Experimental analysis of the temperature conditions of a miniaturized atmospheric pressure plasma jet during thin film deposition

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Abstract: The present work is focused on a temperature analysis of the neutral gas component in an miniaturized atmospheric pressure plasma jet, its radial profile, and the heat impact on the surfaces of the jet capillaries and substrate below the jet. A local, stationary substrate treatment demonstrates a dependence on the radial distance from the axis of the jet and surface temperature.

Keywords: SiO₂, OMCTS, APPJ, PECVD, surface temperature.

1. Introduction

Miniaturized atmospheric pressure plasma jets (APPJ) represent an interesting technique for local thin film deposition under normal pressure, in particular for the covering of 3-D forms, e.g. the inner walls of wells, trenches or cavities [1-6]. The application of such devices for the deposition of SiO₂ films with PE-CVD processes using liquid siloxanes has been reported before [7-11]. A significant enhancement of the film properties is achieved, when the films are deposited with the recently observed homogenized plasma mode (locked mode) of the APPJ, which develops under special conditions and is characterized by regularly ordered and moving discharge filaments [12-14].

It is a well-known fact that the substrate temperature has a critical impact on the composition of the deposited films [13, 15, 16]. Nevertheless, a rigorous temperature analysis focused on the APPJ shows still new aspects of deposition experiments. At higher substrate temperature, the incorporation of volatile components into the film is blocked. On the contrary, the deposition rate increases, and the spectrum of chemical composition is broader, if the surface temperature increases. The control of temperature supports the surface diffusion and leads to an improvement of the lateral homogeneity of films [8, 17]. Moreover, temperature gradients in the substrate can cause inhomogeneous mechanical relaxation stress in films after deposition. Therefore, the non-optimized films that have been deposited under high temperature gradient conditions peel off and show relaxation deformations.

2. Experimental set-up and operating characteristic

The particular plasma source under study here is a capacitively coupled capillary jet (27.2 MHz, applied power 5 - 20 W) with two outer ring electrodes around a quartz capillary between which Ar flows at rates between 10 and 150 sccm (fig. 1a). The distance between electrodes is 7 mm and their temperature is stabilized at 40 °C by a water coolant. The surrounding laboratory temperature stays constant at 23 °C. Downstream the active discharge region a thin film producing compound can be added in small quantities. In our experiments, octamethylcyclotetrasiloxane (OMCTS, 0.05 g/h), diluted in 2 slm Ar, was added for thin film deposition experiments via inner ceramic capillary. The cross-sections of the inner and outer flow are 0.8 mm² and 1.1 mm², respectively.

The plasma of the APPJ during the presented experiments is in locked mode consisting of self organized microfilaments. In this work the locked mode of four microfilaments has been applied (see fig. 1b, c). The filaments rotate at the velocity of 130 mm/s. Visualization of the rotational movement of the symmetric filament pattern requires a minimal shutter speed of 1000 fps. The establishment of the locked mode depends on suitable relations between RF power and argon flux. The operation characteristic (power vs. flux) has been determined for the jet.
geometry (see fig. 2). For the correlation of these two quantities the following empirical relation is received:

\[ P \sim \frac{1}{\sqrt{Q}} \]  

[1]

Here, \( P \) is the RF power and \( Q \) is the flow rate of argon. Along the \( P-Q \) characteristic, no significant changes of the locked mode have been observed, while the temperature of the system varies drastically. Outside of the confidence limits of the operating characteristic, no locked mode could be observed.

3. Material and Methods

Two methods for the temperature analysis have been applied. Both of them achieved a comparable accuracy of the temperature measurements of 0.1 °C. The fiber optic thermometry system has been used for the measurement of the neutral gas temperature [18]. The principle of the probe is based on the fluoro optic effect and does not influence the electrical circuits of the APPJ and therefore causes a minimal perturbation of the plasma. The thin geometry of the probe enables a high spatial resolution (0.5 mm) of the gas temperature. The temperature has been measured along the symmetry axis of the APPJ from the nozzle to the substrate.

