Surface morphology analysis of SiO$_2$ thin film growth on polymeric substrate

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Abstract: The growth front evolution of silica-like thin films deposited in a high current dielectric barrier discharge was investigated using Atomic Force Microscopy subsequently processing the data within the frame work of the Dynamic Scaling Theory. The analysis indicates a transition from a non-self-affine to a self-affine surface morphology. Within the measured thickness range no roughness development is observed.

Keywords: dynamic scaling theory, AFM, surface morphology, surface roughness

1. Introduction
Atmospheric pressure plasma enhanced chemical vapour deposition (AP-PECVD) is recently gaining much interest as a tool to produce functional films. The small footprint and the absence of a complex and expensive vacuum system make the technology attractive, from technological and economic perspective. Thin silica-like films with excellent properties can be deposited by the AP-PECVD process from roll-to-roll. Because of their excellent gas diffusion barrier properties (WVTR < 5 x 10$^{-3}$ g m$^{-2}$ day$^{-1}$). The films can be used in a wide range of applications for example for the protection of flexible electronics, e.g. solar cells and food, beverage and medical packaging [1].

Earlier investigations were carried out using a parallel plate AP-PECVD reactor [2] and it was shown that smooth films up to 350 nm thick can be deposited having a comparable roughness as the polymer substrate. The formation of smooth films was extensively discussed; however the exact mechanisms governing the film growth in the dielectric barrier discharge electrode were not resolved. Precise control of the surface morphology and growth front evolution is important to produce the desired film quality in a cylindrical electrode configuration having non-uniform electric field. The aim of this work is to study the deposition mechanisms and growth front roughness evolution in an industrial relevant roll-to-roll configuration.

2. Experimental
The set of films was grown in a roll-to-roll AP-PECVD reactor with parallel bi-axial cylindrical electrode geometry on Polyethylene-2,6-naphthalate (PEN) foil. Tetraethoxysilane (TEOS) was used as a precursor for silica-like thin films and the process gasses were argon, nitrogen and oxygen. The voltage between the two electrodes was 2-3 kV. The frequency ranged from 180-200 kHz. The Dynamic Deposition Rate (DDR) defined as the product of web speed and film thickness was approximately 10 nm/min. Hence, the film thickness was varied by adjusting the web speed. The estimated energy spent per TEOS molecule is 6.5 keV/molecule which is required to deposit good performing moisture barrier layers [1].

The thickness of the films was measured using spectroscopic ellipsometry (M-2000D, J.A. Woollam Inc.) and X-ray reflectometry (Panalytical X’Pert MRD Pro). The surface morphology of silica-like films was measured using Atomic Force Microscopy (AFM) (Park Systems NX10) in non-contact mode, using a tip with a radius of about 8 nm. In order to investigate film morphology evolution, AFM images of 512x512 pixels were obtained with scanning areas of 2x2 µm$^2$. The surface morphology of each layer was measured on at least 5 different locations. AFM images were analysed by using GWYDDION software [3]. Fig. 1 is an example of the pristine PEN as observed by AFM. In this study, we observe a “rice-like” characteristic structure of pristine PEN with an rms roughness of 1.75±0.04 nm.

Fig. 1. AFM image of pristine PEN scanning area 0.5x0.5 µm$^2$ (Polyethylene-2,6-naphthalate).

3. Method of Data Analysis
Family and Vicsek introduced dynamic scaling to describe both the temporal and spatial scaling behaviour [4]. The growth mechanisms can be understood by
analysing the surface morphology within the framework of the dynamic scaling theory characterized by the roughness ($\alpha$), growth ($\beta$) and dynamic ($z$) exponents. The surface height profile, obtained from AFM, can be characterised by first order statistics (i.e. the root mean square (rms) roughness). This well-known entity provides a measure for the overall roughness of the profile. The rms roughness value describes only the vertical properties of a surface.

Therefore the height-height correlation function (HHCF) is introduced which is defined as $H(r) = \langle [h(x) - h(x + r)]^2 \rangle$, where $h(x)$ represents the surface height at position $x$ on the scanned area. The dynamic scaling approach leads to the following scaling relations [4]:

$$H(r, t) \propto r^{2\alpha}, \quad r \ll \xi(t)$$

$$w^2(t), \quad r \gg \xi(t)$$

where $w(t)$ is the interface width or rms roughness, $\xi(t)$ is the lateral correlation length defined as the largest distance in which the height is still correlated and, $\alpha$ is the roughness exponent. The value of the roughness exponent $\alpha$ is determined by applying a linear regression in the double logarithmic plot at the length scale less than the lateral correlation length beyond which the surface is uncorrelated. The interface width describes the correlation with growth time or thickness as $w(t) \sim t^\beta$.

4. Results

Typical examples of the evolution of the morphology of SiO$_2$ film as a function of thickness for AFM images of 2x2 $\mu$m$^2$ are shown in Fig.2.

First, the rms roughness for each sample was calculated by averaging the values obtained by five 2x2 $\mu$m$^2$ AFM images. Thus, Fig.3 shows the log-log plot of the rms roughness in relation to different thicknesses. We observed that the roughness decreased from $1.73 \pm 0.07$ nm at a thickness of 3.5 nm to $1.61 \pm 0.06$ nm for 16.5 nm thick film, indicating dynamic smoothening growth. It must be pointed out that the surface of the pristine PEN foil used in our experiments is characterized by a high local roughness with specific oriented structures resulting in an rms surface roughness of $1.75 \pm 0.04$ nm. For very thin films ranging in thickness from 3.5-11.4 nm, the local substrate roughness is important and SiO$_2$ films reproduce morphological features very close to the features of the pristine PEN.

The scaling approach is applied for a self-affine range from 16.5 nm to 212.3 nm thick layers. Linear regression was applied to the experimental data of rms surface roughness in the range of thicknesses from 16.5 nm to 212.3 nm to obtain a growth exponent: $\beta = 0.02 \pm 0.01$.

![Fig. 2. AFM images of scanning area 2x2 $\mu$m$^2$ showing the evolution of the surface morphology for (a) bare PEN and for SiO$_2$ films of different thickness: (b) 5.5 nm (c) 51.4 nm, (d) 212.3 nm.](image)

![Fig. 3. Experimental values of the rms roughness SiO$_2$ films as a function of the film thickness.](image)
Fig. 4. Height-height correlation functions determined for the range of thicknesses from 3.5 nm to 16.5 nm.

For films in the range of 16.5–212.3 nm there is a defined discrepancy in surface morphology compared to the pristine PEN surface. Almost no change in the roughness exponent was observed for the films in the 16.5 to 212.3 nm range remaining stable at a value of 0.84 ± 0.01.

Fig. 5. Height-height correlation functions determined for the range of thicknesses from 16.5 nm to 212.3 nm. The red line indicates the slope that determines the \( \alpha \)-value.

5. Conclusions and outlook

In this study, the dynamic morphological evolution of SiO\(_2\) films on PEN foil was investigated in the AP-PECVD roll-to-roll reactor using a specific energy spent per TEOS molecule of 6 keV/molecule. The analyses of AFM images of thin films allowed us to derive the rms roughness and Height-Height Correlation Function as a function of the film thickness. Pristine PEN has a complex directional surface morphology being non self-affine evolving into a self-affine surface at a thickness value of 16.5 nm. Above this film thickness the roughness exponent and the growth exponent were obtained: \( \alpha = 0.84 \pm 0.01, \ \beta = 0.02 \pm 0.01 \). Further analysis of the roughness development of thicker layers (>200 nm) is required.

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7. Reference