{0.5}}{\partial \hat{r}} \quad (18)$$

Here $\hat{n}_e$ is in units of $10^{22}/m^3$, $\hat{r}$ is in mm and $\hat{E}$ in kV/m.
These two equations can be solved to find the radial dependences for n_e and δb_1 for specified E_z. If the total current is specified one has to use eq. (16^a), which reformulated reads:

$$I = \hat{E} \int_0^{\hat{r}_{eff} \propto \hat{a}} 2.6 \hat{n}_e^{0.4} 2\pi \hat{r} d\hat{r} \tag{19}$$

The integration has to be extended up to the radius $\hat{r}_{eff}$, where electron neutral collisions start to dominate; there the conductivity drops faster with the decreasing $\hat{n}_e$ than represented in the numerical expression for $\sigma(\hat{n}_e)$. For argon arcs it is usually safe to replace $\hat{r}_{eff}$ by $\hat{a}$.
The non-equilibrium parameter δb_1 can be eliminated from eqs. (17) and (18) with as result the following differential equation for n_e:

$$41\hat{n}_e^{0.4}\hat{E}^2 = 0.14\hat{n}_e^2 + 0.11\hat{n}_e^{2.4} + 0.48\hat{n}_e^{1.6} - \frac{1}{\hat{r}}\frac{\partial}{\partial \hat{r}}\hat{r}\frac{\partial}{\partial \hat{r}}\left(1.1\hat{n}_e^{1.1} + 14\hat{n}_e^{0.5}\right) \tag{20}$$

If $\hat{E}$ is given and if $\hat{n}_e$(wall, $\hat{r}_{eff}$) is specified (e.g. $\hat{n}_e(\hat{r}_{eff}) \cong 10^{-1}$) then this equation can be solved.
A typical result is shown in fig. 4, in which $n_e(r)$ is shown for $\hat{E}$=1.03 k/m and $\hat{a}$=2.5mm (argon at 1 bar). The result is compared with measurements [19] and with full modeling [13]. It is evident from this figure that the present approximate modeling in which the transport coefficients are represented by a simple power law $\hat{n}_e^x$ is nearly as adequate as the full modeling. We note also that the functional form $\hat{n}_e = \hat{n}_{e0}(1 - \hat{r}^2/\hat{r}_{eff}^{\,2})^{\eta}$ may be a good approximation to the full solution for the central part of the arc. Then it follows from eq. (17) that $\delta b_1(0) = 0.11 + 5.1\eta / \hat{r}_{eff}^2 \hat{n}_{eo}^{1.2}$ which for $\eta / r_{eff}^2 \cong 0.2$ is quite close to the earlier found approximation $\delta b_1 \cong 0.3 + \frac{1}{\hat{n}_e}$
We note that the too small constant (0.11 instead of 0.3) comes from the too simple representation of the radiative recombination term, which underestimates this contribution at higher densities.

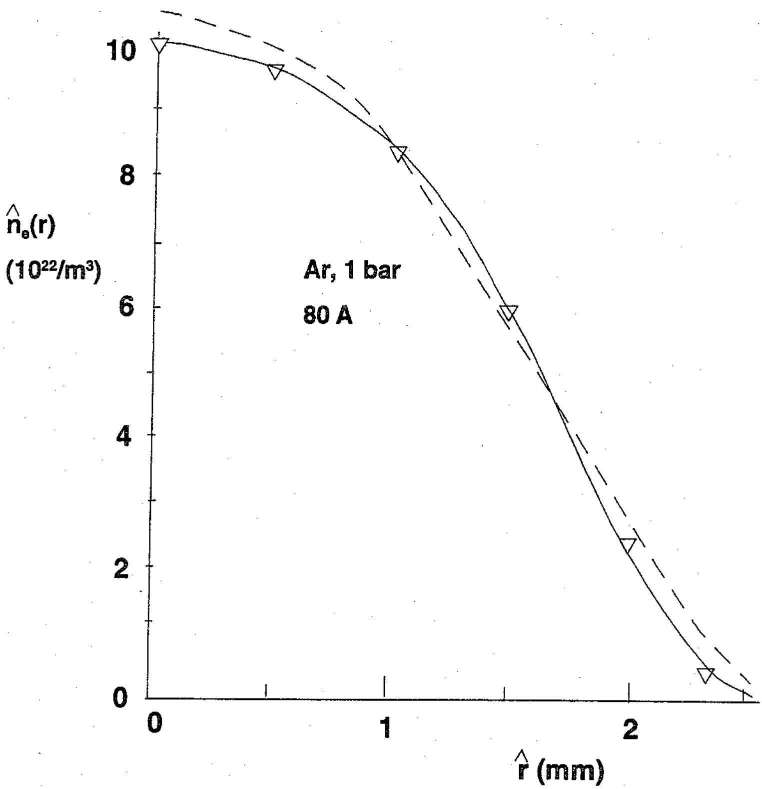

Figure 4. Calculation of $n_e(r)$ profile with the present approximate formulation (full line) and comparison with experiments (Δ, ref [12] and full nummerical modeling (dashed line, ref [13])

