Multi-Scale Modeling of Early-Stage Morphology in Solution-Processed Polycrystalline Thin Films

David L. Patrick*[,1], Cyrus Schaaf,[1] Robell Morehouse,[1] and Brad L. Johnson[2]

[1]*Department of Chemistry, Western Washington University, 516 High St., Bellingham, WA, 98225*

[2]*Department of Physics, Western Washington University, 516 High St., Bellingham, WA, 98225*

SUPPORTING INFORMATION

Computational details. The monomer concentration at each lattice site at each new time $L(i,j,t+\delta_t)$ is found from the concentration at the previous time by numerically solving the 2D diffusion equation,

$$L(i,j,t+\delta_t) = L(i,j,t) + K(L(i-1,j,t) + L(i+1,j,t) + L(i,j-1,t) + L(i,j+1,t) - 4L(i,j,t)) + F\delta_t$$

applying periodic boundaries. Here $K = D\tau/(\Delta_{xy})^2$, $D =$ 4.22×10^{-10} m^2 s^{-1} is the diffusion coefficient, $\delta_t =$ 2.37 × 10^{-5} s is the size of the timestep, $\Delta_{xy} =$ 1 μm is the edge length of a lattice site, and the flux rate $F =$ 1,797 monomers μm^{-2} s^{-1}. For numerical stability the timestep is chosen to make $K \ll$ 1 (here $K =$ 0.01). For lattice sites occupied by a crystal, the concentration is held fixed at the bulk saturation value, $n_0 =$ 3.36×10^{23} m^{-3}.

Video 1. Epifluorescence videomicroscopy sequence acquired *in situ* during film deposition showing a portion of the induction regime, the entire nucleation regime, and the early part of the growth regime. Each bright object is a single TET crystal. Field of view 2.44 × 2.44 mm. The frame rate is 0.2 s^{-1}. Data in Figures 1 and 2 are based on measurements taken from a 1.8 × 1.2 mm subregion centered near the middle of the field of view of Video 1.

Video 2. Computed evolution of the monomer concentration landscape during the nucleation regime. Field of view 2 × 2 mm. Frames are in 2 s intervals beginning at $t =$ 91 s. Brightness is proportional to concentration.

Video 3. Same data as Video 2, rendered in 3D perspective.