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DIFFERENT PRESSURE REGIMES OF A SURFACE-WAVE DISCHARGE IN ARGON: A MODELING INVESTIGATION

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Abstract. In this paper, we present a 2D axisymmetric plasma fluid model for microwave plasmas operating in argon. The effect of the pressure on the plasma parameters is studied over a wide range of pressures going from 10 mbar to atmospheric pressure. The model shows that the discharge contracts with the increase of gas pressure. Additionally, a first reduction of the CO_2 chemistry set of [11,12] from 126 species to 35 species is also presented in this paper. It is shown that this reduction has very little effect on the quantities of interest at the conditions under study.

1. INTRODUCTION

Over the last decades, many studies focused on the description of surface-wave plasmas produced by microwave discharges [1], both from a theoretical and an experimental point of view. These discharges have a wide range of applications, such as gas conversion, plasma medicine, material processing and surface treatment [2], as they offer relative simplicity and low running costs. They can be operated over a wide range of pressure (from a few mtorr to several atmosphere), using different frequencies and various geometries [3]. Among these different geometries, the so-called surfaguide discharges offer the possibility to create low-temperature plasmas using a broad range of operating frequencies, determined by the geometry of the waveguide, and they are able to handle high power coupling between the plasma and the microwave source [3].

A review of the physical processes occurring in low-pressure surface-wave discharges is presented in [4]. More recently, a self-consistent 2D model was proposed in [5], applied to a discharge at atmospheric pressure. Recently, microwave discharges are gaining increasing interest for CO_2 conversion and the pressure has been shown to be a key parameter to obtain better energy efficiency [6], which is crucial in this domain. This type of discharges allows the dissociation of CO_2 through vibrational excitation, requiring less energy than dissociation though electron impact or thermal dissociation [6].

The goal of this study is to get a better understanding of the effect of the pressure on the microwave discharge operation. The presented model is a self-consistent 2D fluid plasma model for argon, applied over a wide range of pressure conditions: from an intermediate pressure of 10 mbar to atmospheric pressure. We did not apply the model to pressures below 10 mbar, because of the limitations of the fluid approach used. A reduction of the chemistry set of [11] and [12] is additionally being carried out, so that the model can be used for CO_2 discharges. The first available results are also presented here.

2. MODEL DESCRIPTION

The model is a time-dependent model implemented in the commercial software COMSOL Multiphysics ® (www.comsol.com). It solves equations to determine each species density (including the electrons), the electron energy, the gas temperature and velocity, the plasma potential and the electromagnetic fields. The model considers the following heavy species: Ar atoms, Ar^+ ions, Ar_2^+ molecular ions, Ar(4s), i.e., all 4s levels considered as a single lumped excitation level, Ar(4p), i.e., all 4p levels and Ar_2^* which includes $Ar_2(^3\Sigma_u^+)$ and $Ar_2(^1\Sigma_u^+)$ excited molecules. The set of reactions between the different species, as well as the transport parameters used, are presented in [7]. The EEDF is precalculated using Bolsig+ [8] and it is used to calculate the rate coefficients of electron impact collisions for different pressure regimes.

The frequency considered is 2.45 GHz and a sketch of the 2D axisymmetric computational domain is shown in Fig. 1. The computational domain is based on the setup used at the University of Mons, presented in [9]. Additionally, two metallic grids were placed inside the quartz tube to prevent microwave energy leaks. These grids are at a distance of 33 cm from each other. It is assumed in the model that these grids only act as a mirror for the microwave field and have no other effect on the plasma.

By comparing the EM field distribution in a 3D geometry and in a 2D axisymmetric geometry, we have made sure that a 2D axisymmetric model could accurately reproduce the EM field created in this configuration.

At intermediate pressure, we have made sure that the trends agree with [10] at similar conditions. At atmospheric pressure, a benchmarking of the model results has been done by comparing the results with literature [5]. It was found that the model is able to accurately reproduce the results of [5].

