INVESTIGATION OF INTERFACIAL STABILITIES WITH A MULTILAYER SLIDE-COATING PROCESS (Speach)

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Abstract

Processing of organic electronics tend towards solution processing, which allows a fast, low cost mass production of devices in small and large area dimension. But current challenges according to wet film processing range from difficulties of thin film processing to re-dissolving of already printed and dried active materials and the high diffusion speeds of these small molecules in the solvents. Further, increasing the overall process speed by coating simultaneously multiple liquid layers might exemplary enables intermixing zones of the liquids depending on the solvent miscibility. There exist a diverse range of instabilities for miscible and immiscible liquid multilayer stack. We are investigating these instabilities experimentally based on fluid properties with regards to solvents for organic electronics but on general systems as well. For the investigation, a slide coating device with an extended incline plate is chosen. The advantage of a slide coater for the investigation is, that after leaving the slots, the liquids form a multilayer stack on the incline. The behavior of the liquids to each other and the stability of the multilayer film can be examined right from their first contact and during the flow down the plate.

Introduction

Newby et al. defined the difficulties for the wet film processing of organic electronics as "The Solvent Problem" (Newby et al., 2013 (1)). Thereby, the difficulties and challenges range from re-dissolving of already printed and dried active materials and the high diffusion speeds of the small molecules in the solvents (1). Still, the motivation exists, to apply simultaneously multiple layers to increase the overall process speed (2; 3; 4). If so, the lowest layer could function as a buffer layer and coated on already dried materials without dissolving it, as Tseng at al. proved for spin coating(5).

The simultaneous application of two liquid layers is investigated with an adjusted slide coating device to understand the mechanism which prevents the successful coating of multiple liquid layers. A slide coating device is chosen for the analysis of the film instabilities due to its unique possibilities in observing inhomogeneity and instabilities directly from the first fluid contacts and during the travelling down the incline.

Flow down an Inclined Plane

After leaving the slot exit of the slide coater, the fluid flows down on the slide until reaching the gap. The adjusted slide coater of this investigation has an extended plate to have a longer investigation time of the fluid behavior. Single and multilayer films are affected by several types of instabilities during this flow. Single and multilayer films are subjected to long-wave instabilities due to gravitational influence while multilayer configurations are affected additionally by types of instabilities due to the introduction of the interface (6; 7; 8). Thereby instabilities based on the fluid properties as liquid-liquid dewetting (9) or Rayleigh Taylor inversion (10) can occur. Further, due to the inclined configuration, jumps of the velocity profile due to viscosity or density stratification can cause perturbations (6).

Experimental Design and Set-Up

The core piece of the experimental set-up is the slide coater with three slots within an extended inclined plane. An overall test set-up including the slide coater and the gear pumps feeding each cavity individually for each slot is illustrated in Figure 1.

