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PRELIMINARY RESULTS OF CALCULATIONS FOR A GLOBAL MODEL DESCRIBING PULSED MICROWAVE DISCHARGE PLASMA IN NITROGEN

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Abstract. This work concerns the modeling of the plasma of a pulsed microwave discharge in nitrogen. The goal is to determine the temporal variations of the densities of the respective plasma species. The ultimate goal is to adopt the methods developed for these calculations for discharges in polluted air or flue gas. A global numerical model is formulated. The object modeled is the plasma of a microwave torch operating at atmospheric pressure. The set of equations for the model consists of particle balance and power balance equations. The most important reactions between the plasma species are only taken into account. The rate coefficients for processes involving electrons are calculated based on a solution of the Boltzmann equation for the electrons. Diffusion and heat transfer are modeled with the aid of terms reflecting also the discharge geometry.

Keywords: pulsed microwave discharge, plasma, global model, torch.

INTRODUCTION

Industrial flue gases and car exhausts contaminate air with pollutants, such as NO_x, SO_x, CO_2 or volatile organic compounds, which are noxious to the environment and to the man. A promising way of decomposing the molecules of pollutants and converting them into harmless gases or reusable products offer non-thermal discharges. Recently, attention was turned to use of microwave discharges for that purpose. Especially pulsed microwave discharges are worth studying. For various reasons they may prove more effective for pollutant abatement than continuous wave discharges of the same mean power.

This paper is concerned with the modeling of the plasma of a microwave discharge in polluted air or flue gas. For preliminary evaluations of the modeling procedures a discharge in N_2 is considered. The object modeled is the plasma of a microwave torch [1] operating at atmospheric pressure. Such a torch is well suited for large-volume treatment of pollutants. Its design is simple and use of multiple torches can be accomplished easily.

THE MODEL

A global, that is a spatially averaged model is formulated. Although such a model can not provide spatial distributions of the plasma parameters, it is useful for establishing their scaling with the operating parameters of the discharge. For a discussion of the model see [2]. It is assumed that all densities are averaged over the plasma volume, the electric field is regarded as uniform and the effective field approximation is applicable. It is also assumed that due to the small size of the microwave plasma torch ionization is balanced mostly by the outward diffusion; the electron-ion recombination may be neglected [3]. The non-equilibrium plasma of the microwave plasma torch is characterized by two temperatures: the heavy particle temperature T and the electron temperature T_e. The vibrational temperature T_v is assumed to be equal to T. Radiation is neglected.

The electrical power input in the plasma is dissipated by the primary electrons in collisions with the molecules of the gas. Electron-ion pairs and free radicals (N atoms in a discharge in nitrogen) are produced through electron impact ionization and dissociation. The radicals, together with the secondary electrons and ions, may subsequently react with the pollutant molecules, leading to their decomposition.

For preliminary calculations we limit the number of species and primary reactions to the most important ones. We use a subset of reactions specified in [1], namely:

- electronic excitation

$$N_2 + e \rightarrow N_2^* + e \qquad (k_1) \qquad (1)$$

- quenching of electronically excited molecule states by N atoms

$$N_2^* + N \rightarrow N_2 + N \qquad (k_2) \qquad (2)$$

- ionization by electron impact

$$N_2 + e \rightarrow N_2^+ + e + e \qquad (k_3) \qquad (3)$$

- dissociation

$$N_2 + e \rightarrow N + N + e \qquad (k_4) \qquad (4)$$

- neutral recombination

$$N + N + N_2 \rightarrow N_2 + N_2 \qquad (k_5) \qquad (5)$$

The species taken into account are: the nitrogen molecules in the electronic ground state, the concentration of which is assumed to be almost equal to the overall concentration of N_2 molecules (n), electronically excited nitrogen molecules N_2^* (n_x), nitrogen atoms N (n_a), electrons e (n_e) and molecular nitrogen ions N_2^+ (their concentration equal to n_e owing to the plasma quasi-neutrality). The rate coefficients for the processes involving collisions with electrons depend on the form of the electron energy distribution function f_e for the discharge plasma. They are determined in the course of calculations by solving the Boltzmann equation for the electrons and calculating the rates as cross section averages over f_e. The ELENDIF code [4] is used for that purpose. The rate coefficient k_5 is taken from [5].

