2D modelling of an Ar microwave sustained discharge at intermediate pressure: a comparison with the experiment

V. Georgieva[1], A. Berthelot[2], T. Silva[3], St. Kolev[2,4], G. Chen[1,3], N. Britun[3], T. Godfroid[5], D. Mihailova[6], W. Graef[6], J. van Dijk[6], R. Snyders[4,5], A. Bogaerts[2] and M.-P. Delplancke-Ogletree[1]

[1] *4MAT, Université libre de Bruxelles, 50 avenue F.D. Roosevelt, 1050 Brussels, Belgium*
[2] *Research Group PLASMANT, Department of Chemistry, University of Antwerp, 2610 Antwerp-Wilrijk, Belgium*
[3] *ChIPS, Université de Mons, 23 Place du Parc, 7000 Mons, Belgium*
[4] *Faculty of Physics, Sofia University, 5 James Bourchier Boulevard, 1164 Sofia, Bulgaria*
[5] *Materia Nova Research Center, avenue N. Copernic 1, 7000 Mons, Belgium*
[6] *Department of Applied Physics, Eindhoven University of Technology, P O Box 513, 5600 MB Eindhoven, the Netherlands*

Abstract: The plasma characteristics of an Ar surface-wave sustained microwave discharge at intermediate pressure have been investigated by 2-dimensional (2D) modelling and by experimental measurements. The initial tests show good agreement between the measured and calculated electron temperature and Ar metastable density at the conditions under study.

Keywords: microwave surfaguide plasma, 2D fluid model, optical emission spectroscopy

1. Introduction

Surface-wave sustained discharges have been extensively studied in the past 20 years [1, 2]. They are well-known for their useful characteristics for molecular gas conversion, that result from (i) the high degree of non-equilibrium ($T_e >> T_{gas}$) [3], (ii) the high level of applied energy absorption by plasma electrons [4], and (iii) relatively easy operation, etc. In particular, these discharges are flexible in terms of continuous or pulsed operation [5] in a wide range of gas pressure (10^{-5} Torr), applied frequency (500 kHz - 10 GHz), and geometry size and shape [2]. The atmospheric pressure plasma characteristics have been studied more extensively both by modelling and experimental measurements [6, 7], while there is no 2D self-consistent model for the intermediate pressure microwave sustained discharges and experimental measurements are scarce in the literature [8]. The application of the intermediate pressure surface-wave sustained discharges for greenhouse gas conversion to valuable chemicals [9] requires a careful investigation of their characteristics. In the present work, we study, both by modelling and experimental measurements, the Ar plasma sustained by a surfaguide wave launcher operating at 0.915 or 2.45 GHz, and at a gas pressure in the range of 250-5300 Pa (2 - 40 Torr). The applied power varies from 50 to 500W in the experiment.

2. 2D model of the microwave set-up

The experimental set-up of the microwave surfaguide discharge system is shown in Fig. 1. The discharge is sustained inside a quartz tube 14 mm in diameter and 24 cm long surrounded by a polycarbonate tube for cooling purposes. The inner tube is cooled down during the experiments by an oil flow at 10 °C. The metallic net (not shown in Fig.1), which surrounds the plasma tube and forms a Faraday cage, has a radius of 40 mm. The center of the quartz tube is positioned in the waveguide gap. The gas mixture injected from the top of the system is regulated by electronic mass flow controllers.

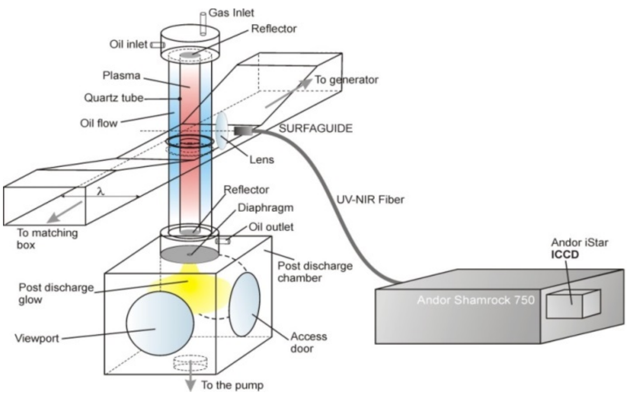

Fig. 1. Schematic representation of surface-wave microwave set-up and the OES system used for plasma diagnostics.

