MEASUREMENTS OF THE ATOMIC OXYGEN CONCENTRATION IN A PULSED OZONIZER DISCHARGE BY LASER INDUCED FLUORESCENCE

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ABSTRACT

Ozone synthesis utilizes the silent discharge (dielectric-barrier discharge) which usually consists of a large number of isolated streamers. Here a discharge is described which produces a single streamer. Measurements of the current, the light emission at 777nm of an OI-transition and of the concentration of O($^5S^0$) using laser induced fluorescence are reported.

1. INTRODUCTION

Numerical calculations show that the efficiency of ozone generation is governed by two factors: the efficiency of generating atomic oxygen and the rate by which atomic oxygen is converted into ozone. The main source of atomic oxygen is the dissociation of O$_2$ by electron impact:

\[ e + O_2 \rightarrow e + O + O^* \]  \hspace{1cm} (1)

where O* represents atomic oxygen in the ground state or in excited states, such as O($^5S^0$) and O($^5P$). The excited states O($^5S^0$) and O($^5P$) are predominantly deexcited by dissociative excitation transfer /1/:

\[ O^*(^5S^0,^5P) + O_2 \rightarrow O + O + O + O + O \] \hspace{1cm} (2)

According to the reaction

\[ O + O_2 + M \rightarrow O_3 + M \hspace{1cm} M = O, O_2, O_3 \] \hspace{1cm} (3)

one should obtain one ozone molecule for every oxygen atom created, but this is valid only for small atom concentrations ($n_0/n_{O_2} \ll 10^{-3}$), low gas temperatures, and zero background ozone concentration /2/.
The aim of our study is to measure local plasma properties in order to demonstrate that the processes listed above are of main importance in ozone synthesis.

2. EXPERIMENTAL

To measure the local density of atomic oxygen in the excited state $^5S^0$ using laser induced fluorescence at $\lambda=777nm$ we have to produce a single streamer at a fixed position and time. The jitter between laser and discharge should be lower than 1ns. The experimental arrangement of Fig.1 is appropriate. A Marx-generator with avalanche transistors (8kV, rise time 5.5ns) serves as power supply. A coaxial set-up is used as ozonizer (Fig.2). The electrons initiating the breakdown are produced by field emission. As feed gas we use pure oxygen at a pressure of 3 to 8hPa. At this pressure the excited states $O(^5S^0)$ and $O(^5P)$ live longer than a few nanoseconds. As light source a $N_2$-laser pumped dye laser (oxazine 750) with a repetition rate of 10Hz is used. A Box-Car averager monitors the scatter signals.

3. RESULTS

The discharge current (Fig.3) of a few nanoseconds duration and a formative time of 11ns was observed. The current density derived from radial light emission profiles (see fig.4) is proportional to the pressure (Fig.5). The duration of the light emission has been found to be typically 10ns. The particle density of the excited state $O(^5P)$, which is proportional to the light emission, is independent of the streamer height z. Using laser induced fluorescence we obtain the absolute density of atomic oxygen in the excited state $^5S^0$ (Fig.6). As the current density $n_O(^5S^0)$ is proportional to the pressure. With time resolved measurements of the light emission and the laser induced fluorescence we can estimate the rate coefficient of reaction (2) to be nearly $4 \times 10^{-10} \text{ cm}^3/\text{s}$. 

-334-
4. THEORY

To get informations about the concentrations of atomic oxygen in the ground state and of ozone from the measured concentration of atomic oxygen in the excited state $O(5\text{S}_0^0)$ we use a reaction scheme including 28 reactions between 8 different reaction partners (ground states of $O$, $O_2$, $O_3$, the excited states $O(5\text{S}_0^0)$, $O(5\text{P})$ and the charged particles $e$, $O^-$, $O^+$). The evolution and decay of different particle species at a pressure of 8hPa shows Fig. 7. The numerical calculations are in a good agreement with the measurements. The relative densities of atomic oxygen in the ground state and of ozone in a streamer are about $10^{-4}$.

REFERENCES


Fig. 1: Arrangement for the laser induced fluorescence
Fig. 2: Ozonizer

Fig. 3: Current pulse of a streamer in oxygen recorded with current probe TEK CT-1
(amplitude: 1A/small div., time: 2ns/small div.,
pressure: 5.3kPa, gap spacing: 4.0mm)
Fig. 4: Radial profile of the particle density $n_O(5P)$
(gap spacing: 5.0mm)

Fig. 5: Temporal maximum of the current density
(gap spacing: 5.0mm)
Fig. 6: Radial profil of the density of atomic oxygen in the excited state $^5S^0$.

Fig. 7: Relative particle densities in a pulsed oxygen discharge.