Hence the present modeling describes the stationary argon arc satisfactorily and could be used as a basis to tackle more difficult problems as flowing non-isobaric arcs and/or other gases. In that case the effective radius, $\hat{r}_{eff}$, may become appreciably smaller than the arc channel radius, â. In section 5 we will address shortly possible ways to obtain a value for $\hat{r}_{eff}$.

We end this section by estimating the flow velocity for which the present non-flowing approximation becomes unvalid. For that purpose we have to investigate the additional terms containing **u** in the electron mass and total energy balances eqs. (8) and (10). In the mass balance we can compare $\nabla.n_e\mathbf{u}$ with $\nabla.D^{amb}\nabla n_e$. The ratio is equal to $\frac{u_z a^2}{D^{amb}L}$, where ∇ has been replaced by 1/L respectively 1/a. As $D^{amb} \cong 10^{-3}\, m^2/s$ and $a \cong 2.5\,\mathrm{mm}$ and $L \cong 10\,\mathrm{cm}$ we find that u_z<10 m/s.

In the energy balance we have to evaluate the first five terms of eq. (10) of which the third ($\mathbf{u}.\nabla$p) and the fifth ($\frac{kT_e}{n_e e}\mathbf{j}.\nabla n_e$) are the most important.

As long as the flow is subsonic $u_z < 1/3\, c_s$, then both terms are small compared to the ohmic term **j.E**. Note, that if sonic conditions are approached, $\mathbf{u}\nabla$p becomes of the same order, where as $\frac{kT_e}{n_e e}\mathbf{j}\nabla n_e$ still remains small compared to **j.E**.

Hence for flowing plasmas with $u_z < 1/3\, c_s$ only the convective term in the mass balance has to be retained. The total pressure remains approximately constant and thus the system is approximately the same for a slowly flowing situation.

5.WALL LAYER PROFILES OF T_e AND T_h

A different approximation procedure can be followed in the periphery, the passive zone of the plasma (r_{eff}<r<a). Here electron production and current dissipation can be ignored as well as the electron contribution to the total pressure. The only important terms are diffusion in the mass balance and heavy particle heat conduction in the -h- energy balance. In other words, transport of mass and heat produced in the center of the discharge are the only processes to be considered in the passive plasma close to the wall.

For non flowing systems the following equations result:

$$\nabla.D^{amb}\nabla n_e = 0; \qquad \nabla.\kappa_h \nabla T_h = 0$$

These equations state, that the electron mass flow and the heavy particle heat flow are constant for radii r_{eff}<r<a. The constants appearing in these equations can be found by applying Gauss theorem on eq. (14) and eq. (13) respectively (here for cylindrical geometry):

$$-2\pi r_{eff} D^{amb} \left.\frac{dn_e}{dr}\right|_{r=r_{eff}} = \int_0^{r_{eff}} \left(\delta b_1 K_1 n_1^{Saha} n_e - k_{+1}\Lambda_{+1} n_e^2\right) 2\pi r dr \qquad (21)$$

$$-2\pi r_{eff} \kappa_h \left.\frac{dT_h}{dr}\right|_{r=r_{eff}} = \int_0^{r_{eff}} \frac{3m_e}{m_h} n_e^2 k(T_e - T_h) k_{ei} 2\pi r dr \qquad (22)$$

Equations (21) and (22), valid for atomic plasmas, state simply that the produced electron mass and the produced not-radiated power have to be carried through the boundary layer. Note, that in the boundary layer $T_e \neq T_h$ and that D^{amb} will be slightly different because of finite δΘ, we will ignore this in the consideration of the mass balance eq. (21), which rewritten in n_e-dependence is ($\hat{n}_e$ in $10^{22}/m^3$):

$$-2.3\cdot 10^{19} \left.\frac{d\hat{n}_e^{1.2}}{dr}\right|_{r\geq r_{eff}} 2\pi r = \int_0^{r_{eff}} \left[2.2\cdot 10^{25}\delta b_1 \hat{n}_e^{2.4} - 0.24\cdot 10^{25}\hat{n}_e^{2.4}\right] 2\pi r dr$$