In the developed 2D models, the fluid description of the plasma is coupled to a self-consistent solution of Maxwell's equations. The computational domain corresponding to the experimental set-up is presented in Fig. 2. It consists of an electromagnetic region (EMR), where Maxwell's equations are solved, and a plasma region (PR), where plasma equations are solved. The EMR (see A.I.J.G.H.K.L.B.A in Fig. 2) includes the PR, the internal quartz tube (CD), the external polycarbonate tube (EF), where the cooling oil flows, and the air between the (EF) tube and the metallic net, forming a Faraday cage. The yellow region presents the wave-launching region. The waveguide gap diameter is set to 15 mm in the model.

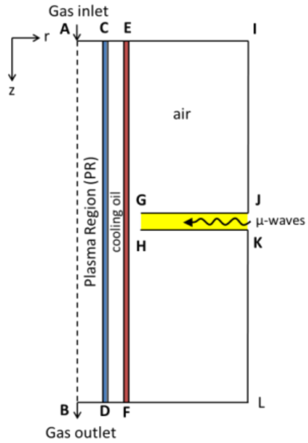

Fig. 2. The computational domain.

Two different software codes are used to model the described system, and a comparison is made. The first software is Plasimo [10], developed by the research group EPG from TU/e, Eindhoven. For each heavy species the balance equations (species density and momentum) are solved. The electron density is determined from quasi-neutrality and the electron temperature from the electron energy balance. The gas flow is taken into account by solving the mass, momentum and energy balance equations for the plasma bulk. The axial velocity at the gas inlet is assumed to follow a parabolic radial profile and is calculated from the gas flow rate. A detailed numerical description can be found in [11].

The second model is developed within the commercial software Comsol Multiphysics ® (www.comsol.com), and solves a set of equations similar to the previous model except that the quasi-neutrality is not assumed and an additional continuity equation is solved to determine the electron density as well as the Poisson equation for the electric potential. Comparing the two models could show the influence of the quasi-neutrality assumption and the sheath importance once the two models are matured. A tube of 1m long, instead of 24 cm, was initially considered in the Comsol model so that the effect of the boundary conditions on the plasma is smaller. All other operating conditions are the same in the two models, however, because of the different volumes, the deposited power density is not the same, which influences the maximum values of plasma density and gas temperature.

The Ar plasma chemistry is based on the set presented in [12]. The following species are considered in the model: electrons, Ar atoms, Ar^+, Ar_2^+, and the lumped states of 4s and 4p groups: Ar(4s) and Ar(4p). The Ar atom density is calculated from the ideal gas law.

3. Plasma diagnostics

Optical emission spectroscopy (OES) provides a non-intrusive way of studying many types of plasmas, being an easy-to-implement technique. Since a considerable amount of plasma physics knowledge can be evaluated from the plasma emission spectra, numerous research works have been dedicated to this diagnostic tool to characterize different discharge properties [13, 14].

In this study, plasma emission from excited argon atoms is used to characterize the microwave surfaguide discharge. The emission spectrum for pure argon plasma in the range of 250 - 920 nm is shown in Fig. 3. The most intense emissions are from the 4p → 4s (or in Paschen notation: 2p → 1s; cf. Fig. 3) transitions in the 600 - 900 nm range followed by the second set of lines belonging to 5p → 4s (or in Paschen notation: 3p → 1s) emission in the 395 - 470 nm spectrum range. See [6] for more details.

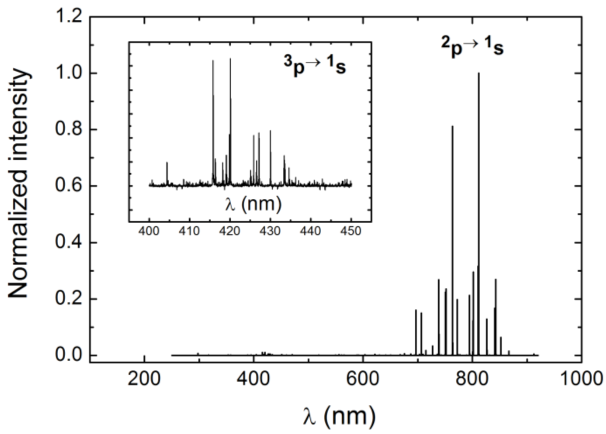

Fig. 3. Emission spectrum from a pure Ar microwave surfaguide discharge. Inset: Zoom of the 3p → 1s (Paschen notation) emissions.