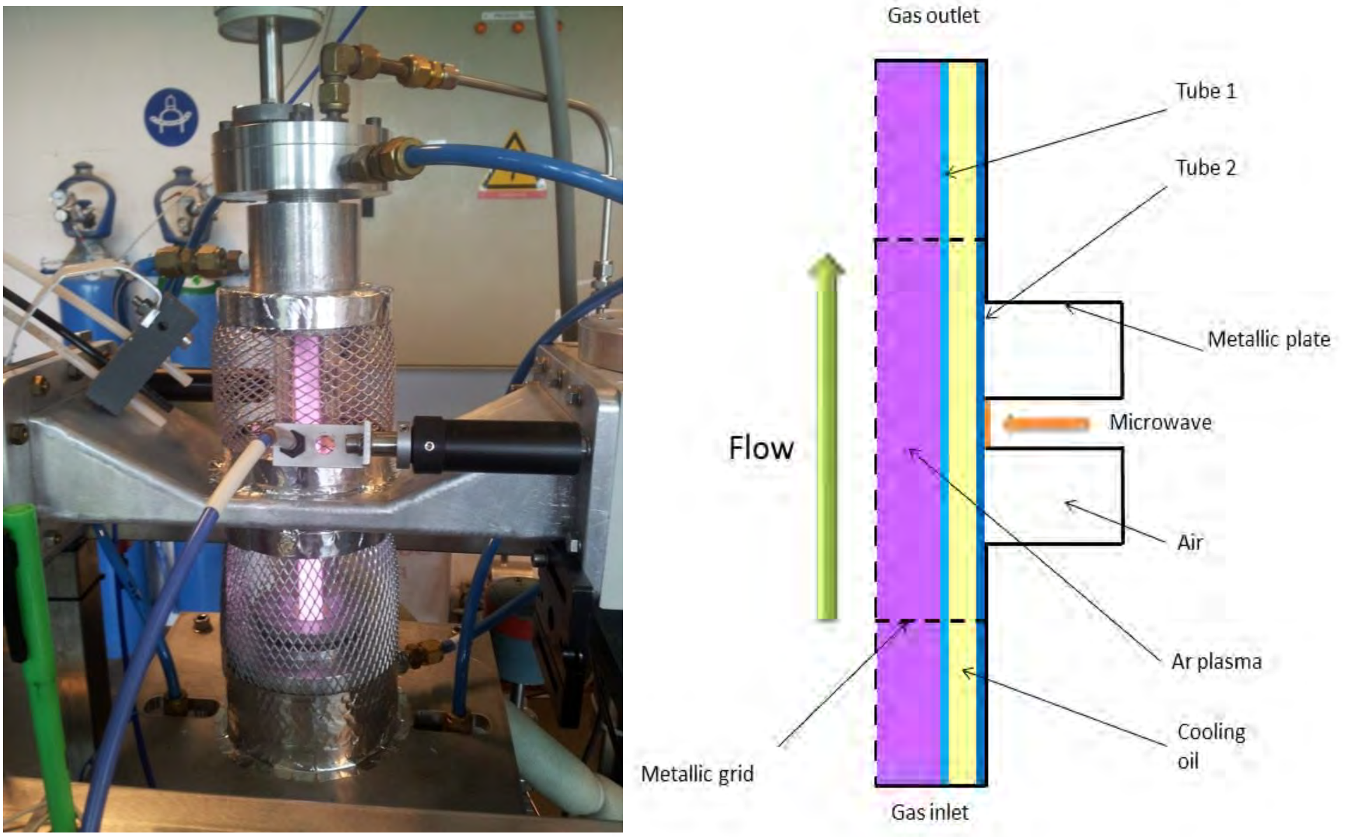

Figure 1. Microwave setup (on the left) and sketch of the computational domain (on the right)

3. RESULTS OF THE ARGON MODEL AND DISCUSSION

First, the model was developed for a pressure of 1000 Pa. Fig. 2 shows the distribution of the electron density (a), the electron temperature (b), the gas temperature (c) and the norm of the microwave electric field (d) at a pressure of 1000 Pa, a flow of 500 sccm and an absorbed microwave power of 100W.

