Supplementary Information

for

A Modular Approach to Studying Polymer Processing Using a Self-Driving Lab

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Chemicals and materials

Hydroxyl propyl cellulose (HPC) was purchased from Sigma Aldrich (molecular weight ~80,000 g/mol, powder). Anhydrous ethanol (EtOH) was purchased from Electron Microscopy Sciences. Food dye (FD&C Blue #1) was purchased from Spice Supreme. Silicon wafers (50.8 mm diameter, 280 µm thick, N-type doped with P, <100>, 1-10 ohm-cm) were purchased from University Wafers. White papers (extra bright, 75 gsm) were purchased from W.B. Mason. Airbrush (Eclipse HP-CS) was purchased from Iwata.

Stock solutions preparation

For the closed-loop optimization campaign, the solution of HPC was prepared by dissolving 5.5 g of the polymer in 7 mL of EtOH. For the spray pattern measurement, the solution of food dye was prepared by mixing 1 part of FD&C Blue #1 and 3 parts of water.

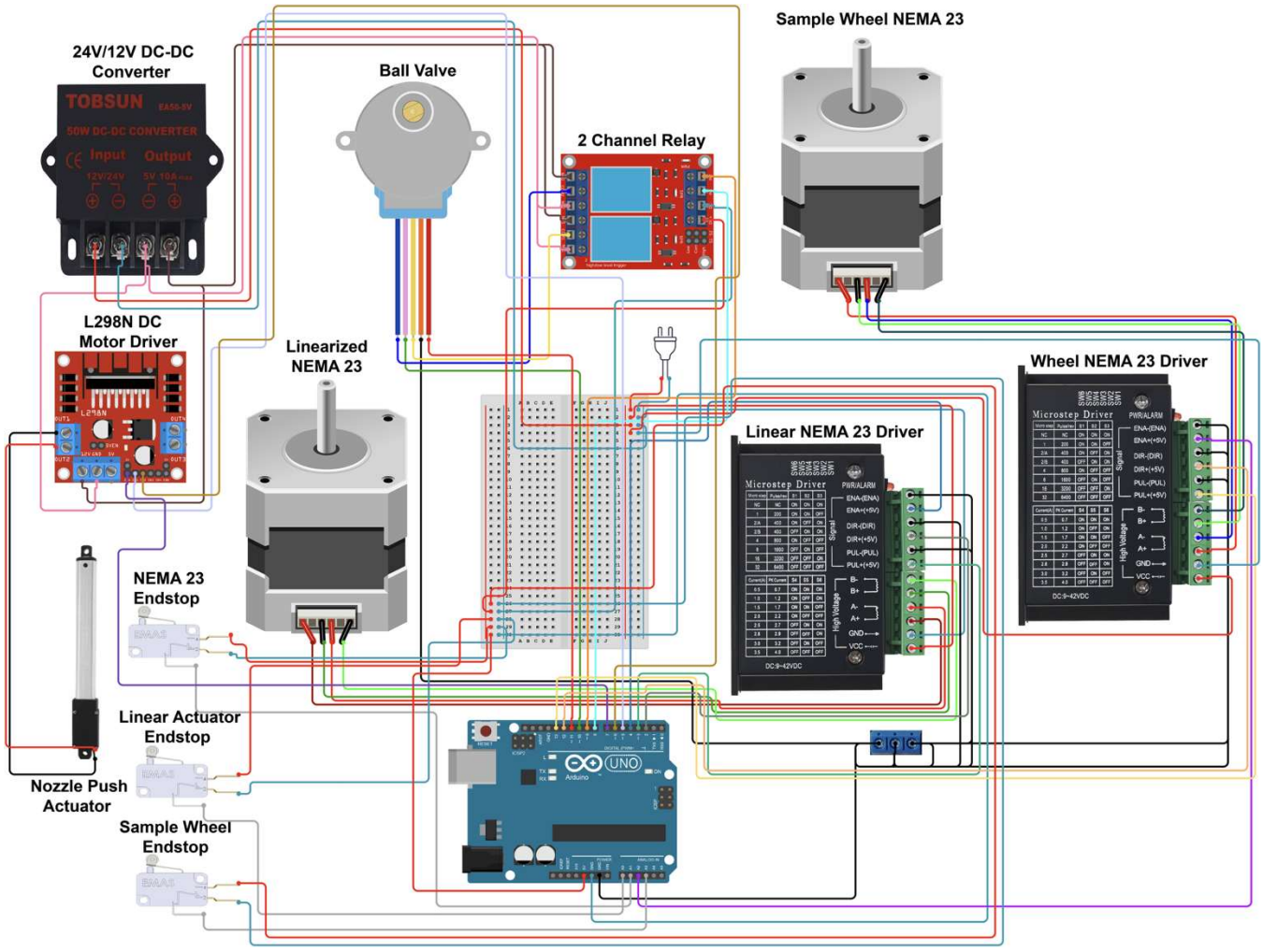

Fig. S1 Schematic of the spray coating station circuitry.

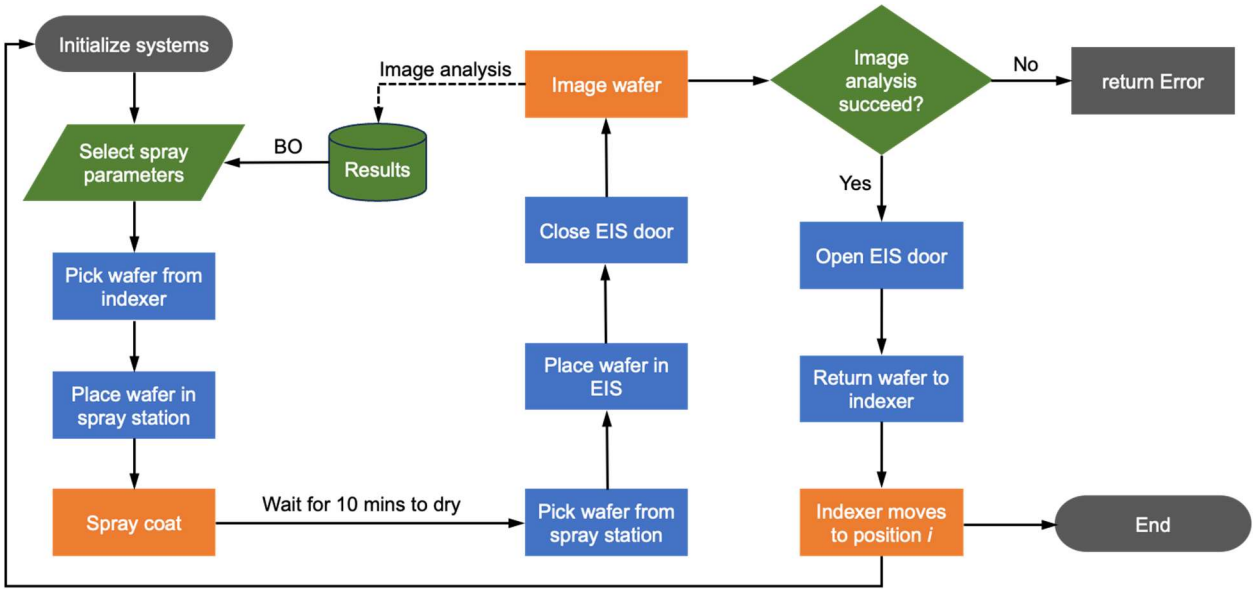

Fig. S2 Workflow diagram of the SDL, illustrating the sequence of operations during closed-loop experimentation. This workflow includes Bayesian optimization (BO) and the use of the custom elliptical imaging station (EIS). Blue elements indicate robotic handling and motion operations, orange elements represent experimental processing steps, green elements indicate decision-making or optimization operations, and gray elements indicate system initialization or termination states.

