

## Characterisation of plasmas produced by the "torche à injection axiale"

**Citation for published version (APA):**

Jonkers, J., Selen, L. J. M., Mullen, van der, J. J. A. M., Timmermans, E. A. H., & Schram, D. C. (1997). Characterisation of plasmas produced by the "torche à injection axiale". In P. Fauchais (Ed.), *Progress in Plasma Processing of Materials 1997 ; Proceedings of the 4th International Thermal Plasma Processes Conference, Athens, July 15-18, 1996* (pp. 109-117). Begell House Inc..

**Document status and date:**

Published: 01/01/1997

**Document Version:**

Publisher's PDF, also known as Version of Record (includes final page, issue and volume numbers)

**Please check the document version of this publication:**

- A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher's website.
- The final author version and the galley proof are versions of the publication after peer review.
- The final published version features the final layout of the paper including the volume, issue and page numbers.

[Link to publication](#)

**General rights**

Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the public portal.

If the publication is distributed under the terms of Article 25fa of the Dutch Copyright Act, indicated by the "Taverne" license above, please follow below link for the End User Agreement:

[www.tue.nl/taverne](http://www.tue.nl/taverne)

**Take down policy**

If you believe that this document breaches copyright please contact us at:

[openaccess@tue.nl](mailto:openaccess@tue.nl)

providing details and we will investigate your claim.

# CHARACTERISATION OF PLASMAS PRODUCED BY THE "TORCHE À INJECTION AXIALE"

*J. Jonkers, L.J.M. Selen, J.A.M. van der Mullen, E.A.H. Timmermans and D.C. Schram  
Department of Physics, Eindhoven University of Technology  
P.O. Box 513, 5600 MB Eindhoven, The Netherlands*

**ABSTRACT:** *Two different kinds of plasmas created by the microwave driven Torche à Injection Axiale (TIA) are investigated: one with helium and the other with argon as the main gas. Using absolute line intensity measurements the densities of the excited states are determined. Applying the ideal gas law gives the ground state density. It is found that both plasmas are ionising and that the excitation temperatures range from 3000 to 11000 K. The electron temperature and the electron density are determined using Thomson scattering. In the plasma with helium as the main gas, average densities between  $0.64$  and  $5.1 \times 10^{20} \text{ m}^{-3}$  and temperatures around 25000 K are found. In an argon plasma the electron temperature is lower and the electron density is higher: 17000 K and around  $10^{21} \text{ m}^{-3}$  respectively. Radial profiles of the electron density, obtained by focusing the laser beam, appear to have a donut-like shape.*

## 1. THE TORCHE À INJECTION AXIALE

The microwave torch called "Torche à Injection Axiale" (TIA), i.e. torch with axial gas injection, was developed by the group of Moisan in 1993 [1]. It consists of a coaxial structure perpendicular to a rectangular waveguide and some elements to ensure optimal power transfer to the plasma, cf. Figure 1. The TIA can, depending on the geometry of the nozzle, excite many kinds of gases or mixtures, such as air, CO<sub>2</sub> and noble gases. Here, we will discuss two plasmas: one with helium and the other with argon as the main gas.

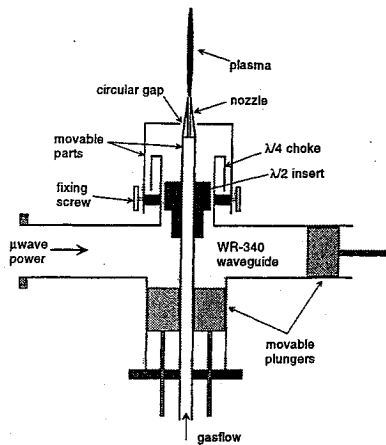


Figure 1: The TIA-structure as proposed by Moisan *et al.* [1] (not on scale).

