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Visualization of surface deformations during thin film drying using a Digital-Image-Correlation method

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ABSTRACT

In this paper, an optical on-line measurement technique for the topography of thin polymer films during the drying process is presented. Based on the refraction of light at the interface between two transparent fluids (cf. free-surface synthetic schlieren method), this technique allows the indirect measurement of the surface slope and thus the time-resolved topography of liquid films from the displacement field of the refracted image of a random pattern below the film using a Digital-Image-Correlation algorithm. A comparison of the topography reconstructed (calculated) from the last picture and a profilometer measurement of the dry film showed that the numerical reconstruction based on linearization of the 2-D nonlinear equation can follow the time-dependent deformation of the drying polymeric film. Thus this measurement technique is a powerful and practical tool for the theoretical understanding of fluid motions and the developing surface topographies during the drying process of polymer solutions.

1. Introduction

Visualizing the development of surface structures during the drying process of thin liquid layers, for example of coatings, is a very powerful tool for understanding the mechanisms which lead to roughness of the surface.

For thin coated products, surface homogeneity is an important quality characteristic, e.g. for LCD films or polarizer films the requirements for deviations in surface roughness are less than 1%. Different heat conductivities of the substrate (e.g. repair with a different material) or non-uniform heat supply or air flow in the dryer can cause lateral temperature gradients in the drying polymer solution, leading to different drying rates and thus lateral solvent concentration gradients. These gradients are driving forces for dynamic instabilities, which result in various structures of the thin film surface. The dynamic instabilities in such thin liquid layers (less than 1 mm thickness) are caused by gradients of surface tension known as Marangoni instabilities. A visualization of the development of these surface structures is necessary in order to understand the mechanisms of the instabilities and the processes leading to unintentional or desired surface structures.

Investigating the development of surface structures on a thin liquid layer requires an optical, non-intrusive measurement technique. There are lots of promising methods, based on light reflection or refraction of collimated light and one-point measurements [1,2], which have the possibility of line measurements as fast scanning technique [3]. Instead of collimated light, scattered light can be used, which simplifies the optical setup because no collimating lens or mirror is needed. The resulting measurement depends on the surface slope and height. A first approach using scattered light emitted from a structured pattern through an interface is discussed by Kurata et al. [4]. Later, Moisy et al. [5] presented a technique based on the optical observation of an unstructured pattern through a thick liquid layer and showed, that it is possible to reconstruct the topography of the layer with very good precision.

This paper will prove this optical measurement technique for thin polymeric solution layers during the drying process. The optical measurement technique consists of two steps. The first step is the measurement of the surface slope and the reference height of the film. The surface slope is obtained from the displacement field of the refracted image of a random pattern using a Digital-Image-Correlation algorithm. The reference film height is measured simultaneously at one point of the liquid layer outside of the area influenced by gradients. The second step is the reconstruction (calculation) of the time-dependent local surface height using a least-squares integration of the surface gradient. For drying and thus shrinking layers the simultaneous measurement of the reference height of one point is necessary due to the fact that the displacement field depends on the local height and the slope of the polymeric surface. From the measured time-dependent shrink-

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Nomenclature

List of symbols

δx	optical displacement of the dot pattern in x-direction [μm]
$\boldsymbol{\delta r}$	optical displacement field of the dot pattern in x- and y-direction [μm]
h	local height [μm]
h_{ref}	reference film height outside the area influenced by any gradients [μm]
n	refractive index
m	mass [kg]
t	time [s]
X_s	solvent content in the polymeric solution $X_s = \frac{m_s}{m_p}$
x, y	fixed coordinate in the plane of the dot pattern [m]

Greek letters

φ_s	volume fraction of the solvent in the polymeric solution
ρ	density [kg/m^3]

Subscripts

a	air
f	polymeric film (varying content of solvent and polymer)
p	polymer
s	solvent
g	glass
end	end of the drying process

age of the film the average solvent content and therefore the average refractive index of the polymeric solution can be calculated. With this information it is possible to reconstruct the surface height and thus the time-resolved topography of the drying liquid film.