It is found that the electron density reaches a maximum value in front of the waveguide. Moreover, the electron density profile exhibits secondary maxima that are located approximately 6 cm, i.e. half a wavelength, from each other. The maximum electron density is found to be $8{,}3\text{x}10^{19}$ m^{-3} with a 34 cm long plasma. The electron temperature profile is almost constant in the entire plasma region with values between 1 and 1.2 eV. The gas temperature (T_g) profile resembles the electron density profile. The maximum value of T_g is 1400 K. Note that T_g drops to almost 300 K close to the quartz tube since the outer surface of the quartz tube is assumed to be cooled at 300 K. The norm of the microwave electric field is at maximum at the boundary between the plasma and the quartz tube, in the sheath region. It decays exponentially in the radial direction, indicating a skin effect. The electron density is indeed too high in the plasma to allow the field to propagate there.

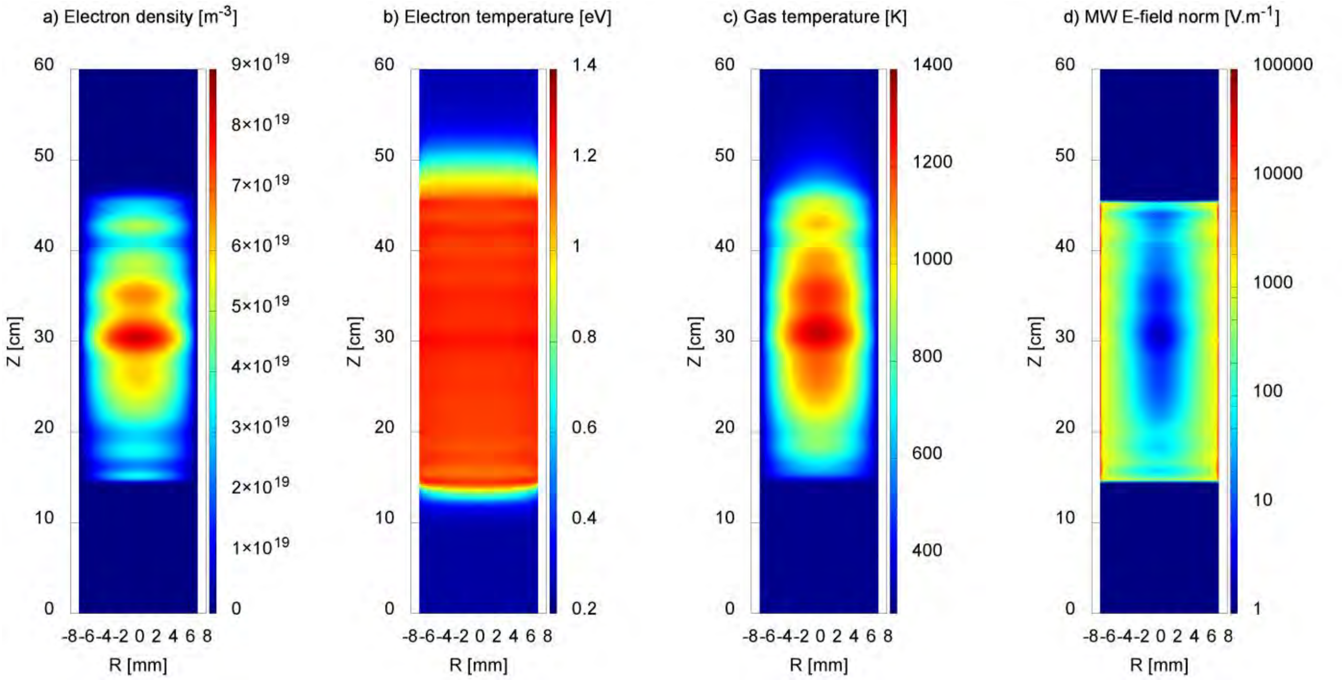

Figure 2. Electron density (a), electron temperature (b), gas temperature (c) and norm of the microwave electric field (d). Pressure: 1000 Pa, Power: 100W, Gas flow: 500 sccm

