IN-SITU MEASUREMENT OF SURFACE STRUCTURE FORMATION CAUSED BY MARANGONI FLOWS DURING DRYING OF THIN FILMS

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Introduction

For thin coated products, surface homogeneity is an important quality characteristic. The roughness of the surface coating, which develops during drying, is caused by interfacial dynamic instabilities due to gradients of surface tension, called Marangoni instabilities. The main forces – the surface tension and the viscosity – as functions of solvent content and temperature alter as drying proceeds and freeze the structures remaining in the dry film. The concentration and temperature gradients can be induced to the film e.g. by inhomogeneous flow conditions in the gas phase as well as different heat conductivity / capacity. This is a problem in printed electronic production in particular. An in-situ measurement technique visualizing the developing surface structures during the drying process is a powerful tool to give us a deeper understanding of the proceeding mechanism.

Experimental setup

The measurement technique to observe the appearances of surface structures during thin film drying is based on the analysis of the refracted images of a dot pattern through the surface of transparent fluid (*Moisy et al. 2007; Krenn et al. 2010*). A schema of the measurement technique is shown in figure 1. A CCD-camera perpendicular above the film is used to take sequences of images during the drying. The

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polymer film is casted on a thin glass substrate with a dot pattern on its bottom side. A simultaneous measurement of the height of the polymeric solution unaffected by gradients is necessary because of the shrinkage of the layer during the drying process. To induce lateral temperature gradients on the glass substrate a plate with different thermal conductivities ($\kappa_1 > \kappa_2$; for the shown experiments aluminum / Teflon) is placed between the heating plate and the glass substrate (see figure 1 left).

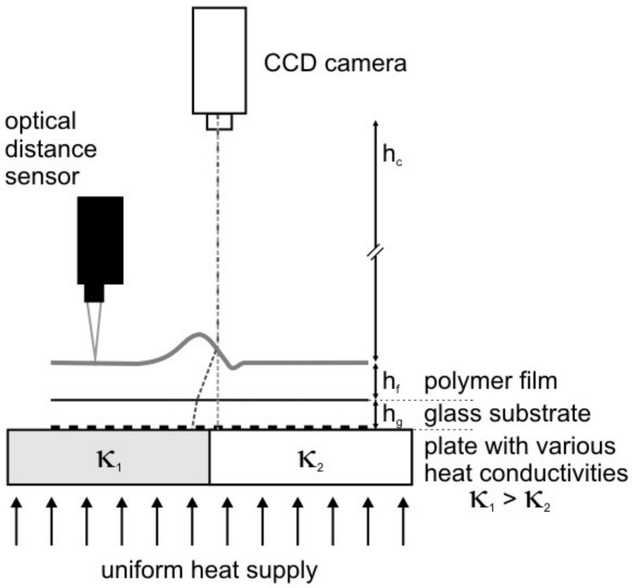
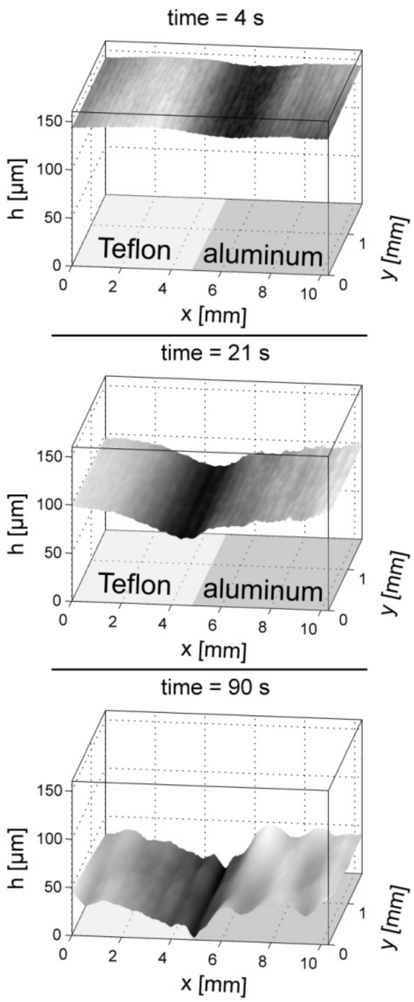


Figure 1:
Schematic principle of the measurement technique and the optical path through the deformed polymeric film and the glass substrate. Two independent measurements are needed to reconstruct the time-resolved surface topography: The measurement of the surface slope is obtained by measuring the displacement of the pattern of dots underneath the glass substrate and the unaffected height of the polymeric solution (Krenn et al. 2010).

