Atomic oxygen density distributions in micro cavity plasma arrays measured by helium state enhanced actinometry (SEA)

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Abstract: Atomic oxygen density distributions were determined in a micro structured dielectric barrier discharge at atmospheric pressure. Since typical diagnostics such as two photon absorption laser induced fluorescence cannot be applied due to limited optical access, a new diagnostics of optical emission spectroscopy, helium state enhanced actinometry, was developed. After a benchmark against TALIF measurements was performed, the diagnostics were applied to a micro cavity plasma array.

Keywords: atmospheric pressure, micro discharges, dielectric barrier, atomic oxygen, optical emission spectroscopy

1. Introduction

Micro discharges operated at atmospheric pressure are known for their wide range of potential applications, such as the treatment of volatile organic components (VOCs) or plasma catalysis [1,2]. Key to these applications are reactors in which gases are decomposed, reactive species are generated, and the plasma interacts with catalytic surfaces. One class of discharges that offers this are micro cavity plasma arrays [3]. These consist of thousands of cavities in the 100µm range in which the plasma is ignited. High electric fields of about 30 kV/cm [4] and the possibility to use a catalytic surface in direct contact with the plasma provide optimal conditions for fundamental research on plasma catalysis. To optimize or monitor the processes, diagnostics are required. However, due to their small dimensions, micro discharges have the disadvantage of limited optical access. This makes the application of diagnostics very challenging. For example, the use of laser spectroscopic methods to determine densities of reactive species such as atomic oxygen is not possible. Optical emission spectroscopy (OES), or more precisely actinometry, offers an alternative. Here, atomic oxygen densities can be determined directly from the emission of the discharge. Nevertheless, unlike laser spectroscopic methods as for example two photon absorption laser induced fluorescence (TALIF), only excited states can be observed with OES methods. For this reason, a model is needed that links the measured excited state with the ground state density. Since several assumptions are necessary for this, there may be some uncertainties. For this reason, helium state enhanced actinometry (SEA) was introduced as an extension of the classical actinometry approach to reduce the uncertainties and to provide a valid alternative to TALIF measurements in plasmas difficult to access [5,6]. In the following, the SEA diagnostics will be explained and verified with a benchmark. Subsequently, the method will be applied to micro cavity plasma arrays and the results will be discussed.

2. Helium state enhanced actinometry (SEA)

The classical actinometry approach uses two spectral lines, an oxygen line (λ = 844nm) and an argon line (λ = 750nm). The atomic oxygen density can be determined from their intensity ratio. However, this method is based on the corona model, which assumes that only electron impact excitation from the ground state and de-excitation by spontaneous emission takes place. In the case of oxygen, the excitation can partially occur through a dissociative channel. This is not taken into account in the model, thus the densities are overestimated. By using another oxygen line (λ = 777nm) and the Boltzmann solver BOLSIG+, the dissociative channel can be included. Furthermore, this extension offers the possibility to determine the mean electron energy. Therefore it is called energy resolved actinometry (ERA). Even if the dissociative channel is taken into account, uncertainties can arise here as well, since the atomic state associated with the 777nm line can be excited not only by electron impact reactions but also by metastable atoms or molecules. In this work, therefore, a helium line (λ = 706nm) is used instead of the 777nm oxygen line. This has several advantages. First, the uncertainties due to metastables are reduced. Secondly, a larger range of the electron energy distribution function (EEDF) is probed, which improves the measurement of the mean electron energy. Two intensity ratios can be formed from the intensities of the three spectral lines:

$$\frac{I_{844}}{I_{750}} = \frac{f_{O_2}}{f_{Ar}} \frac{\nu_{844}}{\nu_{750}} \frac{a_{844}}{a_{750}} \frac{2r_O k_{844,d}(\varepsilon) + k_{844,de}(\varepsilon)}{k_{750,d}(\varepsilon)}$$

$$\frac{I_{706}}{I_{750}} = \frac{f_{He}}{f_{Ar}} \frac{\nu_{706}}{\nu_{750}} \frac{a_{706}}{a_{750}} \frac{k_{706,d}(\varepsilon)}{k_{750,d}(\varepsilon)}$$

These depend each on the fraction *f* of the respective species in the total gas flow, the photon energy *hν*, the optical branching ratio *a*, the dissociation degree *r* and the mean electron energy-dependent effective excitation rates *k(ε)* calculated by BOLSIG+. The direct electron impact excitation is indexed by *d* and the dissociative excitation by *de*. The ratios result in a system of equations with two unknowns, the degree of dissociation and the mean electron energy. It can be solved to determine both parameters. Figure 1 shows a benchmark of measured atomic oxygen densities against TALIF measurements depending on the applied voltage in the COST micro plasma jet [7]. As expected, the classical actinometry

approach significantly overestimates the density. In contrast, taking into account the uncertainties of the different diagnostics SEA shows good agreement with the TALIF measurements. Further details about the SEA diagnostics can be found in [5].

