Influence of local gas phase mass transport coefficients on the drying rate of polymer films – a fundamental study

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Introduction

Throughout the last decades the demand for high quality coatings has strongly increased. Optical foils and printed electronics have to be produced with thickness deviations below 1 μ m. Recent studies about surface tension driven convection flows [1] show that laterally non-uniform drying of polymer coatings might lead to an undesirable surface roughness. Besides temperature inhomogeneities, varying gas phase mass transport coefficients are the main reason for significant differences in the local drying rate of polymer coatings. For a better understanding of the polymer drying process - especially in the case of laterally small structures (e.g. printed electronics) - the influence of the gas phase mass transport coefficient on the local drying rate was investigated in a fundamental study.

Experimental Setup

Representative for polymer coatings whose drying process is mainly defined by the gas phase mass transport resistance and not by the diffusion of the solvent in the film, the drying of waterborne Polyvinylalcohol (PVA) coatings was investigated by means of Inverse Micro Raman Spectroscopy (IMRS) [2]. Polymer films with a dry thickness of approximately 10 μ m were isothermally dried on a glass plate in a temperature-controlled flow channel at 40°C and air flow velocities between 0.2 m/s and 0.5 m/s. Figure 1 shows the flow channel with IMRS measuring technique. A laser beam ($\lambda = 533$ nm) is focused via an edge filter and a system of mirrors into the drying polymer film inside the flow channel.

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Figure 1: Inverse Micro Raman Spectroscopy (IMRS) for the experimental investigation of the local solvent concentration profiles during thin film drying processes.

In the focal point of the objective, the laser light is elastically and inelastically scattered. A part of the scattered light is collected again by the objective and is reflected towards the edge filter. Only the inelastically scattered Stokes-Raman light - which is frequency shifted towards the laser light - passes the filter and is detected on a CCD chip. By moving the focal point of the objective with a piezo-focus stepwise through the film, the concentration profiles during the drying process can be obtained with a spatial resolution of $1 - 2 \mu m$ and a time resolution of $\sim 1 s$.

Modeling

Depending on the lateral position of the measurements in a 60 x 60 mm film, the water-PVA solution showed different local drying rates and the dryings rates were "accelerated" during the drying process. To find an explanation for these observed phenomena, different approaches for the mass transport in the gas phase were used in a simulation program and the simulation results were compared to the experimental data. The local mass transport coefficient in the gas phase can be calculated from Sherwood correlations. For a drying surface with an offset between the beginning of the hydrodynamic and the concentration boundary layer Brauer [3] supposes the following approach:

$$Sh_{x+x_0} = \frac{\beta_{x+x_0} \cdot (x+x_0)}{D_g} = 0.332 \cdot \sqrt{Re_{x+x_0}} \cdot \sqrt[3]{Sc} \cdot \left[1 - \left(\frac{x_0}{x+x_0}\right)^{3/4}\right]^{-1/3}$$
(1)

with: x = distance btw. beginning of concentration boundary layer and point of measurement

 x_0 = distance btw. beginning of hydrodynamic and concentration boundary layer β_{x+x_0} = local mass transport coefficient Re_{x+x_0} = local Reynolds number $(Re_{x+x_0} = u \cdot (x+x_0)/v_g)$ Sc = Schmidt number $(Sc = v_g/D_g)$

In the case of the experimental setup from figure 1 the question arises, if the upstream edge of the temperature-controlled glass substrate (figure 3a, with initial offset) or the edge of the film (figure 3b, without initial offset) is the stagnation point of the hydrodynamic boundary layer.



Figure 3: Schematic illustration of different approaches to describe the mass transport in the gas phase.

In both cases, the upstream edge of the polymer film will be the starting point of the concentration boundary layer, at least at the beginning of the drying process (figure 3 top). As can be seen from equation 1, the mass transport coefficient has a numerical singularity for x = 0, so the polymer film will dry much faster at its upstream edge than anywhere else. According to equation 1, the mass transport coefficient - and therefore the drying rate - decreases in flow direction along the film. This is the explanation for the different local drying rates at different film positions. As soon as the drying rate at the upstream edge of the film falls below a certain value, the evaporation of the solvent at this position does not contribute to the formation of the concentration boundary layer any more and the starting point of the concentration boundary layer shifts downstream. At a specific position within the area of the film, this moving "drying front" causes the observed acceleration of the drying rate.

Results and Discussion

Figure 4 left shows a standard drying experiment at $T = 40^{\circ}C$ and an air flow velocity of u = 0.4 m/s. The position of the measurement is in the center of the 6 x 6 cm film. The experimental data show a strong increase of the drying rate during the drying process.



Figure 4: Drying of water-PVA solutions, $X_0 = 8 g_{water}/g_{PVA}$, $T = 40^{\circ}C$, left: comparison with different simulation approaches (u = 0.4 m/s), right: influence of air flow velocity on the drying process.

The simulation with a constant mass transport coefficient in the gas phase does not represent the observed acceleration at all. Simulation results taking into account the changing local mass transport coefficient due to a laterally moving drying front with initial offset between the beginning of the hydrodynamic and the concentration boundary layer (see figure 3a) show an acceleration of the drying rate, but there are still significant deviations from the experimental data. Without an initial offset (see figure 3b) the simulation results predict the experimental data very well. According to this, the upstream edge of the thin polymer film is the stagnation point for the hydrodynamic boundary layer in flow direction. Figure 4 right shows, that also the influence of the air flow velocity can be well predicted by the corresponding simulation results (again without initial offset).

For future works these findings are significant and crucial to predict the drying process of laterally structured polymer films accurately with regard to the local mass transport coefficients e.g. in the field of printed electronics.

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