If the optical properties of the system (i.e. refraction index of the polymer solution and the glass substrate) are known, the instantaneous deformation of the polymer film can be reconstructed from the displacement field $(\delta x, \delta y)$ between the refracted image of the deformed surface and the reference image obtained at planar surface. The reference picture at planar surface is taken before the polymer solution is casted on the glass substrate. For detailed information on the procedure and mathematical equations please see *Krenn et. al. (2010)*. Measurements of the topography are shown in the next section.

Results and Discussion

The shown experiments were performed with a polymer solution of poly(vinyl acetate) (PVAc) with methanol as solvent. For this polymer system the drying process is well known from former works (Krenn et al. 2008). The polymer solution is prepared with an initial solvent content of $X_0 = 2\ \frac{g(methanol)}{g(PVAc)}$ and then casted on the glass substrate. The experiments has been performed in a temperature controlled drying channel at a constant gas velocity of $0.5\,\frac{m}{s}$. A result is shown in figure 2.



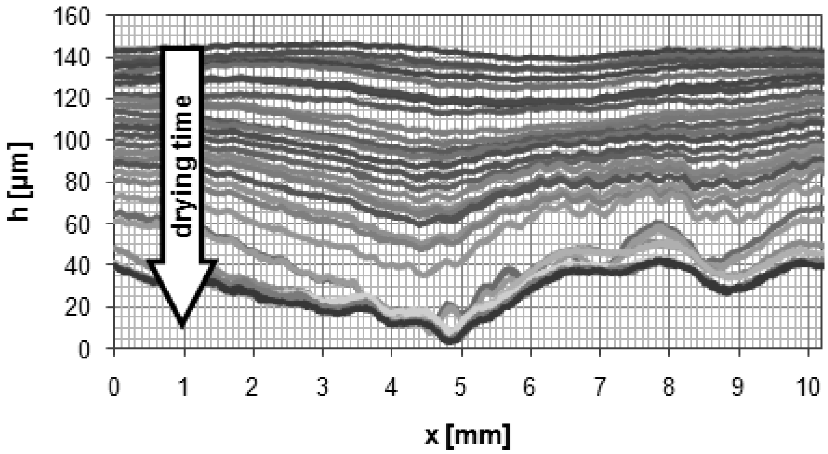


Figure 2:

Shrinkage and surface deformation during the drying process of a thin polymer film (methanol/PVAc) on a plate with different thermal conductivities (Teflon/aluminium). The polymer solution is casted on a thin glass substrate and observed with the presented measurement technique.

left: Perspective view of the surface at different time steps

right: cross section of the surface during drying along x-direction in the middle of the observed part of the film.

Due to the evaporation of the methanol the film shrinks and cools down above the Teflon block due the low thermal conductivity of the Teflon. A numerical simulation of the temperature field in the substrate at constant rate evaporation of the solvent shows the developing temperature gradient on the border between Teflon and aluminum side. Thus the drying rate slows down on the Teflon supported side and lateral solvent concentration gradient occurs. The surface tension is a strong function of the solvent concentration, thus a resulting driving force induces a convective flow in the polymer solution from the Teflon supported side to the aluminum side. Although the film dries slower on the Teflon supported side, this polymer flow leads to thinner polymer film on the Teflon side and a wave along the border between Teflon and aluminum. At a certain solvent content, the surface structures “freeze” due to the increasing viscosity of the polymer solution.

A comparison of the topography reconstructed (calculated) from the last picture and a profilometer measurement of the dry film showed that the reconstruction based on weak slope linearization of the 2-D non linear equation can follow the time-dependent deformation of the drying polymeric film. However, it lightly overestimates the depth of the occurring structures for the shown extreme case of deformed surfaces.

Conclusion and Prospect

The results in this contribution show, that the presented optical measurement technique is suited well to investigate the development of surface structures during the drying of thin films. At the present point of this work, the results are qualitatively in good agreement with mechanically measured surface profiles. For a quantitative evaluation, the measurement technique has to be improved and the linearization of the mathematical solution of the 2D problem has to be analyzed. Nevertheless, for this research topic the information obtained by the described measuring technique is fundamental due to the fact, that time-resolved processes of the occurring convection flows can be visualized and discussed.

Measuring the development of surface structures during thin film drying is a necessary step to validate future CFD simulation results of Marangoni convection flows.

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