The gas flows through the inner conductor of the coaxial waveguide and the plasma is created just above the nozzle. Note that no discharge tube is used, the plasma expands in the open air. The plasma has the shape of a needle: it is approximately 2 mm in diameter and 15 to 50 mm long depending on the carrier gas and the applied power. The spectrum below 470 nm is dominated by molecular bands which arise from (association products of) molecules from the surrounding air (like the  $N_2^+$  First Negative system, the  $C_2$  Swan system and the CN Violet system). This indicates towards a strong interaction between plasma and the surrounding air [2].

## 2. ABSOLUTE LINE INTENSITIES

A helium plasma is created using an input power of 300 W and a flow of 5.0 slm. The length of this plasma is approximately 15 mm of which the turbulent upper 3 mm being is not taken into account in this paper. Only five helium lines can be measured accurately. The other lines are either too weak, or outside the sensitive range of our detection system (400 .. 800 nm), or superimposed on molecular bands. Since the plasma is very thin, it is not straightforward to measure radial distributions. Using the data listed in Reference 3 the absolute densities of the five states can be determined. The result is depicted in Figure 2a. It looks as if the atomic state distribution obeys a Boltzmann relation which results in a temperature  $T_{\text{spec}} = 3800 \pm 200$  K. This spectroscopically determined temperature is slightly higher than the temperature found by Quintero *et al.* using relative line intensities:  $3100 \pm 150$  K [4].

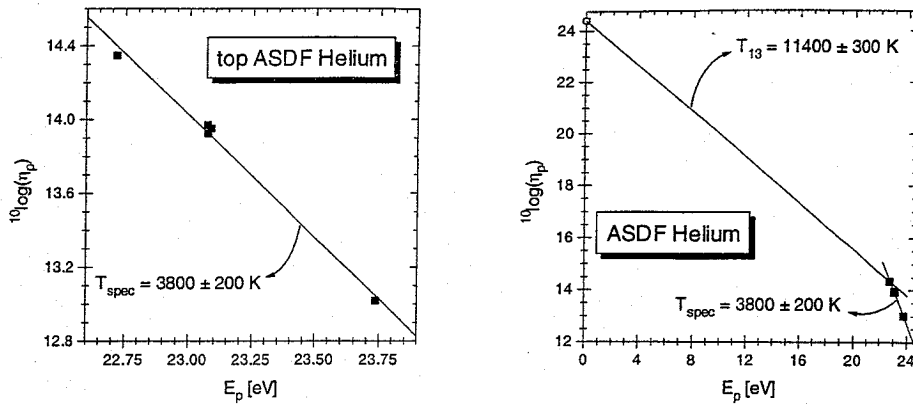


Figure 2: The measured ASDF of the helium plasma created by the TIA (measured at 5 mm above the nozzle). The state occupations depicted in figure a seem to obey a Saha-Boltzmann relation. In figure b the ground state density based on the ideal gas law (hollow circle) is added to the ASDF: strong deviations from Saha-Boltzmann can be observed.

Since absolute densities were measured, it is possible to include the density of the ground state using the ideal gas law. The gas temperature is unknown, but it can be roughly estimated by the rotational temperature of the  $\text{N}_2^+$  molecule:  $3000 \pm 1500$  K [4,5,6]. A large uncertainty margin is taken, since it is not known whether the rotational temperature is a good indication of the gas kinetic temperature. This yields a ground state density equal to  $n_1 \approx (2.4 \pm 1.2) \times 10^{24} \text{ m}^{-3}$ . In Figure 2b the value of the ground state density is combined with results obtained from the spectroscopic measurements. The temperature deduced from the ratio of the densities of the lowest measured excited state (i.e. the third level) and the ground state is found to be  $T_{13} = 11400 \pm 300$  K. Its accuracy is purely determined by the unknown gas temperature.

Since at least two temperatures are necessary to describe the Atomic State Distribution Function (ASDF), it does not obey a Saha-Boltzmann relation. The ASDF depicted in Figure 2b is characteristic for an ionising plasma [7,8]. It can be shown that in this case the obtained excitation temperatures are underestimations for the electron temperatures. Moreover, measuring  $T_{\text{spec}}$  in a strongly ionising plasma using these levels will always yield a temperature around 3800 K [8].