(23)

If $\delta b_1 \cong 0.11 + \frac{1}{\hat{n}_e^{1.2}}$ we find (with $\hat{r}$ in mm):

$$-\left.\frac{d\hat{n}_e^{1.2}}{d\hat{r}}\right|_{r\geq r_{eff}} = \hat{n}_e^{1.2}\frac{r_{eff}}{4}$$

$$\hat{a} - \hat{r}_{eff} = \frac{4}{\hat{r}_{eff}}\left(\frac{\hat{n}_e(\hat{r}_{eff})}{\hat{n}_{eo}}\right)^{1.2} \qquad (24)$$

Since the wall layer thickness is small ($\hat{a} \cong \hat{r}_{eff}$), we will find a weak gradient for low currents (low $\hat{n}_{eo}$) and a moderate gradient at higher currents (high $\hat{n}_{eo}$). Since electron heat conduction is relatively a stronger process than electron diffusion the electron temperature gradient is relatively small and T$_e$ remains relatively close to corresponding (p)LTE-value, which for 1 bar and $n_e \cong 10^{21}/m^3$ is approximately 9000 K. This is exactly which E.Pfender and his co-workers found in the probe measurements evaluation [26]. He finds also that the boundary layer thickness decreases with increasing arc current (from 0.4mm at 100A to

<0.1mm for currents above 200A, in a 10mm diameter arc). This finding is in agreement with eq. (24).

By evaluating the heavy particle heat balance one finds that coupling between electrons and heavy particles becomes weaker for electron densities below $10^{22}/m^3$. Hence most of the non-radiated dissipated power has to be carried away by the heavy particle heat conduction in the boundary layer. This will only be possible by virtue of a strong heavy particle temperature gradient in the wall boundary layer, causing T_h to deviate strongly from T_e there.

In molecular plasmas significant changes will occur in the boundary layer: recombination of electrons through molecular processes becomes important in mass balance eq. (21) and the heat conductivity will increase in the heavy particle balance eq. (22). Both effects will lead to an increase of the wall layer thickness. Then the eqs. (21) and (22) have to be solved to obtain wall layer thickness, $\hat{a}-\hat{r}_{eff}$. The effective radius $\hat{r}_{eff}$ can then be used as a boundary condition for the active atomic plasma in the center along the lines indicated in section 4. This is a possible strategy to tackle the more difficult problem of molecular plasmas without the need for full kinetic modeling.

6.CONCLUSIONS

The transport of high density plasma has been reformulated in an approximate way. The main plasma parameters are electron density and pressure and non-equilibrium has been taken into account by using the deviation from equilibrium as other parameters, instead of T_e. It appears that a distinction can be made between an active part, which is Coulomb collision dominated and a passive part.

In the active part the ionization ratio n_e/n_1 is high and Coulomb collisions dominate the transport. In the passive part electron-neutral collision dominate and current conduction can be neglected. The boundary between both regions is defined by a critical ionization degree $(n_e/n_1)_c$ for which e-i and e-o collision rates are equal. For atmospheric arcs in argon this critical ratio is about 10^{-3} and thus $n_{ec} \propto 10^{21}/m^3$.

The basic background for this approximate formulation is the possible extension to the flowing (non-isobaric) arcs and to plasmas in molecular gases. In the latter case the central part will still be atomic of nature and a similar approach as presented above for argon will be valid.

The molecular processes play a rôle in the passive part, which for molecular plasmas will be larger and thus be more important. In the passive part dissipation can be neglected, but some kinetic processes as molecular ion formation, dissociation and dissociative recombination have to be considered. Also the dissociation non-equilibrium can probably be characterized by one additional non-equilibrium parameter which describes the molecular overpopulation with respect to the atomic density at the heavy particle temperature. In this way a first step can be taken on the non-equilibrium route, without the need to consider the numerous kinetic processes between the various molecular components in the periphery of the plasma and by treating the central active part as atomic.
It is with this contribution, that the authors will honour the strong contribution of E.Pfender to the further understanding and technological use of thermal plasmas.