2. Experimental set-up and evaluation

2.1. Experimental set-up

The measurement technique to observe the appearances of surface structures during drying of thin films is based on the analysis of the refracted images of a randomized dot pattern observed through the surface of a transparent fluid as shown by Moisy et al. [5]. A schematic of the measurement technique is shown in Fig. 1. A CCD-camera with a telecentric objective placed above the film is aligned perpendicularly to the surface. In contrast to Moisy et al., the telecentric objective with a high accuracy is necessary for these thin film measurements since very small displacements have to be detected. The camera system takes sequences of images during the drying of the polymeric solution. The polymeric film is cast on a thin glass substrate, which has a dot pattern underneath. An additional measurement of the height of the polymeric solution unaffected by gradients is necessary because of the shrinkage of the layer during the drying process. Different thermal conductivities ($K_1 > K_2$) under the substrate induce lateral temperature gradients in the polymeric solution during the drying process (see Fig. 1), due to the evaporation of the solvent. The temperature gradients and the simultaneously occurring concentration gradients caused by the temperature-dependent drying rate result in surface tension driven flows within the polymeric solution. These flows form surface structures during the drying process, which can be observed by the presented measurement technique and are also present in the dry film.

In case of a deformed surface, the dots of the randomized pattern at the bottom of the glass substrate appear to be shifted (Fig. 1). Therefore, the time-resolved topography of the polymeric solution can be observed by the displacement of the dot pattern. The relation between the topography and the observed displacement field is described below.

2.2. Relation between the local surface height and the displacement field

To calculate the current topography we have to determine the apparent displacement of the dots $\boldsymbol{\delta r}(x, y)$ (for x-direction

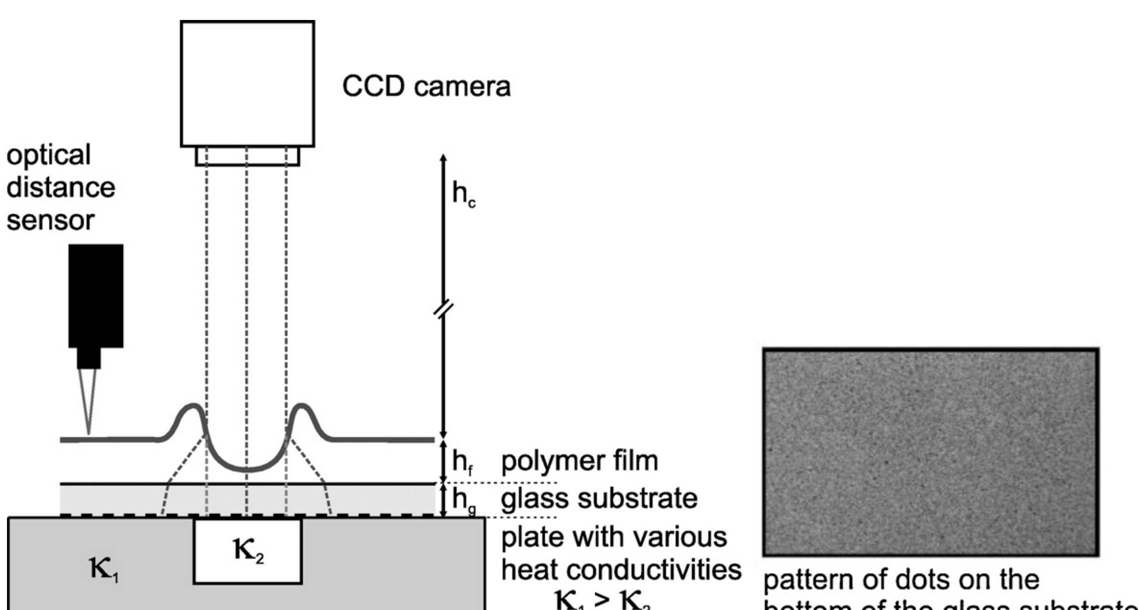

Fig. 1. Schematic principle of the measurement technique and the optical path through the deformed polymeric film and the glass substrate. Two independent measurements are needed to reconstruct the time-resolved surface topography: the measurement of the surface slope is obtained by measuring the displacement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