In Figure 3 the temperatures describing the ASDF are depicted as a function of height above the nozzle (AN), i.e. the axial position. At every point, two

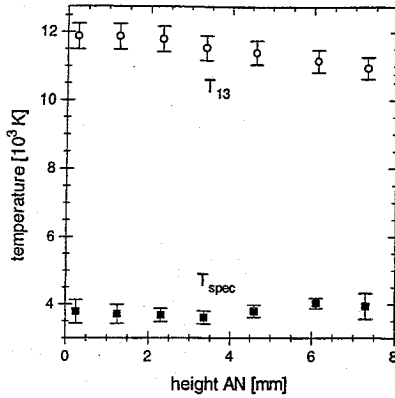


Figure 3: The temperatures describing the ASDF (see Figure 2) versus the height above the nozzle (AN). The error bars of  $T_{spec}$  correspond to the standard deviations in the fitted slopes; those in  $T_{13}$  stem from the uncertainty in the gas temperature.

temperatures are needed to describe the ASDF. Therefore it can be concluded that at every measured position the plasma is not in Local Saha Equilibrium (LSE).

Also the ASDF in argon plasmas is described by two temperatures: one for the states determined by spectroscopy  $T_{spec} = 4860 \pm 150$  K and one for the ratio between the ground state and the first measured excited state (i.e. the third level):  $T_{13} = 8700 \pm 300$  K (at 1 mm AN using 330 W input power and a flow of 3.0 slm).

### 3. THOMSON SCATTERING

In contrast with absolute line intensities, Thomson scattering [9] is not influenced by the absence of equilibrium. Therefore this technique was used to obtain the electron density and temperature. The setup that was used is treated in Reference 9.

In Figure 4a the electron temperature in the helium plasma is depicted as a function of the height above the nozzle (AN). The temperature is more or less stable around 25000 K and drops to 19100 K at 5 mm AN. The accuracy of the fits is approximately 300 K. As can be seen in Figure 4b, it appears that the electron density decreases from  $(5.1 \pm 0.8)$  at 1 mm to  $(0.64 \pm 0.10) \times 10^{20} \text{ m}^{-3}$  at 5 mm AN. Rodero *et al.* [5,6,10] used the Stark broadening of  $H_{\beta}$  to determine  $n_e$  in this helium plasma. Under similar conditions (3.0 slm, 300 W) they found a slightly lower density:  $(4.2 \pm 0.2) \times 10^{20} \text{ m}^{-3}$  just above the nozzle.

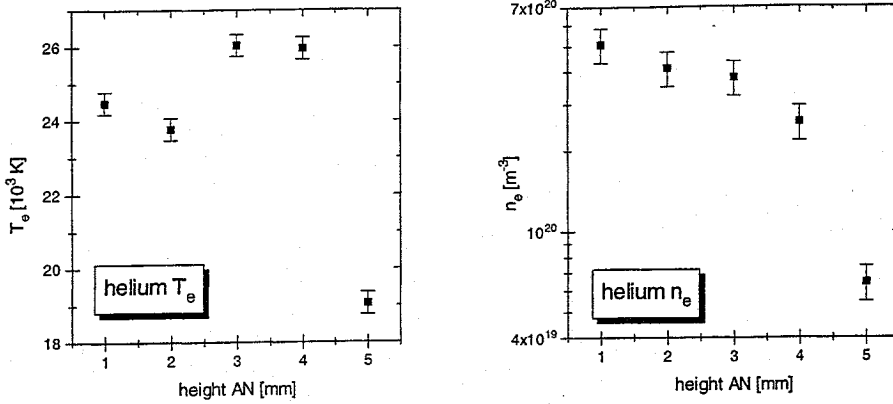


Figure 4: The electron temperatures and densities in the helium plasma.

In the argon plasma the electron temperature remains stable around 17000 K, except for a strange dip between 5 and 6 mm AN (cf. Figure 5a). On the other hand the electron density decreases from  $(22 \pm 3)$  to  $(3.5 \pm 0.5) \times 10^{20} m^{-3}$ , as can be seen in Figure 5b.