4. Results and discussion

4.1. Simulation results

The initial investigation is focussed on benchmarking of the two simulation models. The output of the models gives information on the spatial distribution of a number of characteristics, i.e., species density and temperature, electric and magnetic field intensity, deposited power density, diffusion coefficients, etc. The following operating conditions are used as an input: applied power of 80 W, pressure 1000 Pa, and gas flow rate of 500 sccm. Fig. 4 presents the calculated electron temperature T_e (a), electron density n_e (b), gas temperature T_g (c), and Ar(4s) density (d), calculated by Plasimo (first column) and by Comsol (second column). The main Ar^+ ion density has a similar profile and values as the electron density, and therefore is not presented.

It is found that the T_e calculated by both models is in the order of 1 - 1.1 eV and does not change considerably in the volume, having maximum values in front of the waveguide (Fig. 4a). Note that the results from Plasimo

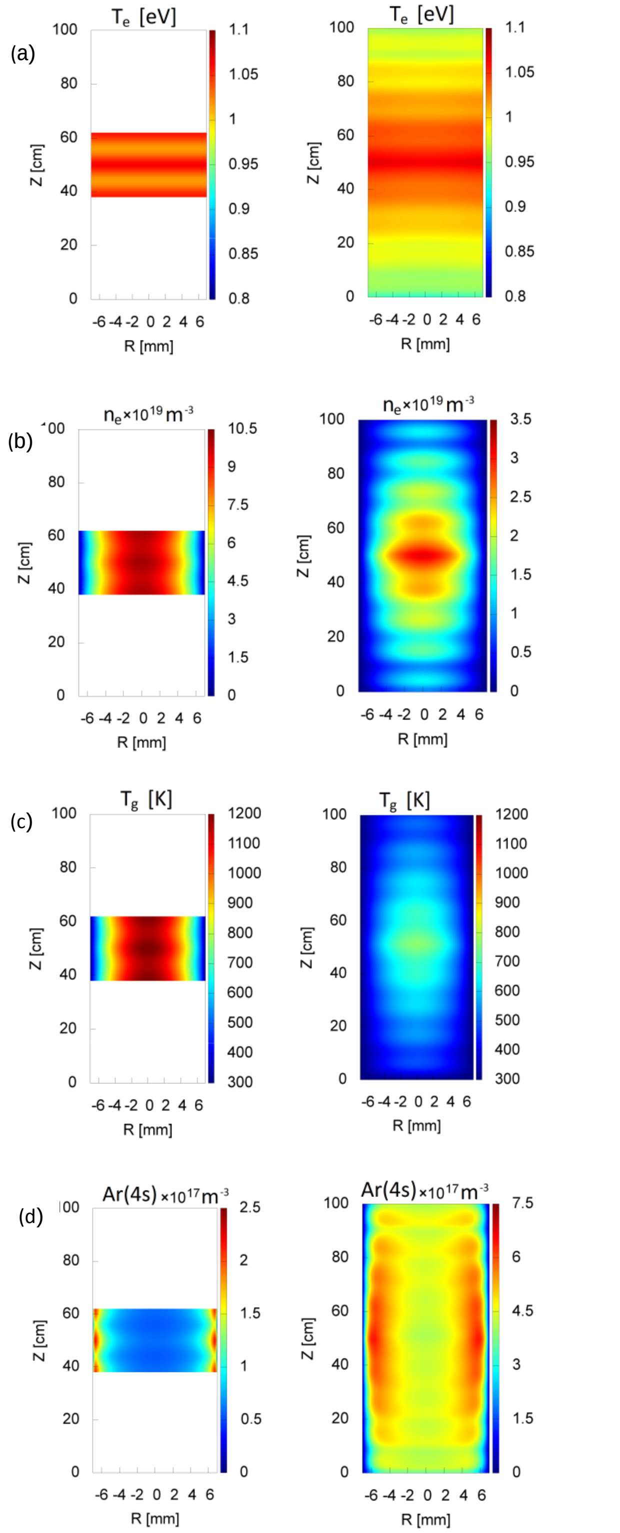

Fig. 4. Electron temperature (a), electron density (b), gas temperature (c), and Ar(4s) density (d), calculated by Plasimo (first column, tube length 24 cm) and by Comsol Multiphysics ® (second column, tube length 100 cm) in PR (see Fig. 2) at pressure of 1000 Pa (7.5 Torr), power of 80 W, and gas flow rate of 500 sccm.

considering 24 cm tube are presented in the same scale as the results from Comsol considering 1m tube, taking into account that the tube centre in both models is positioned in the waveguide launcher. It is clear that the difference in the simulation geometry size can lead to different values of plasma density and gas temperature since the same power is distributed in different volumes. The maximum electron (and Ar^+ ion) number density is calculated in Plasimo in the order of 10^{20} m^{-3}, while in Comsol it is calculated to be a 3 times smaller, i.e., 3.5x10^{19} m^{-3} (note scale difference by a factor of 3 in Fig. 4b). The maximum gas temperature is calculated to be 1200 K in the tube centre in the Plasimo model, while it is about 800 K in the Comsol model. In order to improve the benchmarking, we are currently implementing the same simulation domain in both models by limiting the electromagnetic field to the central 24 cm of a