Ultrahigh resolution scanning electron microscopy (UHR-SEM) images taken using a JEOL JSM-7400F are shown in Fig. S1 and Fig. S2 for interrupted growth runs deposited at 27°C and 30°C, respectively. For each temperature, images were taken at low magnification (right column) and high magnification (left column). Film growth that was interrupted well before the transition point indeed showed a distinct nanocrystalline structure (Fig. S1a,b and Fig. S2a,b), while films left to grow up to the transition region showed a mixed nature, where pyramidal edges of large columnar crystals co-exist together with nanocrystals (Fig. S1c,d and Fig. S2c,d). Finally, film growth that was interrupted well after the transition region exclusively showed large, faceted crystals, as shown in Figs. S1e,f and S2e,f. As expected, the transition point shifted to shorter times and grew sharper at higher temperature due to the strong dependence of reaction rate on deposition temperature.
Figure S1. Surface morphology of films deposited at 27°C for 4hr (a) x50000 (b) x20000. Films deposited for 5hr (c) x50000 (d) x20000. Films deposited for 7hr (e) x50000 (f) x20000.
Figure S2. Surface morphology of films deposited at 30°C for 2hr (a) x100000 (b) x25000. Films deposited for 3hr (c) x80000 (d) x33000. Films deposited for 6hr (e) x80000 (f) x25000.