Evolution of active species downstream of a low pressure 90%Ar-10%(N_2-O_2) flowing microwave discharge

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In this study we present the evolution of the densities of different active species downstream of a low pressure surface-wave microwave discharge in a 5 mm diameter tube at different pressure conditions in the range 2-8 mbar and 90%Ar-10%(N_2-O_2) mixture compositions, as determined by self-consistent modelling. The different pressure conditions have been realized under the same flow condition. We show that the density of O-atoms behaves similarly for all the conditions under study, with a significant density decrease occurring at about 0.5 ms afterglow time. In the case of N-atoms the role of the pressure and mixture composition is more significant, i.e. the density in the afterglow decreases slowly in the higher N_2 content mixtures and lower pressures.

1. Introduction

Low pressure plasmas that contain N and O atoms and excited NO molecules have a wide range of applications, such as metal surface cleaning, medical sterilization, etching and grafting of polymers, thin film synthesis, to increase surface adhesion and textile material modification. In numerous cases instead of the active discharge region the afterglow is used, where the density of charge species is negligible. For sensitive materials it is also important to keep the gas temperature as low as possible. This can be achieved by using mixtures containing predominantly noble gases with some addition of molecular gases. Our study focuses on a mixture containing 90%Ar and 10%(N_2-O_2).

The afterglow of a surface-wave microwave discharge has been used in several applications and to study the interaction of plasma with surfaces and macromolecules [1-3]. This system has several advantages. The composition of the plasma required by a given application can be controlled by the discharge conditions and initial gas mixture composition. Further tuning possibilities are provided by the flowing afterglow, since the lifetime of the various species in the afterglow is very different [4].

The aim of our work is to determine the effect of the discharge pressure and the initial mixture composition on the evolution of species densities in the flowing afterglow of a surface-wave microwave discharge ignited in 90%Ar -10%(N_2-O_2).

2. Afterglow system

The afterglow system here investigated is based on a surface-wave microwave discharge generated in flowing gas in a 5 mm diameter tube of 50 cm length. Downstream the discharge – that has a length of a few cm depending on the mixture composition – an afterglow region develops with the active species being carried through by the gas flow. For application purposes the small diameter tube is further connected to a 36 mm diameter reactor, where the active species are transported.

The gas pressure in the system is measured at the entrance of the tube and in the reactor. Due to the small diameter of the tube and the relatively high gas flow rate used (for application purposes gas flow rates in the range of 500 sccm are used) a pressure drop occurs along the tube. Since the surfatron used for coupling the microwave power into discharge can be moved along the tube, discharges at different pressure conditions can be generated under the same flow condition and system configuration. Meanwhile, the length of the afterglow varies with the surfatron's position, which defines the composition of the gas entering the reactor.

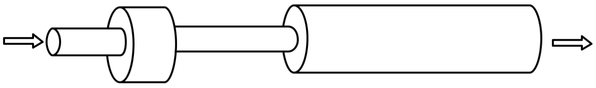

Figure 1. The flowing afterglow system consisted of a 5 mm diameter 50 cm long tube, with a movable surfatron encircling the tube, and a cylindrical reactor of 36 mm diameter.

For our investigations we have chosen three surfatron positions assuring in the discharge region pressure values of 8 mbar, 4 mbar and 2 mbar. A self-consistent kinetic model has been used to study the densities of the different species in the discharge region and their evolution along the afterglow up to the entrance of the reactor.

3. Modelling

The creation of the different species in the discharge is described with a zero dimensional self-consistent kinetic model that is based on the solutions of the electron Boltzmann equation for the microwave field, coupled to a system of rate-balance equations for the neutral and charged heavy species.