plasma tube with a length greater than 24 cm. The new simulation geometry allows imposing correct boundary conditions of zero species density gradients at the inlet and outlet, where there is no plasma, similar to the experiment. Despite of the discrepancy in the absolute values, which might be due to the different power density, the presented initial benchmarking shows reasonable agreement between the profiles of the electron and heavy species densities, gas temperature, and the electric field (not shown here). The excited species and the molecular ion Ar_2^+ densities show maxima close to the cold tube surface (skin effect) in both models, where the T_g is close to room temperature and the power, absorbed by the plasma, is maximal. Further careful analysis of the production and loss processes and species transport might clarify the observed profiles.

4.2. Experimental results

Using a corona model method associated to a simple emissive spectroscopy technique [15], the metastable argon density and electron temperature have been evaluated. In particular in this approach, the 425.9/750.4 ($3p_1/2p_1$) and the 737.2/750.4 ($4d_4/2p_1$) neutral/neutral line pairs of atomic Ar are used to determine the electron temperature. The effect of the operating parameters such as the applied power and total gas pressure were studied with an effort to characterize the plasma system. As an illustration of the applied method, Fig. 5 shows the evolution of the electron temperature and density for Ar metastable $1s_3$ (in Paschen notation). In general it was found that in our system the electron temperature decreases slightly with increasing power and with increasing gas flow rate.

The measured electron temperature of 1.3 eV (Fig. 5) at a power of 100 W and a pressure of 267 Pa (2 Torr) is in reasonably good agreement with the predicted value of 1.1 eV from the simulations (Fig. 4a) at similar operating conditions. In the simulation we calculate the lumped state of the 4s group density. The statistical weight of the Ar metastable $1s_3$ in the 4s group is 1/12 [16].

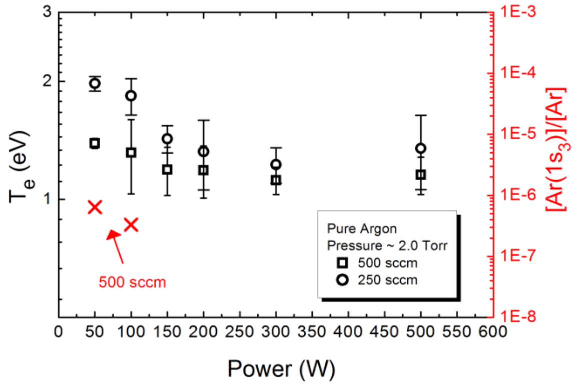

Fig. 5. Left scale: evolution of electron temperature in pure Ar for 500 sccm (black squares) and 250 sccm (black circles) as a function of power. Right scale: density of Ar metastable relative to the ground state (red crosses).