We consider a microwave plasma torch as described in [1], with a length $L = 5 \times 10^{-2}$ m and a radius $R = 2 \times 10^{-3}$ m, located in a quartz tube with a radius $R_{out} = 4 \times 10^{-2}$ m. For all species present in the plasma concentrations averaged over the plasma volume $V = \pi R^2 L$ are considered. We attribute all losses to the diffusion and forced convection. The discharge is fed with microwave power pulses superimposed on a continuous wave microwave background.

The set of equations used in the global model consists of:

- particle balance equations,
- power balance equations.

We write the particle balance equations according to reactions (1)-(5) as

$$dn_x/dt = k_1\, n\, n_e - k_2\, n_x\, n_a \tag{6}$$

$$dn_a/dt = 2k_4\, n\, n_e - 2k_5\, n\, n_a\, n_a \tag{7}$$

$$dn_e/dt = k_3\, n\, n_e - (2D_a/3R^2)\, n_e \tag{8}$$

where D_a is the coefficient of ambipolar diffusion. In the first approximation we neglect the flow.

The electron energy balance equation is written as

$$\frac{3}{2} k n_e \frac{dT_e}{dt} = P_v - n_e v_{iz} \varepsilon_L \tag{9}$$

where P_v is the density of the microwave power absorbed by the plasma, k is the Boltzmann constant and ε_L is the plasma energy loss per electron-ion pair created due to all electron-neutral collision processes (cf [2]).

The energy balance equation for the heavy particles reads

$$\left[\frac{7}{2} kn + \frac{5}{2} kn_a\right] \frac{dT}{dt} = \frac{3}{2} \delta_e v n_e k (T_e - T) - \frac{2\lambda}{3R^2} T \tag{10}$$

where v is the effective collision frequency for momentum transfer, δ_e is the fraction of energy lost by an electron in a collision with a heavy particle, and λ is the thermal conductivity. Values of the coefficients δ_e, λ and D_a are taken from the literature, v is calculated. Equations (6)-(10) for 5 unknowns n_x, n_a, n_e, T and T_e form the set to be solved.

CALCULATION METHOD

To solve the set of ordinary differential equations (6)-(10) one must determine, first of all, the initial values of the unknowns for given discharge conditions corresponding to the cw microwave background. This is done by putting $dn_i/dt = dT_e/dt = dT/dt = 0$ ($i = x,a,e$) in the respective equations and solving the resultant set of nonlinear algebraic equations. For the model presented, some equations may be solved independently of the others.

First, for given T, taken from experiments [1], the electric field reduced intensity E/n is calculated from the balance between ionization and diffusion (8). This is done by iteration, with the Boltzmann equation for the electrons solved in each step using the ELENDIF code. Next, the electron concentration n_e is calculated from the energy balance equation for the heavy particles (10). Simultaneously the microwave power density P_v is determined based on the formula

$$P_v = e\, n_e\, v_d\, (E/n)\, n \qquad (11)$$

where v_d is the electron drift velocity. That way one obtains, for given gas temperature T, the electric field reduced intensity E/n, the electron concentration n_e , the electron temperature T_e and the microwave power density P_{v0} characterizing the initial conditions. Finally, particle balance equations for the stationary initial state are solved, providing the initial values for populations of the electronically excited nitrogen molecules N_2^* (n_x) and nitrogen atoms N (n_a). The rate coefficients are again determined based on the solution to the Boltzmann equation for the electrons.