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REFERENCES

1. M.I. Boulos, P. Fauchais, E. Pfender, *Thermal plasmas Fundamentals and application,* vol.1, (Plenum, New York, 1994).
2. P.C. Kong, E. Pfender, *Plasma synthesis of ceramics, A review. Materials processing, Theory and Practices* (Elsevier, Amsterdam, 199?).
3. H. Zhu, Y.C. Lau, E.pfender, *J. Appl. Phys.* 69 (1991) 3404.
4. E. Pfender, Q.Y. Han, T.W. Or, Z.P. Lu, J. Heberlein, *Diamond and Related materials* 1 (1992) 127.
5. T. Yoshida, invited paper in Loughborough I.S.P.C.-10. (1993) *Pure App. Chem.*
6. J.J. Beulens, A.J.M. Buuron, D.C. Schram
J.J. Beulens, *Surface modification using a cascade arc plasma source,* (thesis Eindhoven University of Technolgy 1992).
7. A.J.M. Buuron, M.C.H van de Sanden, W. Ooij, R.M.A. Driessens, D.C. Schram, *J. Appl. Phys.* 78 (1995) 528.
A.J.M. Buuron, *Plasma deposition of carbon materials,* (thesis Eindhoven University of Technolgy, Einhoven 1993).

8. J.J. Beulens, A.T.M. Wilbers, M. Haverlag, G.S. Oehrlein, G.M.W. Kroesen, D.C. Schram, *J. Vac. Sci. Technol.* 10, (1992) 2387-2393.
9. M.J. de Graaf, R.J. Severens, M.J.F. van de Sande, H.J.M. meyers, H. Kars, M.C.M. van den Sanden, D.C. Schram, *J. Nucl. Mat.* 200-380 (1990).
10. Q.Y. Han, J. Heberlein, E. Pfender, *Journal of materials synthesis and processing* 1, 25 (1990).
11. P. Proulx, J. Mostaghimi, M.I. Boulos, *Int. J. heat and mass transfer* 34 (1991) 2571.
12. T.L. Eddy, K.Y. Cho, A multitemperature model for plasmas in chemical nonequilibrium (1991) ASME Heat Transfer Conference Minneapolis, Minnesota, July 28-31
13. J.J. Beulens, D. Milovic, D.C. Schram, P.M. Vallinga, *Phys. Fluids* B3 (1991) 2548.
14. D.A. Benoy, *Modeling of thermal argon plasmas* (thesis, Eindhoven University of Technology, Eindhoven 1993). D.A. Benoy, E.C.J.N. de Jong, F.H.A.C. Fey, J.A.M. van der Mullen, D.C. Schram, *J. of High Temp. Chem. Processes* 1,367(1992).
15. J.C.M. de Haas, *Non-equilibrium in flowing atmospheric plasmas* (thesis Eindhoven University of Technology, Eindhoven 1986).
16. J.A.M. van der Mullen, *Phys. Rep.* 191, 109(1991).
17. D.C. Schram, I.J.J.M. Raaymakers, B. van der Sijde, H.J.M. Swenkelaars, P.W.J.M. Boumans, *Spectrochim. Acta* 38B, 1545(1983).
18. R.J. Rosado, *An investigation of non-equilibrium effects in thermal argon plasmas* (thesis Eindhoven University of Technology, Eindhoven 1981).
19. C.J. Timmermans, R.J. Rosado, D.C. Schram, *Z. Naturforsch* 40A 810(1985); C.J. Timmermans, *An investigation of pulsed high density plasmas* (thesis Eindhoven University of Technology, Eindhoven 1984).
20. H.W. Drawin, *Phys. Letters* 42A, 423 (1973).
21. A.T.M. Wilbers, J.J. Beulens, D.C. Schram, *Proc. ISPC-10*,I,1.1-4, (1991), *J. Quant. Spectrosc. Radiat. Tranfer* 46 385(1991).
22. S. Nowak, J.A.M. van der Mullen D.C.Schram, *Spect. Acta* 43B, 1235 (1985).
23. J.M. de Regt, R.A.H. Engeln, F.P.J. de Groote, J.A.M. van der Mullen, D.C. Schram, *Rev. Sci. Instrum* 66, 3228 (1995).
24. G.M.W. Kroesen, D.C. Schram, C.J. Timmermans, J.C.M. de Haas, *IEEE Transactions on Plasma Science* 18, 985 (1990).

25. J.M. de Regt, J. van Dijk, J.A.M. van der Mullen, D.C. Schram, *J. Appl. Phys.* 28(1995) 40-46; D.A. Bennoy, J.A.M. van der Mullen, D.C. Schram, *J. Phys. D: Apll. Phys.* 26(1993) 1408-1413.
26. E. Leveroni, A.M. Rahal, E. Pfender, *Proc. ISPC-8*, 346, Tokyo, 1987.
27. E. Pfender, *Pure Appl. Chem.* 60, 590 (1988).
28. W.L.T.Chen, J. Heberlein, E.Pfender, *ISPC-10*, Bochum; 1991, I 1.2-12.