SUPPLEMENTARY MATERIAL

Experimental Verification of the Formation Mechanism for Pillar Arrays in Nanofilms Subject to Large Thermal Gradients

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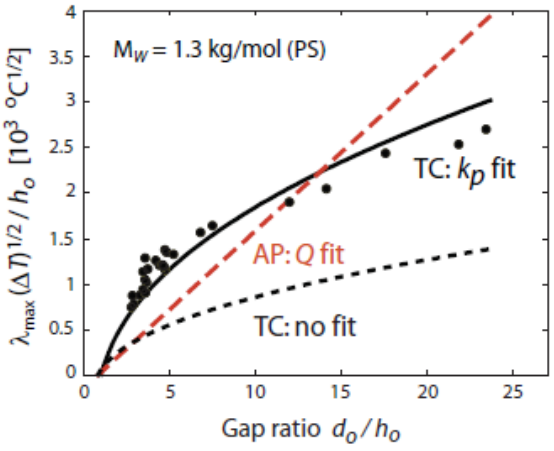
(1) The video sequence depicts structure formation in an initially flat nanofilm of molten polystyrene (2.46 kg/mol) with initial thickness h_o= 160 nm inserted within the experimental assembly shown in Fig. 1 of the paper with T_H = 130 °C and T_C = 60 °C. Structure formation is being viewed through a transparent cylindrical mesa (diameter 400 μm) fabricated from SU-8 where d_o^m = 630 nm. The time counter denotes hrs : min. Fig. 1(a) in the main text contains additional information regarding the experimental setup. The relevant temperature values for comparison to theoretical predictions were obtained from finite element simulations described in the text, which for this run correspond to to T_2 = 124 °C and $\Delta T = T_2 - T_1^m$ = 17.1 °C.

Structure formation at early times is characterized by a constant wavelength. Once the fluid protrusions make contact with the top the silanized mesa, however, there occurs rapid fluid reorganization with a subsequent decrease in wavelength. Only measurements prior to contact with the mesa (or prior to any coalescence or film depletion in the region outside the mesa) were used for comparison to theoretical predictions. The still images shown in Figs. 2(a) and (b) of the paper correspond to this experimental run.

(2) Measurements of the fastest growing wavelength for lower molecular weight nanofilm samples (1.3 kg/mol PS) are shown in the figure below. The data correspond to initial film thicknesses ranging from 90 nm ≤ h_o ≤ 260 nm and gap widths ranging from 600 nm ≤ d_o ≤ 2400 nm. The data for d_o/h_o < 10 were obtained from the region beneath the SU-8 cylindrical mesa (1 mm diameter). The data for d_o/h_o >10 were obtained from the region outside the mesa beneath the sapphire window. The temperature differences, $\Delta T = T_2 - T_1^m$ and $\Delta T = T_2 - T_1^S$, were computed from finite element simulations as described in the text.



Superimposed on the experimental data are the predictions of the AP and TC models. The (black) dashed line presents the TC model using the bulk material constants in Table I of the text, which yields an unacceptable fit. The (orange) dashed curve represents the AP model with a fitting constant Q=2.6 - the predicted linear increase with gap ratio d_o/h_o is not supported by the data. The solid (black) curve represents the TC model with fitting constant k_p = 0.62 W/m-K. This enhancement in the polymer thermal conductivity k_p over its bulk value 0.13 W/m-K produces excellent agreement with Eq. (1c). Within the measurement errors reported, however, the difference in the values of the fitting constants k_p for the 2.46 kg/mol and 1.3 kg/mol PS films is not statistically significant. A more comprehensive study of this fitting constant obtained from a much larger range in molecular weight is required in order to establish the correlation between the molecular weight and film thermal conductivity.