Superposition of relatively narrow microwave pulses on a microwave cw background, with the pulse peak power density P_{v1} exceeding several times the background power density P_{v0} (in practice the background is needed to avoid problems with discharge ignition), does not change significantly the gas temperature. Therefore, in preliminary calculations for the pulsed input power the gas temperature T may be regarded as constant. Then it is only necessary to solve the set of differential equations (6)-(9). The respective rate coefficients in these equations may differ by several orders of magnitude. For solving this set the Gear's method for stiff equations is used.

In each step of integration the instantaneous value of E/n is found from formula (11) by iteration. This is justifiable on assumption that the efficiency of power transfer from the microwave field to the plasma is constant. Any rise of P_v must result in a rise of E/n, as the electron concentration can not change instantaneously. The rise of E/n causes an increase of the ionization rate, resulting in an increase of the electron concentration and decrease of E/n. In the numerical code each change of E/n is followed by solving the Boltzmann equation for the electrons and updating the rate coefficients.

RESULTS OF CALCULATIONS

Calculations were made for a 2.45 GHz microwave discharge in nitrogen at atmospheric pressure. The following initial data were assumed: continuous wave background with a power density P_{v0} = 39 Wcm^{-3}, gas temperature T = 1500 K, pulse repetition rate of 1 kHz, rectangular pulses of length and peak power equal to t_p = 10 μs, P_{v1} = 195 Wcm^{-3}, and t_p = 6 μs, P_{v1} = 273 Wcm^{-3}. Hence the mean power density was equal to $P_{v\,mean}$ = 40.6 Wcm^{-3} and $P_{v\,mean}$ = 40.4 Wcm^{-3}, respectively.

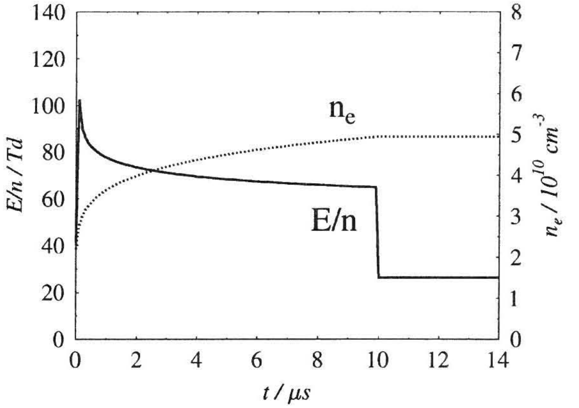

Fig. 1. Variation of the electric field reduced intensity *E/n* and electron concentration n_e for the first pulse; t_p = 10 μs, P_{v1} = 195 Wcm^{-3}.

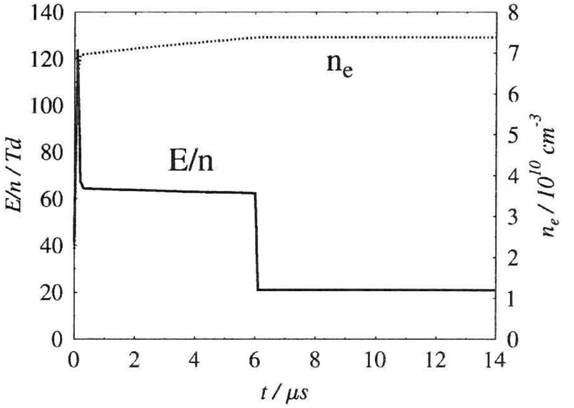

Fig. 2. Variation of the electric field reduced intensity *E/n* and electron concentration n_e for the first pulse; t_p = 6 μs, P_{v1} = 273 Wcm^{-3}.