Axially, omitting the secondary maxima, the electron density decreases linearly from the maximum value in the centre of the tube to the end of the plasma, which is consistent with literature [10]. Note that these two secondary maxima are due to an additional resonance of the EM field caused by the two horizontal metallic grids shown in Fig. 1. The two metallic grids also confine the plasma in between them.

Fig. 3 shows the results obtained at atmospheric pressure in the same configuration and with the same conditions of flow and power. It is found that the maximum of electron density is $4{,}5 \times 10^{19}$ m^{-3}. It is located on the symmetry axis but it is shifted axially by 3 cm compared to the intermediate pressure case. An off-axis secondary maximum is also present in front of the waveguide. The electron temperature shows values between 1.2 and 1.4 eV in the plasma. The electron temperature is at maximum around the plasma. The plasma is about 6 cm long in this case. The gas temperature reaches higher values at atmospheric pressure: between 1500 K and 3000 K in the plasma. The norm of the microwave electric field is still at maximum at the interface between the quartz tube and the plasma, also showing a skin effect. However, it exhibits two weak secondary maxima outside the plasma. The presence of a microwave electric field outside the plasma is also visible on the electron temperature profile: a small number of electrons receive a small power deposition, which results in a local increase of the electron temperature. The shape of the microwave field is explained by the presence of the two metallic grids which act together as a resonator. It is more visible than at intermediate pressure since the plasma does not occupy the whole space.

By comparing the results of the model at intermediate and atmospheric pressure, it is clear that the discharge is more contracted axially at atmospheric pressure (6 cm vs 34 cm) but also radially, as the plasma does not occupy the whole radial space. This effect has been observed experimentally and can be quite challenging to reproduce in a model [5]. Note that the filamentary structure that can be observed at some pressure regimes cannot be reproduced in a 2D axisymmetric model.