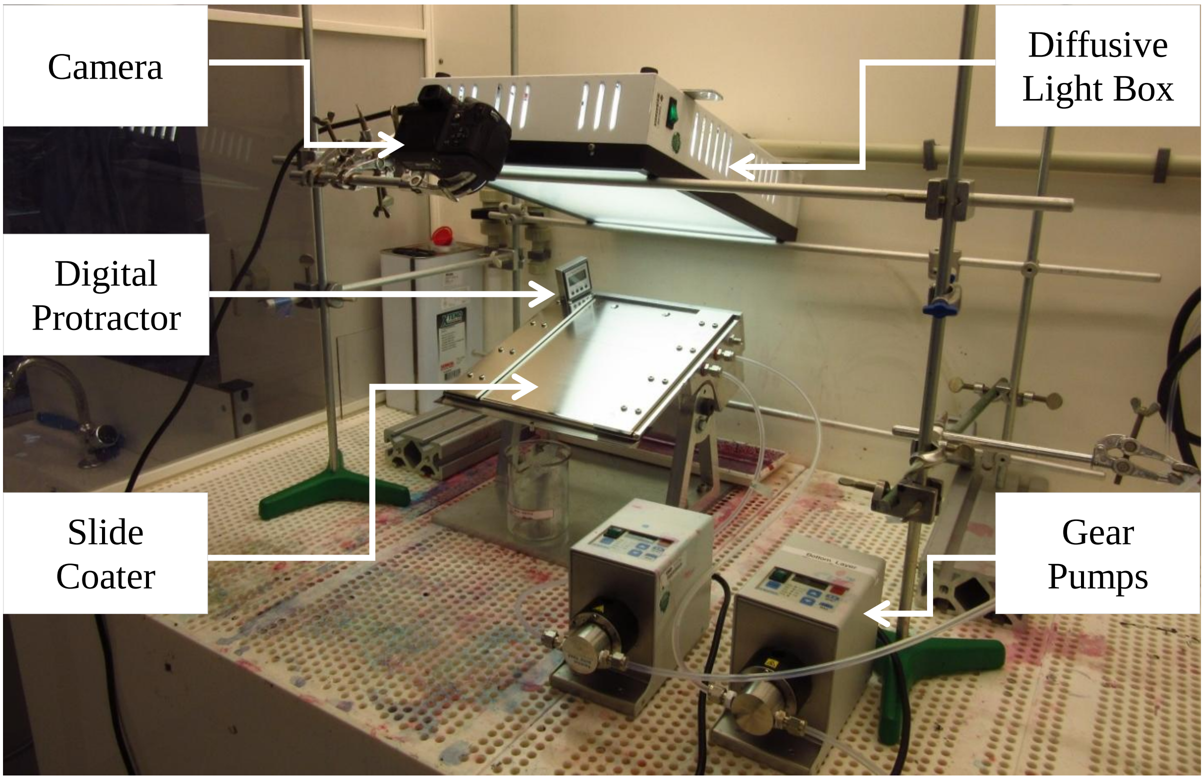


Figure 1: Experimental set-up with the slide coater as core piece, light box, camera, digital protractor, and gear pumps (own figure).

Solvent Selection

Several solvents are defined for the first screening. The combinations for two layer tests are selected by the following criteria. Major criterion is the difference in the top to bottom layer surface tension where the top layer should have the lower one. Further, several ranges in the surface tension difference are covered. A second criterion is, that immiscible, fully and partly miscible systems are covered. Third, regarding the viscosity, configurations with lower and higher viscous bottom layers are selected. The solvents are colored with dyes to allow a distinction of the flow.

Results and Conclusion

Major influence for a successful liquid stack is given by the capability of the top layer spreading on the bottom layer. Hereby gives the spreading factor as a balance of surface and interfacial tensions an indication, if the top layer will spread on the bottom one. Following, the interfacial tension measurement of Anisole in Propane-1,2-diol and the slide coating combination of these liquids is illustrated (Figure 2).

Figure 2: Anisole and Propane-1,2-diol. Left: Droplet is formed by the interfacial tension measurements. Right: Anisole does not spread on Propane-1,2-diol (own figure).

The spreading factor S for this combination is negative, indicating dewetting (see eq. 1).

Liquid-liquid-gas interface:

$$S = \sigma_{v,l1} - (\sigma_{l1,l2} + \sigma_{v,l2}) \quad \text{eq. 1}$$

with: $\sigma_{v,l1}$*: Surface tension bottom layer liquid [mN/m]*
$\sigma_{v,l2}$*: Surface tension top layer liquid [mN/m]*
$\sigma_{l1,l2}$*: Interfacial tension [mN/m]*

Relating the spreading factors for all performed slide coating test to the actual onset of spreading or dewetting shows a clear correlation, as illustrated in Figure 3.