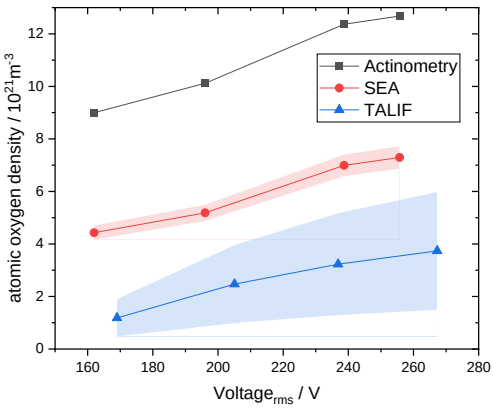

Fig. 1. Atomic oxygen density determined by various methods as a function of applied voltage [5].

3. Experimental Setup

SEA is now applied to a micro cavity plasma array. It consists of a grounded electrode, a dielectric and a powered metal grid with thousands of cavities with diameters in the 100µm range which can be described as single surface DBDs. A quartz cover closes off the reactor and allows a gas flow along the cavities. The emission of the plasma can be observed using an ICCD camera and a tuneable filter (550-1000nm). All components of the experimental setup are shown in figure 2. The setup allows temporally and 2d spatially resolved measurements of the atomic oxygen density.

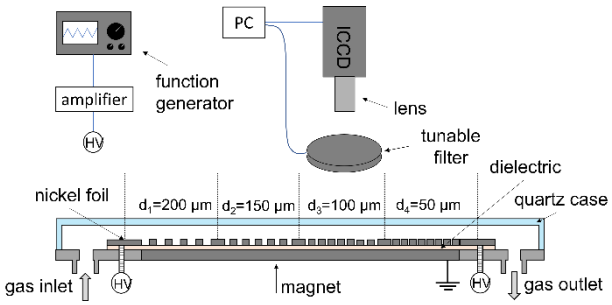

Fig. 2. Experimental setup [8].

4. Results and discussion

The setup offers the advantage of being able to image a complete subarray structure onto the ICCD chip. Thus, the atomic oxygen density can be calculated for each pixel. It shows that oxygen distributions can be observed over the entire array as well as within individual cavities. Especially ring structures within the cavities are noticeable. These can be explained by higher electric fields at the edges of the cavities. This can be examined in more detail by accumulating over all cavities of a sub-array structure, resulting in an average cavity (fig. 3). In this way, for example the influence of different cavity diameters on the density can be investigated. In addition, time-resolved measurements are possible. Typically, the micro cavity plasma array is operated with a triangular voltage with an amplitude of up to 800V at a frequency of 15kHz. One period can be divided into two different phases. The increasing potential phase (IPP) and the decreasing potential phase (DPP). The difference between the phases is visible in the density and energy measurements. During the IPP the electrons move outwards and a ring structure is formed. In the DPP, on the other hand, the electrons move inward. The high concentration in the centre of the cavity results in higher atomic oxygen densities and mean electron energies. Overall, the measurements show very high dissociation degrees of almost 100%. This fits to voltage variations in which the density remains constant independent of the applied voltage. A variation of the oxygen admixture also shows that oxygen is nearly completely dissociated at low admixtures and the density saturates only at higher admixtures of several percent.

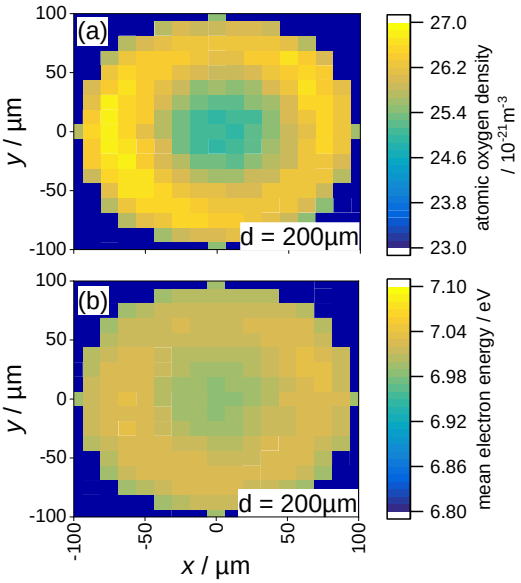

Fig. 3. 2D resolved oxygen density (a) and mean electron energy (b) for a single 200µm cavity [8].

5. Acknowledgement

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6. References

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