Time-Resolved Optical Emission Spectroscopy of a Surfaguide Flowing Gas Microwave Discharge

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Pulsed surfaguide microwave discharge operating at 2.45 GHz in different Ar-N_2 and Ar-N_2-H_2 gas mixtures was investigated by time-resolved optical emission spectroscopy. The emission lines of Ar, N_2, N_2^+, N, and H were studied. Besides this, the N_2 vibrational temperature as well as the gas temperature were also analyzed. The measurements confirm that the efficient N_2 dissociation in the discharge depends mainly on the presence of the nitrogen molecular ions (dissociative recombination). It was also found that the negligible addition of H_2 (~ 0.2 %) dramatically changes the mechanism of N_2 dissociation.

Microwave surfaguide discharges (MSGD) generated in the flowing gas promote so called “remote plasma processing” usually performed in the post-discharge area. Such plasma processing is of a great interest because the chemically active radicals can be easily delivered to the post-discharge region and used e.g. for surface treatment [1, 2]. The actual chemical composition in the post-discharge, however, is defined in the plasma area, where molecular dissociation or/and recombination, as well as the other processes, happen. The pulse mode of operation further enhances the efficiency of these processes [3].

This work is focused on time-resolved study of the MSGD plasma by means of optical emission spectroscopy (OES). The MSGD was generated at 2.45 GHz using a pulsed microwave generator. The discharge was sustained in the quatrz tube with a diameter of 1.4 cm, and length of 24 cm cooled down by the oil flow (Fig. 1). The pulse duration of 600 μs, and duty ratio of 0.4 were utilized. The OES measurements were performed using an Andor SR750 monocromator equipped with an Andor iStar740 series ICCD camera through an optical fiber. For the gas temperature (T_g) determination the rotational analysis of the First Positive Band of N_2 was undertaken following [4].

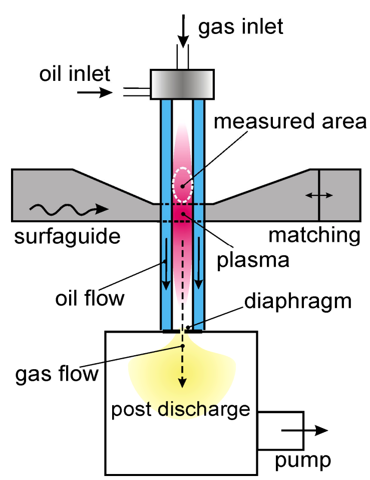

Fig. 1: Schematics of the microwave discharge studied in this work.

As a result of this work it was found that the dissociative recombination process plays a key role in the N_2 dissociation in the MSGD, i.e. it is directly defined by the presence of the N_2^+ ions in the discharge ($e + N_2^+ \rightarrow N(^4S) + N(^4S)$) [5]. This is illustrated in Fig. 2, where the typical Ar, N_2, N_2^+, and N emission spectra are presented at two different moments of time during the plasma pulse. It was found, that the N peak emission increases during nearly 200 μs after the beginning of the plasma pulse, and saturates afterwards. This peak with a certain delay follows the time-evolution of the N_2^+ emission peak (see Fig. 3(a)). During the pulse the T_g increase might be as high as 600 K (at the max. applied power). Generally, the saturation times of the different studied emision lines are related as follow (see Fig. 3(a)):

$$\tau_{Ar} \approx \tau_{N_2} < \tau_{N_2^+} < \tau_N \qquad (1)$$

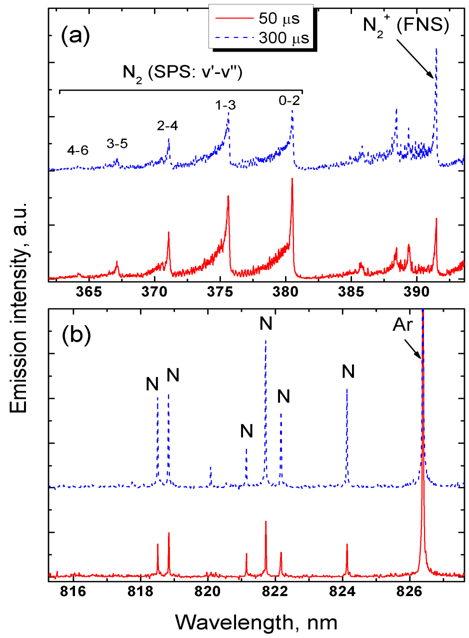

Fig. 2: The appearance of the N_2 and N_2^+ (a), as well as Ar and N (b) emission peaks/bands taken at the beginning and in the middle of 600 µs plasma pulse. N_2 content = 5%.

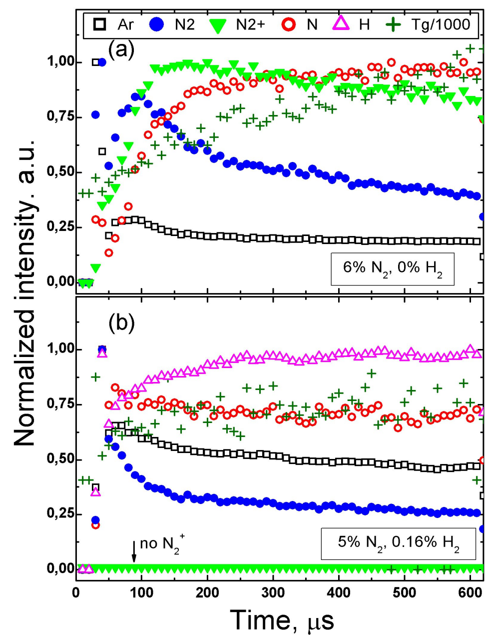

Fig. 3: Time-evolution of the (separately) normalized Ar, N_2, N_2^+, N, and H peaks along with T_g measured in different gas mixtures: (a) 6% N_2, 0% H_2, (b) 5% N_2 , 0.16% H_2.

At the same time, after a small addition of H_2 to the gas mixture, the production of N_2^+ gets almost totally suppressed (Fig. 3(b)) which, we believe, is due to the quenching of the $N_2(A)$ metastable level by molecular hydrogen (dissociation energy is 4.5 eV). As a result, the N peak emission intensity drops by nearly one order of magnitude (so does the production of N atoms, not shown), which points out the importance of the $N_2(A)$ metastables in the nitrogen ionization ($e + N_2(A) \rightarrow N_2^+ + 2e$) following by the N_2 dissociative recombination. In the presence of hydrogen the dissociation rate of N_2 is supposed to drop down to the level defined by the direct electron impact dissociation ($e + N_2\,(X) \rightarrow 2\,N(^4S) + e$), and the effect of gradual saturation of the N emission peak disappears, as illustrated in Fig. 3.

It was also found that the minor additions of H_2 affect the gas temperature values in saturation, presumably as a result of the suppression of disociative recombination, which is known to be an exothermic reaction [6]. This effect may provide a way for the gas temperature control during the plasma processing if the high level of the N_2 dissociation in the discharge is not required.

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