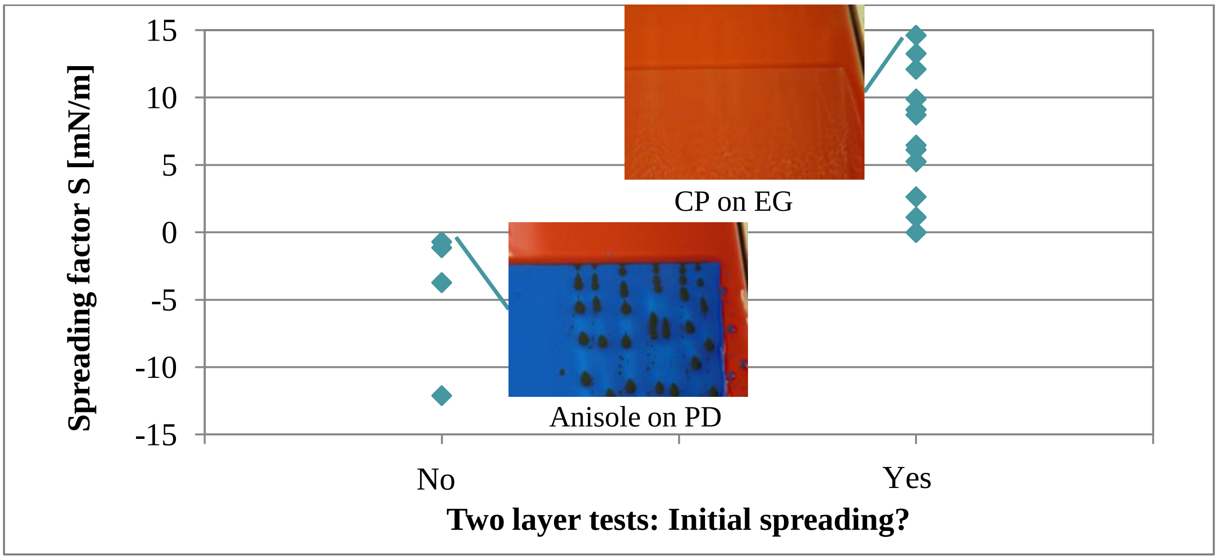


Figure 3: Calculated spreading factor S as function of the two classes spreading and non-spreading. Two examples are highlighted: Anisole and Propane-1,2-diol (PD) for non-spreading and Cyclopentanone (CP) and Ethylene Glycol (EG) for spreading (own figure).

Besides the crucial factor of spreading, further instabilities as shear waves (6; 11) for liquid combinations with a large viscosity difference are detected. Figure 4 shows the shear waves resulting in a break up in vortices for the combination of Cyclopentanone on Ethylene Glycol.