As expected in both plasmas the electron temperatures determined by Thomson scattering are much higher than the excitation temperatures presented in the previous section ( $T \leq 11900$  K and  $T \leq 8700$  K for helium and argon respectively). Again this indicates that the plasmas produced by the TIA are far from equilibrium as will be discussed more quantitatively in the next section.

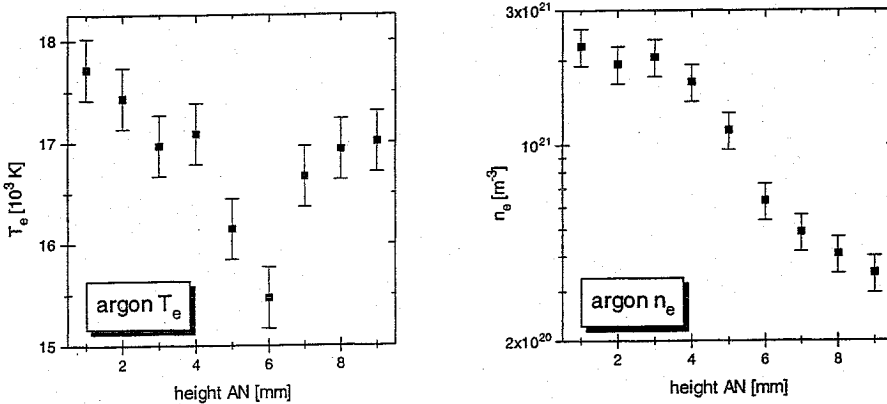


Figure 5: The electron temperatures and densities in the argon plasma.

## 4. PARTICLE BALANCE

The electron temperatures found in the previous section for the helium plasma (19100 to 25000 K) are much higher than the temperatures found by Rodero *et al.* using a modified  $2\lambda$  method: 11000 to 12500 K [5,6]. In this section we will compare the production of free electrons with their losses due to recombination, diffusion and convection. It will be shown that a high electron temperature is required to sustain the plasma.

The particle balance for charged particles is given by

$$n_e n_+ S_{CR} - n_e n_+ \alpha_{CR} = \nabla \cdot (D_a \nabla n_+) + \nabla \cdot (n_+ \mathbf{w}_p) \quad (1)$$

In this equation the terms on the right hand side represent the efflux due to ambipolar diffusion (dominantly in radial direction) and to convection respectively and the terms on the left hand side the net production. The ionisation ( $S_{CR}$ ) and recombination ( $\alpha_{CR}$ ) coefficients can be estimated using a collisional radiative model [7]. For helium we use the model of Drawin and Emard [11] and for argon the one by Benoy *et al.* [12]. The losses of free electrons due to convection, i.e. the second term at the right hand side of (1), can be estimated (assuming no multiple ionised particles) by

$$\nabla \cdot (n_+ \mathbf{w}_p) \approx \frac{n_e}{h_p} w_p \quad (2)$$

In which  $h_p$  is the height of the plasma. The plasma flow velocity  $w_p$  is estimated by the gas flow, the cross section of the nozzle and the heating of the gas by the plasma. The other loss term, i.e. the efflux due to diffusion can be written as

$$\nabla \cdot (D_a \nabla n_+) \approx \frac{n_e}{d^2} D_a \quad (3)$$

The ambipolar diffusion coefficient  $D_a$  is given by [13]. We assume that the characteristic diffusion length  $d$  equals the radius of the plasma. Using these approximations and  $T_e$  and  $n_e$  determined by Thomson scattering, the magnitude of the terms of the particle balance can be estimated. These results are listed in Table 1.

	helium	argon
ionisation	$2 \times 10^{26}$	$3 \times 10^{27}$
recombination	$3 \times 10^{21}$	$7 \times 10^{22}$
convection	$6 \times 10^{24}$	$1 \times 10^{25}$
diffusion		$3 \times 10^{24}$

Table 1: Typical values of the separate terms of the particle balance (in  $m^{-3}s^{-1}$ ), using  $T_e$  and  $n_e$  of Section 3. The diffusion loss rate is obtained using a gradient length of 1 mm.