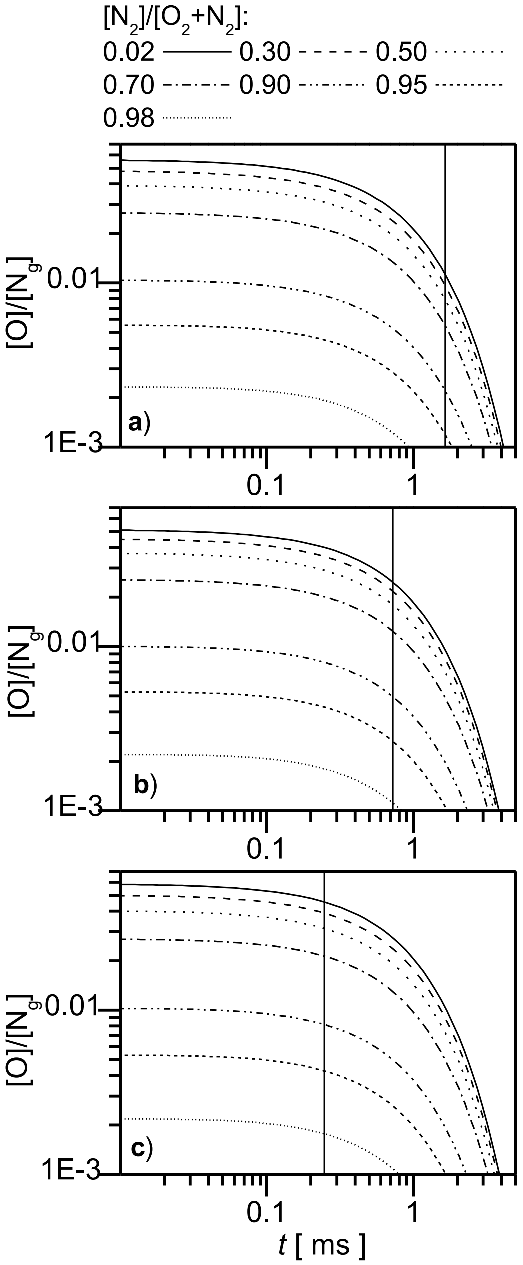

Figure 2. Relative density of oxygen atoms along the afterglow at different discharge pressure conditions: a) 8 mbar, b) 4 mbar and c) 2 mbar. The vertical line indicates the afterglow time when the species enter the reactor.

The concentrations obtained for the steady-state discharge are used as initial values to the early afterglow taking place in the same tube, where the same system of equations is solved in time under zero electric filed [4, 5].

In the model the following species are taken into account: Ar($^1S_0$, $^3P_2$, $^3P_1$, $^3P_0$, $^1P_1$), $O_2(X^3\Sigma_g^-,v)$, $O_2(a^1\Delta_g, b^1\Sigma_g^+)$, O($^3P$, ^1D), O_3, Ar^+, Ar_2^+, O_2^+, O^+, O^-, $N_2(X,v)$, N(^4S, ^2D, ^2P), N_2(A,B,B',C,a',a,w), NO_2(X, A), NO(X, A, B), N_2^+(X, B), N_4^+ and NO^+. The set of gas phase and surface reactions describing the kinetics of species as considered in the model has been presented in [5-7] and the references therein.