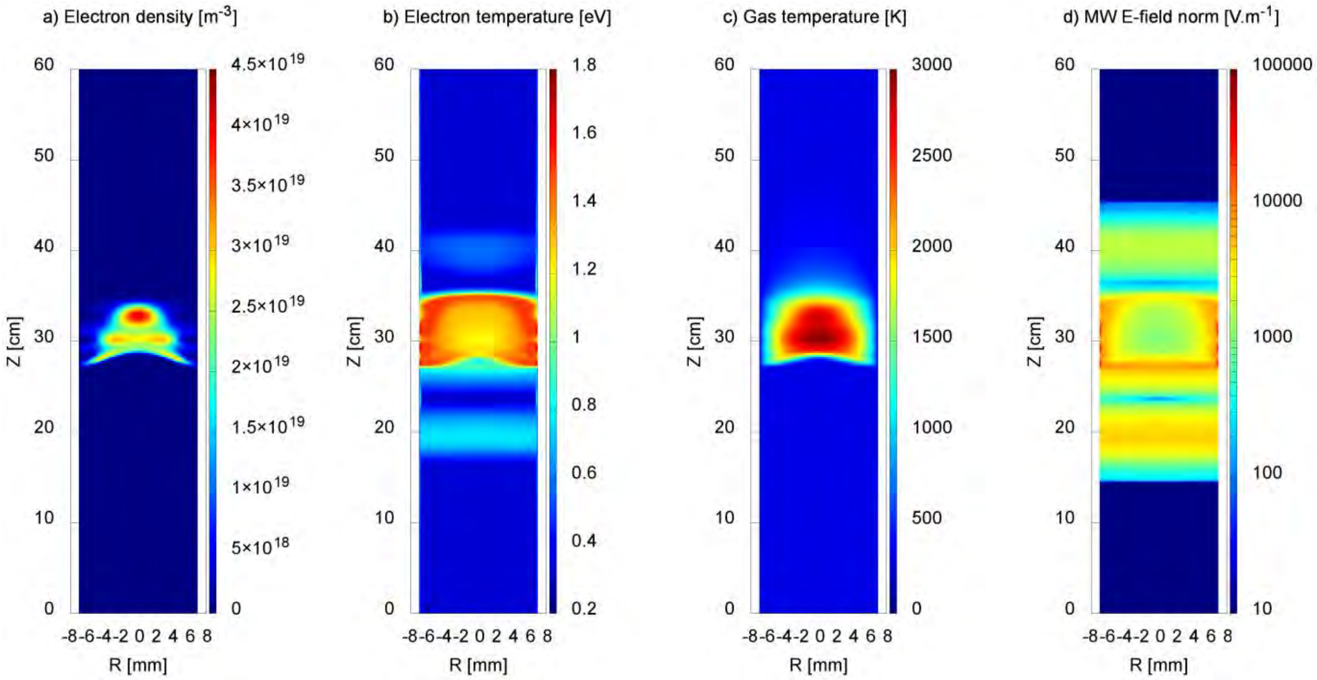

Figure 3. Electron density (a), electron temperature (b), gas temperature (c) and norm of the microwave electric field (d). Pressure: 10^5 Pa, Power: 100W, Gas flow: 500 sccm

4. REDUCTION OF THE CO_2 CHEMISTRY SET

The CO_2 chemistry set available in our group and presented in [11,12] contains 126 species and more than 9000 reactions. This is not suitable for 2D modelling because of the limited amount of computational resources. Therefore, a first reduction of this chemistry set was carried out.

Molecules	Charged species	Radicals	Excited species
CO_2, **CO**	**CO_2^+**, CO_4^+, CO^+, $C_2O_2^+$, $C_2O_3^+$, $C_2O_4^+$, C_2^+, C^+, **CO_3^-**, CO_4^-	C_2O, **C**, C_2	**CO_2(Va, Vb, Vc, Vd)**, **CO_2(V1-V21)**, **CO_2(E1**, E2), CO(V1-V63), CO(E1-E4)
O_2, O_3,	O^+, O_2^+, O_4^+, **O^-**, O_2^-, O_3^-, O_4^-	**O**	O_2(V1-V4), O_2(E1-E2)
	electrons		

Table 1. Species considered in the full model. The species in bold characters are the only ones considered in the reduced model.

Table 1 shows the species considered in the chemistry set of [11,12]. A zero-dimensional model presented in [11] and implemented in ZDPlaskin [13] was used to find out which species have little influence on the plasma at the conditions under study. At every step of the reduction, the CO production and the vibrational distribution of CO_2 were monitored, as these quantities constitute the most important output of the model. In all cases, a power deposition of 250 W was used. The final chemistry set of the reduced model is indicated by the species in bold in Table 1.

Table 2 shows the difference in CO production predicted by the full and the reduced model at two different pressures: 5000 Pa and 50000 Pa. It is found that the difference in predicted CO production is between 0.6% and 3% in the range under study.

Fig. 4 shows the vibrational distribution of CO_2 predicted by the model with the full and the reduced set, again at 5000 Pa and 50000 Pa. The distribution is shown both in the plasma and in the afterglow.

The difference between the predictions of the two sets is negligible. This comparison shows that this 35-species chemistry set can accurately describe the production of CO through the vibrational excitation of CO_2 at the conditions under study.