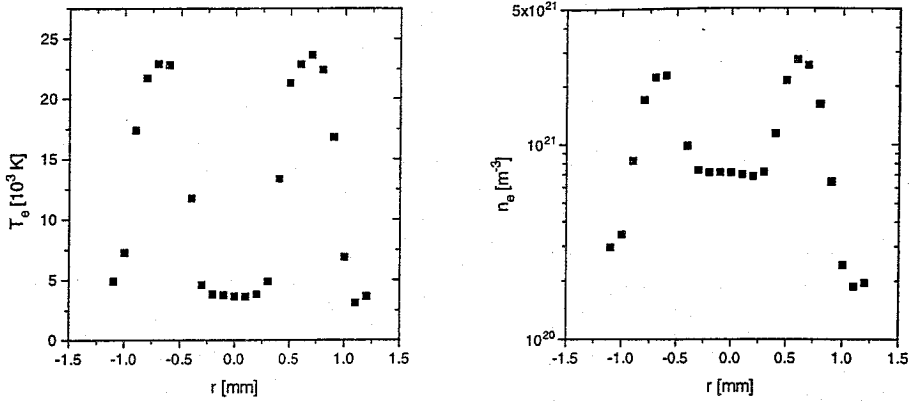


Figure 6: The radial electron temperature and density profiles in the argon plasma at 2 mm AN, obtained by Thomson scattering. The spatial resolution is determined by the entrance slit of the monochromator (0.3 mm). The density profile seems to have a donut like shape, which enhances the losses due to diffusion.

much shorter than 1 mm. Note that the spatial resolution is 0.3 mm, so that the actual diffusion length is probably shorter. This means that the diffusion losses become of the same order of magnitude as the production by ionisation, see Table 1.

Although Rodero *et al.* found similar electron densities in the helium plasma, they found electron temperatures around 12000 K using a modified  $2\lambda$  method [5,6]. As is discussed in section 0 using passive spectroscopy to determine  $T_e$  in these plasmas is questionable. Moreover, for this temperature the ionisation rate becomes  $2 \times 10^{22} \text{ m}^{-3} \text{ s}^{-1}$  [11], which is even too small to balance the losses due to convection ( $6 \times 10^{24} \text{ m}^{-3} \text{ s}^{-1}$ , cf. Table 1). So a much higher  $T_e$  is needed to sustain this plasma.

## 5. CONCLUSIONS

The plasmas created by the *Torche à Injection Axiale* are far from equilibrium. The electron temperature is much higher than the gas temperature. Moreover, its small effective size causes huge losses of charged particles due to diffusion (typical residence time of a free electron is in the order of 1  $\mu\text{s}$ ). In order to sustain the plasma these losses have to be balanced by a strong ionisation rate. This flow through the excitation space gives the atomic state distribution function its typical shape for an ionising plasma, which causes that the measured excitation temperatures are much lower than the electron temperature.



Comparing the ionisation with the loss terms it seems that the production outranges the destruction by two orders. However, the actual losses can be higher due to:

1. mixing with the surrounding air,
2. formation of molecular argon ions,
3. a smaller gradient length.

*ad 1*

As is discussed by Timmermans *et al.* [2] extra destruction channels for the free electrons are created if the surrounding air is mixed with the plasma. For instance, for argon and nitrogen this mechanism is charge transfer followed by dissociative recombination. The charge transfer reaction is resonant since the ionisation energies of argon and molecular nitrogen are almost equal (respectively 15.68 and 15.58 eV). The second reaction is fast due to the Coulomb interaction between the positively charged molecule and the electron. The presence of this mechanism is supported by the fact that in the argon plasma the First Negative system of  $N_2^+$  and many atomic nitrogen lines can be observed [2,4,5,6].

*ad 2*

A second way of losing charged particles is the formation of rare gas molecular ions followed by their destruction via dissociative recombination. Normally, the formation of the molecular ion is the limiting process, which rate is around  $10^{27} \text{ m}^{-3}\text{s}^{-1}$  in the argon plasma (neutral density equal to  $2.4 \times 10^{24} \text{ m}^{-3}$ ). However, since the formation of molecular ions is accompanied by its inverse process, the effective destruction rate is much lower [14].