Hence based on the simulation result, the ratio $[Ar(1s_3)]/[Ar]$ is calculated (averaged over the radial profile in front of the waveguide) to be $1x10^{-7}$ (Plasimo) and $3.3x10^{-7}$ (Comsol). The value calculated by Comsol is in good agreement with the measured ratio of $3.5x10^{-7}$ at similar operating conditions.

5. Conclusions

Microwave sustained Ar discharges at intermediate pressure are simulated by 2D self-consistent models employing either Plasimo software (quasi-neutrality assumption) or Comsol Multiphysics ®. The initial tests show reasonably good agreement between the two models. It is found that the electron density has a maximum in the centre, along the axis of symmetry in the tube; the electron temperature does not change considerably in the volume and is in the order of 1 - 1.1 eV, having maximum values in front of the waveguide. The excited Ar atoms and the molecular ions have maximum densities near the tube walls. In addition, the plasma characteristics are measured using the OES technique. The comparison of the simulated and measured electron temperature and Ar metastable density to Ar atom density ratio shows good agreement, taking into account that both simulation models and experimental measurements of the Ar plasma characteristics are still under development. The influence of the applied power, pressure and gas flow rate is being studied and is expected to help in understanding the interconnection between electromagnetic surface waves and plasma.

6. Acknowledgments

This research was carried out in the framework of the network on Physical Chemistry of Plasma-Surface Interactions - Interuniversity Attraction Poles, phase VII (http://psi-iap7.ulb.ac.be), and was supported by the Belgian Science Policy Office (BELSPO). N. Britun is a post-doc researcher of the FNRS, Belgium.

7. References

[1] M. Moisan and Z. Zakrzewski. *J. Phys. D: Appl. Phys.*, **24**, 1025 (1991)
[2] H. Schlüter and A. Shivarova. *Phys. Rep.*, **443,** 121 (2007)
[3] A. Fridman and L.A. Kennedy. *Plasma Physics and Engineering*. (New York: Taylor & Francis) (2011)
[4] Yu.A. Lebedev. *J. Phys.: Conf. Ser.*, **257**, 012016 (2010)
[5] T. Godfroid, J.P. Dauchot and M. Hecq. *Surf. Coat. Technol.*, **174-175**, 1276 (2003)
[6] M.C. Garćıa, A. Rodero, A. Sola and A. Gamero. *Spectrochim. Acta B*, **55**, 1733 (2000)
[7] Y. Kabouzi, D.B. Graves, E. Castaños-Martínez and M. Moisan. *Phys. Rev. E*, **75**, 016402 (2007)
[8] C. Boisse-Laporte, A. Graniert, E. Dervisevic, P. Leprince and J. Marec. *J. Phys. D: Appl. Phys.*, **20**,197 (1987)
[9] G. Chen, T. Silva, V. Georgieva, T. Godfroid, N. Britun, R. Snyders and M.-P. Delplancke-Ogletree. *Int. J. Hydrogen Energy*, **40**, 3789-3796 (2015)
[10] J. van Dijk, K. Peerenboom, M. Jimenez, D. Mihailova and J. van der Mullen. *J. Phys. D: Appl. Phys.*, **42,** 194012 (2009)
[11] M. Jimenez-Diaz, E.A.D. Carbone, J. van Dijk and J.J.A.M. van der Mullen. *J. Phys. D: Appl. Phys.*, **45**, 335204 (2012)
[12] St. Kolev and A. Bogaerts. *Plasma Sources Sci. Technol.*, **24,** 015025 (2015)
[13] N. Britun, T. Godfroid and R. Snyders. *Plasma Sources Sci. Technol.*, **21,** 035007 (2012)
[14] T. Silva, N. Britun, T. Godfroid and R. Snyders. *Plasma Sources Sci. Technol.*, **23,** 025009 (2014)
[15] J. Boffard, C. Lin and C. A. DeJoseph Jr. *J. Phys, D: Appl. Phys.*, **37**, R143-R161 (2004)
[16] A. Bogaerts and R. Gijbels. *J. Appl. Phys.*, **84**, 121 (1998)