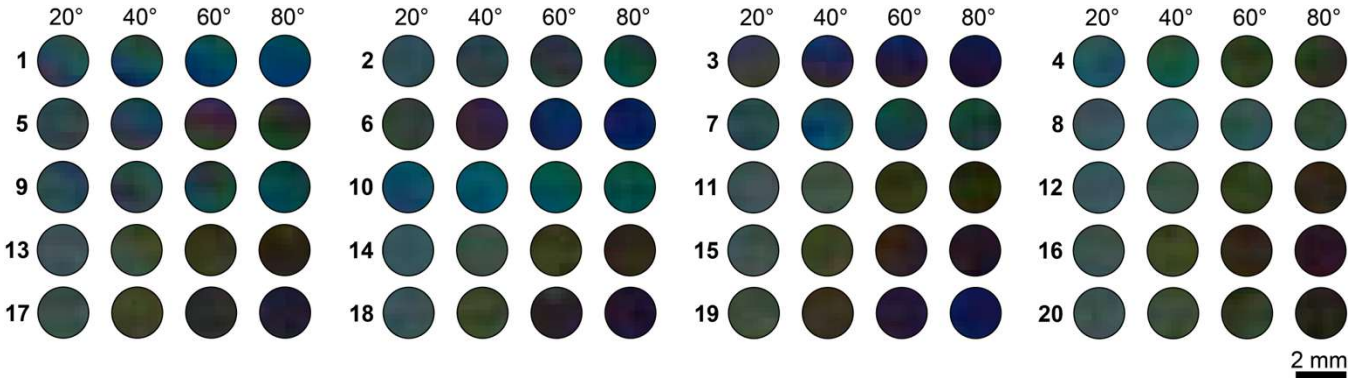

Fig. S3 Images of produced HPC films during the SDL campaign at different grazing angles.

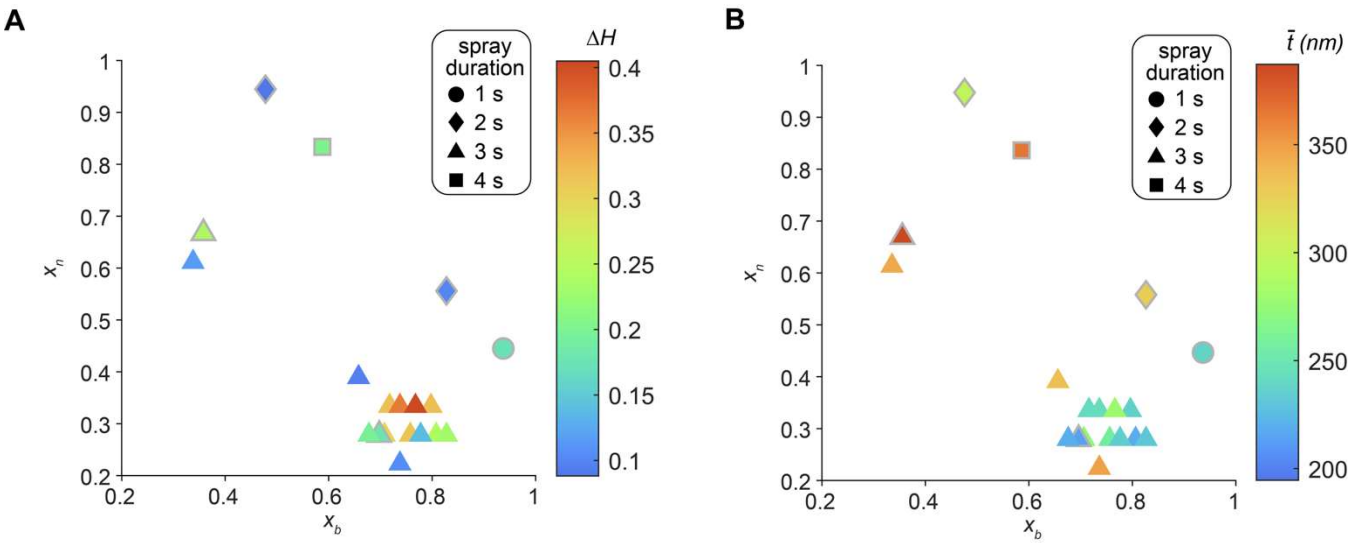

Fig. S4 (A) Iridescence strength as a function of spray coating parameters. (B) Thickness as a function of spray coating parameters. Data points outlined in gray correspond to initial experiments generated with Latin hypercube sampling (LHS).

