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Polymer and DNA droplets: drying dynamics and deposit nanostructuring

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Polyethylene oxide (PEO) is a semicrystalline polymer with important biomedical applications due to its biocompatibility and water solubility. DNA is an important biomolecule and the drying of its suspension droplets is relevant to various applications such as biosensing DNA microarrays or inkjet bioprinting for microelectronic devices. We used drying droplets of PEO and DNA (several molecular weights) suspensions to study in some depth the pinning/de-pinning transition which is a determining factor for the final morphology of the deposit. In case of PEO, it is well-known that this process can lead to pillar formation under specific conditions. [1-3] We report on the drying process of sessile droplets of aqueous PEO solutions studied by contact angle analysis. [4] PEO droplets were left to evaporate. Residues were formed with either a disk-like puddle or a distinctive tall conical pillar shape. This occurred following a four-stage deposition process: pinned drying, pseudo-dewetting, bootstrap building, and late drying. Contact angle analysis allowed us to monitor all stages during drying and consider transitions between stages for different molecular weights. We reveal the effect of adhesion and contact line friction on the final morphology of the dried PEO solute. For high molecular weights the results are compatible with a pinning mechanism based on the interdigitation of the loops and tails of an adsorbed polymer layer with the polymer gel network inside the droplet that forms as water evaporates. DNA "biodrops" were deposited on substrates and allowed to dry. [5] Initially, the triple line (TL) of both types of droplet remained pinned but later receded. The TL recession mode continued at constant speed until almost the end of drop lifetime for the biodrops with short DNA strands, whereas those containing long DNA strands entered a regime of significantly lower TL recession. We discuss our observations based on free energy barriers to unpinning and increases in the viscosity of the base liquid due to the presence of DNA molecules. We show the importance of DNA length by comparing the nanostructuring of DNA deposits at the interior and contact line (Figure 1).

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[2] K. A. Baldwin, M. Granjard, D. I. Willmer, K. Sefiane, D. J. Fairhurst, Soft Matter 7, 7819 (2011)
[3] K. A. Baldwin, D. J. Fairhurst, Colloids and Surfaces A: Physicochemical and Engineering Aspects 441, 867 (2014)
[4] D. Mamalis, V. Koutsos , K. Sefiane, A. Kagkoura, M. Kalloudis, M. E. R. Shanahan Langmuir 31, 5908 (2015)
[5] A. Askounis, Y. Takata, K. Sefiane, V. Koutsos, M. E. R. Shanahan, Langmuir 32, 4361 (2016)

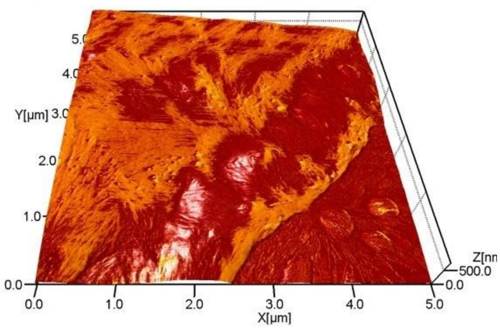

Figure 1 : 5 × 5 μm^2 AFM 3D topography image of the edge of the DNA deposit left behind after the drying of the water droplet containing long 1000 bp DNA strands. Z-scale ranges 0−770 nm. [5]