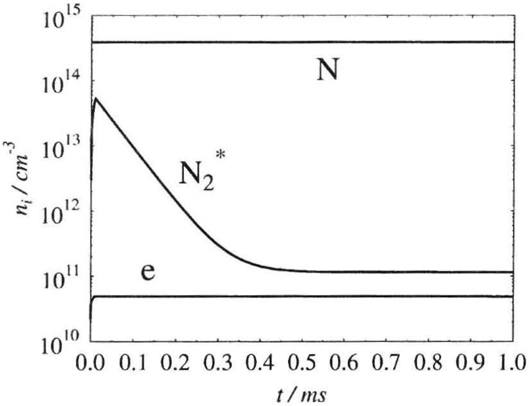

Fig. 3. Variation of the plasma species concentrations during the first cycle of pulsed operation; conditions as for Fig. 1.

Figures 1 and 2 show changes of *E/n* and n_e at the very beginning of pulsed operation. As it is seen, a stepwise rise of the power density input to the plasma causes a rapid rise of the electric field. The resulting rise of the ionization rate causes the electron concentration increase, which in turn allows *E/n* to decrease gradually. Understandably, the variation of the electric field intensity is steeper for the pulse of higher power. The calculations showed a similar behaviour for the electron temperature; its peak value reached almost $19 \cdot 10^3$ K and $24 \cdot 10^3$ K for conditions of Figs. 1 and 2, respectively, to drop to about 10^4 K at the pulse end, and $8 \cdot 10^3$ K behind the pulse.

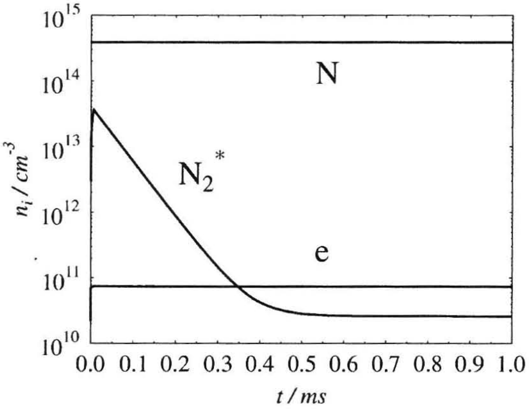

Fig. 4. Variation of the plasma species concentrations during the first cycle of pulsed operation; conditions as for Fig. 2.

Population changes for the plasma species considered in the calculations are shown in Figs. 3 and 4. The number density of the N atoms remains almost unaffected by the pulse, while that of electronically excited nitrogen molecules increases rapidly during the pulse, to decrease behind it to a low level in about 0.4 ms. The number densities of the electrons and N_2^+ ions remain constant behind the pulse.

CONCLUDING REMARKS

It should be emphasized that these results are correct only within the limits imposed by the model adopted. For more precise calculations a more comprehensive global model is needed. Such a model is now under development. The numerical code for this model is based on a CORONA code developed for modeling the plasma chemistry for streamer corona discharges [6]. This code, making extensive use of a PLASMAKIN library [7], may easily handle any reasonable number of species and reactions.

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WSTĘPNE WYNIKI OBLICZEŃ DLA GLOBALNEGO MODELU PLAZMY IMPULSOWEGO WYŁADOWANIA MIKROFALOWEGO W AZOCIE

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Streszczenie. Praca dotyczy modelu plazmy impulsowego wyładowania mikrofalowego w azocie pod ciśnieniem atmosferycznym, którego celem jest wyznaczenie stałych czasowych zmian gęstości składników plazmy co umożliwi zastosowanie użytych metod dla obliczeń w zanieczyszczonym powietrzu lub gazach odlotowych. Wykorzystano model globalny składający się z zestawu równań opisujących bilans cząsteczek i energii dla najważniejszych reakcji. Współczynniki szybkości reakcji z udziałem elektronów oblicza się z wykorzystaniem równania Boltzmana. Przepływ, dyfuzję i wymianę ciepła uwzględniono wprowadzając człony modelujące w uproszczony sposób dla konkretnej geometrii wyładowania.

Słowa kluczowe: impulsowe wyładowanie mikrofalowe, plazma, model globalny, torch.