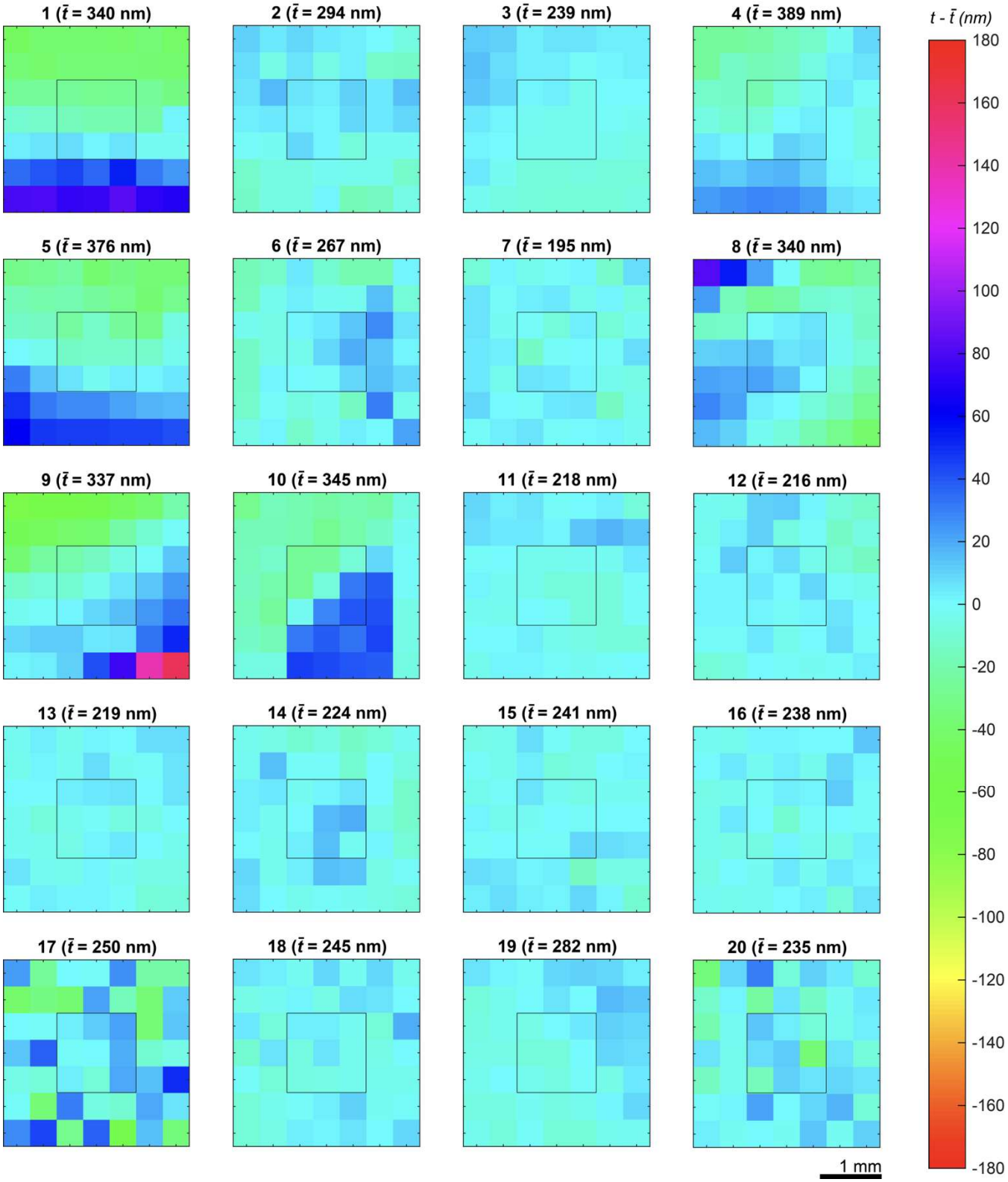

Fig. S5 Thickness deviation maps for samples 1-20. Each panel shows the local film thickness t, relative to the mean thickness of that sample $\bar{t}$.

The CAD files for the sample indexer, spray-coating station, and elliptical imaging station, along with the scripts used for thin-film interference modeling and SDL operation, are publicly available at: https://doi.org/10.5281/zenodo.18812450

Movie S1. Video showing one cycle of the automated experiment.