*ad 3*

An other possible explanation for the large discrepancy between the production and the destruction of ions and free electrons might be that the actual radial gradient length is smaller than the value of 1 mm, as used for estimating the diffusion losses in Table 1. An indication of the gradient length is obtained by the skin effect: the penetration of the microwaves into the plasma is limited, since they are absorbed by the free electrons. According to Jackson [15] the skin depth is given by:

$$\delta = \sqrt{\frac{2}{\mu\omega\sigma}} \quad (4)$$

For an argon plasma at our conditions the relative magnetic permeability  $\mu_r$  equals 1 and its conductivity  $\sigma$  approximately  $10^4 \text{ } \Omega^{-1}\text{m}^{-1}$  [16]. Due to the high frequency of the microwaves (2.45 GHz) the skin depth is only 0.1 mm. In order to measure the radial gradient length Thomson scattering measurements are performed using a focused laser beam. It is found (cf. Figure 6) that the radial gradient length is indeed

## ACKNOWLEDGEMENTS

The authors want to thank Prof. M. Moisan, who kindly gave us permission for using the TIA design. Our TIA was build by F.J. Overberg. The microwave equipment was donated by Philips Research Laboratories Eindhoven. The discussions with D.A. Benoy improved our insight in the processes involved in these plasmas.

## REFERENCES

1. M. Moisan, G. Sauvé, Z. Zakrzewski and J. Hubert, *Plasma Sources, Sci. and Technol.* **3** 584 (1994).
2. E.A.H. Timmermans, J. Jonkers, J.A.M. van der Mullen and D.C. Schram, "Microwave Induced Plasmas for the analysis of Molecular Components in Incinerator Gases", **P3/15**, this conference.
3. W.L. Wiese and G.A. Martin, "Wavelengths and Transition Probabilities for Atoms and Atomic Ions", part 2: "Transition Probabilities", U.S. Government Printing Office, Washington (1980).
4. M.C. Quintero, A. Rodero, A. Gamero and A. Sola, *Europhysics Conference Abstracts* 18E ESCAMPIG XII 342 (1994).
5. A. Rodero, M.C. Quintero, A. Sola and A. Gamero, *Spectrochim. Acta B.* **51** 467 (1996).
6. A. Rodero, M.C. García, M.C. Quintero, A. Sola and A. Gamero, *J. Phys. D: Appl. Phys.* **29** 681 (1996).
7. J.A.M. van der Mullen, *Phys. Rep.* **191** 109 (1990).
8. J.A.M. van der Mullen, J.M. de Regt, R.D. Tas and J. Jonkers, "New Transition Probability Values for Argon for mapping Open and Closed ICPs", **P3/16**, this conference.
9. J.M. de Regt, R.A.H. Engeln, F.P.J. de Groote, J.A.M. van der Mullen and D.C. Schram, *Rev. of Sci. Instrum.* **66** 3228 (1995).
10. A. Rodero, M.C. Quintero, M.C. García, A. Sola and A. Gamero, *Europhysics Conference Abstracts* 18E ESCAMPIG XII 203 (1994).
11. H.W. Drawin and F. Emard, *Z. Physik* **243** 326 (1971).
12. D.A. Benoy, J.A.M. van der Mullen and D.C. Schram, *J. Quant. Spectrosc. Radiat. Transfer* **46** 195 (1991).
13. R.S. Devoto, *Phys. Fluids* **9** 1230 (1966).
14. J.A.M. van der Mullen, J.M. de Regt, J. Jonkers and F.P.J. de Groote, "Recombination and Diffusion in an ICP during Power Interruption", plenary session **III**, this conference.
15. J.D. Jackson, "Classical Electrodynamics", John Wiley Inc., New York (1975).
16. J. Aubreton, C. Bonnefoi et J.M. Mexmain, *Revue Phys. Appl.* **21** 365 (1986).