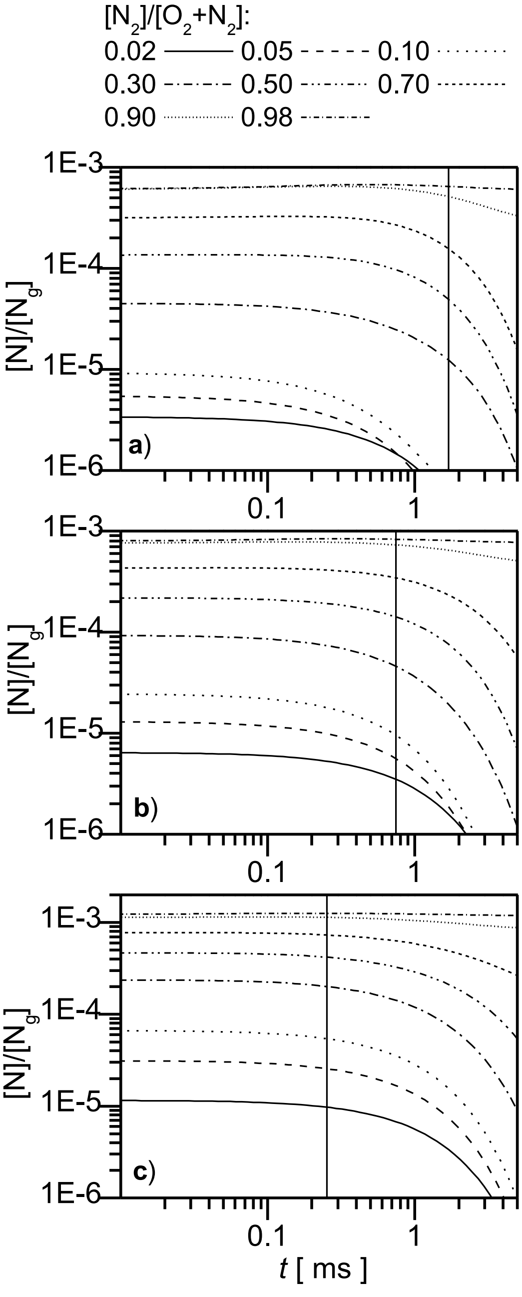

Figure 3. Relative density of nitrogen atoms along the afterglow at different discharge pressure conditions: a) 8 mbar, b) 4 mbar and c) 2 mbar.

According to our measurements performed on the investigated system, when using a gas flow rate of 500 sccm 11 mbar is achieved at the tube entrance and 1 mbar in the reactor. In accordance to this 8 mbar can be found at 15 cm from the gas entrance, 4 mbar at 35 cm and 2 mbar at 45 cm. As a consequence, the afterglow parameters concerning its length in space and time, and its medium pressure for the different discharge conditions are as follows: (*i*) p_{dis} = 8 mbar, p_{aft} = 4.5 mbar, L_{aft} = 35 cm, t = 1.7 ms, (*ii*) p_{dis} = 4 mbar, p_{aft} = 2.5 mbar, L_{aft} = 15 cm, t = 0.75 ms and (*iii*) p_{dis} = 2 mbar, p_{aft} = 1.5 mbar, L_{aft} = 5 cm, t = 0.25 ms.

4. Results

Here we present the evolution along the afterglow of the two most important species, namely the O-atoms and N-atoms. The relative densities of atoms are shown in the case of the three discharge pressure conditions for different gas mixture compositions, that is 90%Ar with 10% varying composition of N_2-O_2. A vertical line on each figure indicates the time when the species are expected to enter the reactor in case of that specific condition. We remind that with changing the position of the surfatron along the discharge tube we assure different gas pressure in the discharge region, and as a consequence the pressure in the afterglow region, as well as the length of the afterglow, i.e. the flight-time of species in this region also changes, as indicated in the previous paragraph.

The results show that in the case of O-atoms the pressure has only minor effect on the relative density and its evolution along the afterglow. On the other hand, in the case of N-atoms the relative density significantly increases in the discharge (t = 0 ms) with decreasing pressure, and the atoms survive longer in the afterglow.

In what concerns the relative densities of atoms at the reactor entrance, we identify the position of the surfatron, thus the length of the afterglow, as being crucial. In the case of the 8 mbar condition, where the length of the afterglow is around 1.7 ms, the relative density of O-atoms decreases with a factor of 5 from the discharge to the reactor entrance. Meanwhile in the case of 2 mbar condition – the length of the afterglow being 0.25 ms – the density decrease is minor. In the case of the N-atoms the density decrease in the afterglow also depends on the mixture composition, i.e. it becomes less significant with increasing the nitrogen content of the mixture.

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3. References

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