	CO produced [10^{21} m^{-3}]		
	Full set	Reduced set	Difference
5000 Pa	6,73	6,53	3 %
50 000 Pa	0,184	0,182	0,6 %

Table 2. CO production predicted by the model with the full and the reduced set at 5000 Pa and 50000 Pa.

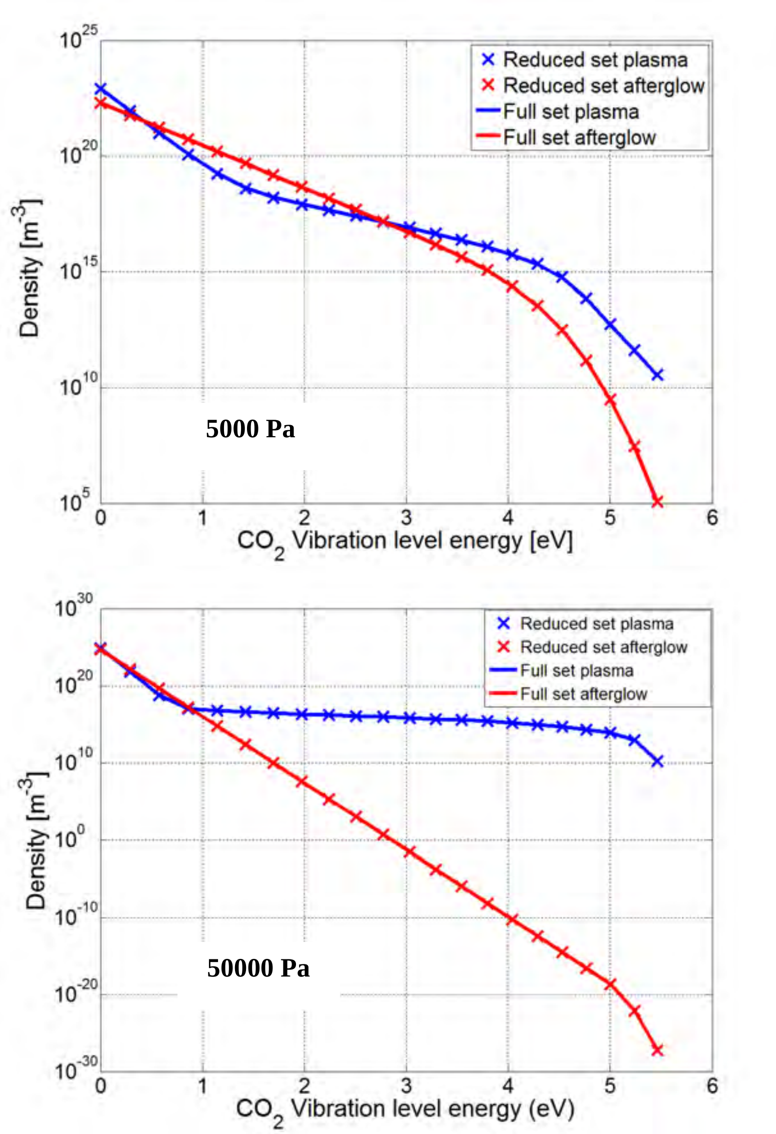

Figure 4. CO_2 vibrational distribution in the plasma (blue) and in the afterglow (red), using the full model (lines) and the reduced model (crosses).
Top panel: 5000 Pa; Bottom panel: 50000 Pa

5. CONCLUSIONS

A microwave discharge in argon is modelled using Comsol Multiphysics ® at different pressures. A comparison of the model results at different pressures shows an axial and a radial contraction of the plasma.

The results at intermediate and atmospheric pressure appear to be in good agreement with previous research conducted on similar setups.

A reduced CO_2 chemistry set is being made from the set of [11,12]. This chemistry set will undergo a further reduction to be used in a 2D model. This will give insights on the effect of pressure on the efficiency of CO_2 dissociation and especially on the behavior of the vibrational excitation in a CO_2 discharge.

6. ACKNOWLEDGEMENTS

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