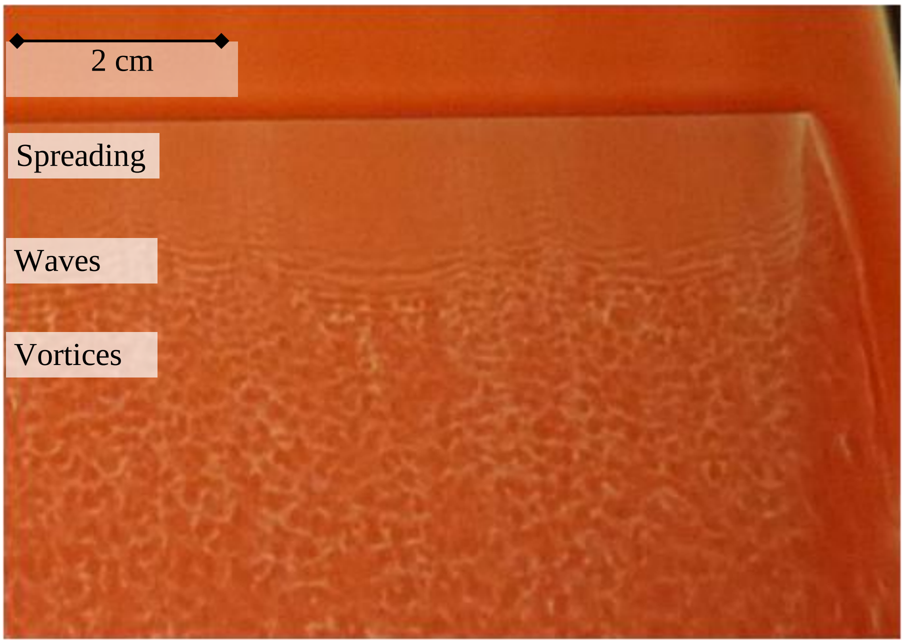
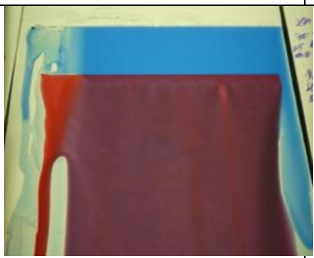
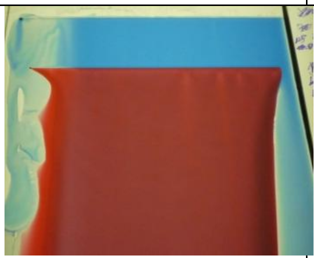
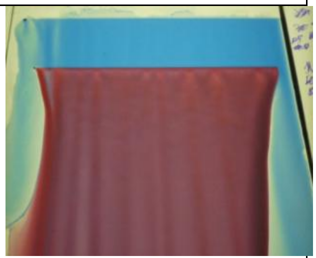
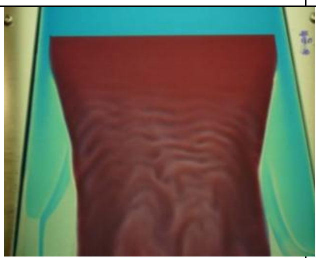
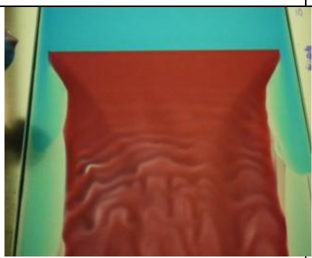
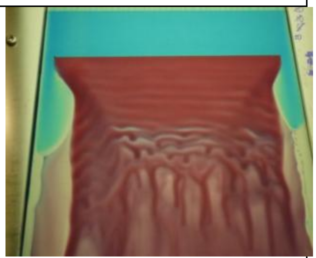


Figure 4: Shear wave instability by red Cyclopentanone on light blue Ethylene Glycol. Different characteristic areas from spreading to vortices are labeled (own figure).

Finally, miscible systems show different trends for stable and instable effects, which are mostly expressed as waves. Process characteristics as the wet layer thicknesses, inclination angle or even pump pulsation can enhance or damp these instabilities. The following table summarizes two slide coater test series for several volume flow ratios and absolute layer thicknesses (Table 1).

Table 1: Selected two layer slide coating test indication the effect of the volume flows or thickness ratios on the existence of waves.

Combination	Volume Flow Ratio 1:1	Volume Flow Ratio 1:2	Volume Flow Ratio 1:1
Cyclopentanone on Anisole			
Layer Thickness	141 on 135 µm	112 on 135 µm	112 on 107 µm
Isopropanol on Anisole			
Layer Thickness	183 on 134 µm	145 on 134 µm	145 on 106 µm

Summary

Within this investigation, the capability of the slide coating device to examine different interfacial and overall flow stabilities was proven. Several organic solvents combinations were tested from completely immiscible to fully miscible ones highlighting the major influence criteria on stable films, the spreading factor. Even the successful spreading of immiscible solvents can be explained with this theory. With the findings about spreading, a three dimensional plot is developed including further fluid properties to indicate expected stable and instable two layer coating tests.

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