## Electronic Supplementary Information Stable polydisperse free-standing porous films made by mechanical deformation

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The time-dependent lateral dimensions  $L_x(t)$ ,  $L_y(t)$ , film thickness h, and three diagonal components of pressure tensor,  $P_{\alpha\beta}(t)$  with  $\alpha = x$ , y, and z for the free-standing polydisperse film subject to biaxial expansion are shown in Figs. S1 and S2. The effective strain rate is estimated according to the definition  $L_{x,y}(t)/L_{x,y}(0) = \exp(\dot{\varepsilon}t)$ , see Fig. S2b.



Figure S1. Time series of two lateral dimensions  $L_x(t)$ ,  $L_y(t)$ , and thickness h(t) of film for the film at  $T = 1.0\epsilon/k_B$  upon biaxial expansion with  $\dot{\epsilon}\tau_e \approx 2.61$  (see Fig. S2b). Values of dimensions for the expansion ratio of  $\lambda = L_{x,y}/L_w \approx 4.0$ , i.e.,  $L_x(t) = L_y(t) \approx 526\sigma$  are indicated by arrows. Here  $L_{x,y}(0) = L_w \approx 134\sigma$ .



Figure S2. (a) Time series of three diagonal terms of pressure tensor  $P_{\alpha\beta}$  for the film at  $T = 1.0\epsilon/k_B$  upon biaxial expansion. (b) Change in lateral dimensions  $(L_x(t) \times L_y(t))/L_w^2$  plotted versus the relaxation time t on a semi-log scale. The effective strain rate  $\dot{\epsilon}$  is determined by the slope in (b). The expansion ratio of  $\lambda = L_{x,y}/L_w \approx 4.0$ , i.e.,  $L_x(t) = L_y(t) \approx 526\sigma$  is indicated by an arrow.

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Figure S3. Six selected chains of  $N_2 = 100$  out of the free-standing polydisper film at  $\lambda = 1.0$  and 4.0 are shown as indicated.

The effective film thickness h can either be estimated from the monomer density profiles  $\rho(z)$ , Fig. S4, or the scattering function in the direction perpendicular to the interfaces of films, Fig. S5.



Figure S4. Monomer density profiles rescaled to the bulk melt density for all monomers,  $\rho(z)/\rho_0$  (a), monomers in chains of  $N_1 = 1900$ ,  $\rho_1(z)/\rho_0$  (b), and  $N_2 = 100$ ,  $\rho_2(z)/\rho_0$  (c), respectively.



Figure S5. Two components of collective structure factor,  $S_{||}(q_{||})$  (a) and  $S_{\perp}(q_{\perp})$  (b), in the directions parallel and perpendicular to the expanding direction, plotted versus  $q_{||}$  (a), and  $q_{\perp}/(2\pi/h(\lambda))$  (b).  $S_{||}(q_{||}) \sim q_{||}^{-2}$  in (a), and the Porod law  $S_{\perp}(q_{\perp}) \sim q_{\perp}^{-4}$  in (b) are also shown by straight lines for comparison. Data for an unperturbed monodisperse film at  $\lambda = 1.0$  are included in (a), for comparison.

Time-dependent morphological changes of expanded thin polydisperse films at  $\lambda \approx 4.0$  and  $T = 1.0\epsilon/k_B$  indicating relaxation retardation are shown in Fig. S6. Detailed internal structures of films at  $t = 0\tau$  and  $1.2 \times 10^6 \tau$  are investigated in Fig. S7. Fig. S8 shows the monomer density profile  $\rho(z)$  at several selected temperatures T for the expanded film at  $\lambda \approx 4.0$  subject to cooling.



Figure S6. Snapshot configurations of thin polydisperse porous films at  $\lambda \approx 4.0$  subject to relaxation at several selected relaxation times t and assumed thicknesses h, as indicated where all 1000 chains of  $N_2 = 100$  are marked in orange color (a), and the very same six selected chains of  $N_1 = 1900$  are marked in different colors (b).



Figure S7. Snapshot configurations of slices of thickness  $3.0\sigma$  cut along the lateral dimensions from the expanded film shown in Fig. S6 at  $t = 0\tau$  (a) and  $t = 1.2 \times 10^6 \tau$  (b). Subchains belonging to long chains are shown in the background while belonging to short chains are marked in orange color with thicker bonds. The center of films is fixed at  $z = 0\sigma$ 



Figure S8. Rescaled monomer density profiles for all monomers,  $\rho(z)/\rho_0$  (a), monomers in chains of length  $N_1 = 1900$ ,  $\rho_1(z)/\rho_0$  (b), and  $N_2 = 100$ ,  $\rho_2(z)/\rho_0$  (c), plotted as a function of z at several selected temperatures T, as indicated. The centers of thin porous films in the z-direction are matched at  $z = 0\sigma$ . The interfaces located at  $Z_G^{(\text{lower})}$  and  $Z_G^{(\text{upper})}$  determined from  $\rho(z)$  in (a) for films  $k_B T/\epsilon = 1.0$  and 0.5 are indicated by dashed and sold arrows, respectively.