For the measurement of the surface temperature an IR-camera has been used. The camera achieves a comparable spatial resolution of 0.6 mm and it operates in the MIR region. The correct contact-less measurement of temperature requires the IR emissivity of the surfaces [16, 19]. For the present measurements the emissivity of the outer quartz capillary (0.82) and inner ceramic capillary (0.75) has been estimated and the corresponding temperature has been corrected. The APPJ has been monitored by IR-camera from two views: The side view provides the axial temperature profiles of the outer jet capillary. The axial view provides the radial temperature profiles of both, inner and outer capillaries.

The morphology of the deposited films has been explored by scanning electron microscopy. The analysis enables to estimate the differences between the structure of films deposited under different temperature conditions. Particularly this method makes the observation of fracture patterns from defined indentation of films possible, which is useful for qualification of the relaxation processes in the films after deposition.
4. Temperature analysis

The IR temperature images of the APPJ have been collected under stationary conditions related to the operating characteristic of the locked mode. The present data are evaluated for two exemplary conditions denoted as A and B in fig. 2. The axial view in the APPJ under condition A is shown in fig. 3. The corresponding temperature profile is plotted in fig 4 compared to the conditions B.

A maximal surface temperature of about 200 °C on the end of inner ceramic capillary was obtained, which is approximately 100 °C hotter than the outer capillary which is cooled partially by the electrodes. The outer capillary achieves its maximal temperature in the space between the electrodes. The effect is caused by the smaller heat capacity of the inner capillary and its good isolation against the conductive heat exchange. The additional reason for the maximal axial heating might be the fast rotation of the hot filaments, which limit the heat dissipation of the gas cylinder encapsulated inside of the jet.

The RF power dependence of the temperature shows the conservation of the temperature conditions in the center of the jet, while the temperature of the outer capillary increases by an increase of the RF power. The possible explanation of the effect is based on the equilibrium between the reactive heating and dissipation losses in the volume unit conserving the thermal energy density in the APPJ at the locked mode. Each increase of the applied power extends the ionization volume, but it does not increase the temperature in the center of the jet (fig. 4).

The heating of the outer parts of APPJ by increased RF power leads to an increase of the axial temperature gradients, which present a higher stress to the capillary. However, it suppresses the radial temperature gradients, which are considered disadvantageous for a stable and homogeneous film deposition.

5. Influence of the surface temperature on the films

Generally, the correct monitoring of the surface temperature during a deposition poses some technical problems. The conductive methods of the temperature mea-
measure influence the heat balance of the deposition process. The contact-less measurement of the surface temperature requires information about the IR emissivity of the film which changes with the deposition. An example of a correct solution has been discussed previously [20].

Here, a heat conductive substrate (aluminum foil) has been chosen. Its temperature was controlled by means of an external thermostat in order to quantify the influence of surface temperature. We can assume a heat accommodation coefficient of 1 for this system and we approximate the real surface temperature by the temperature of the thermostat.

Plasma polymer films of SiO₂ have been locally deposited at two different surface temperatures (20 and 70 °C) and under constant deposition conditions A. The temperature difference between gas and surface is 70 °C. The temperature of the deposition coefficient of 1 for this system and we approximate the real surface temperature by the temperature of the thermostat.

6. Conclusion

The temperature conditions of a miniaturized APPJ have been analyzed in the regime of four-filament locked mode during deposition of SiO₂-like thin films. The measurement revealed that at the inner capillary feeding the plasma with a mixture of precursor the highest temperature of the plasma source develops. Its temperature reaches 200 °C and more at 10W. Moreover, the inner capillary of APPJ can be used as a temperature probe for the central gas flow during the deposition, due to the local thermal equilibrium in the centre of the jet.

The increase of RF power supports lateral heat transfer and minimizes radial temperature gradients in the APPJ. Generally, the deposition at lower temperature gradients on the surface produces films, which are mechanically highly stable.

The temperature control of the outer capillary at 200 °C and heating of the substrate to 100 °C can improve the mechanical and structural quality of the SiO₂-like film deposition.

References