# Supplementary Information

# Tailoring the crystal growth of quartz on silicon for patterning piezoelectric films

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**Table S1**. Correspondence between the molarity of the $SrCl_2$ aqueous solution and the Sr to $SiO_2$ molar ratio in the final solution used in the dip-coating process.

| **Concentration of the $SrCl_2$ aqueous solution (Solution B)** | **Sr/Si molar ratio in the final solution (Solution C)** |
|---|---|
| 0.4 M | 0.02 |
| 0.5 M | 0.025 |
| 0.6 M | 0.03 |
| 0.65 M | 0.0325 (shown as 0.033 in paper) |
| 0.7 M | 0.035 |
| 0.75 M | 0.0375 (shown as 0.038 in paper) |
| 1 M | 0.05 |
| 1.5 M | 0.075 |
| 2 M | 0.1 |

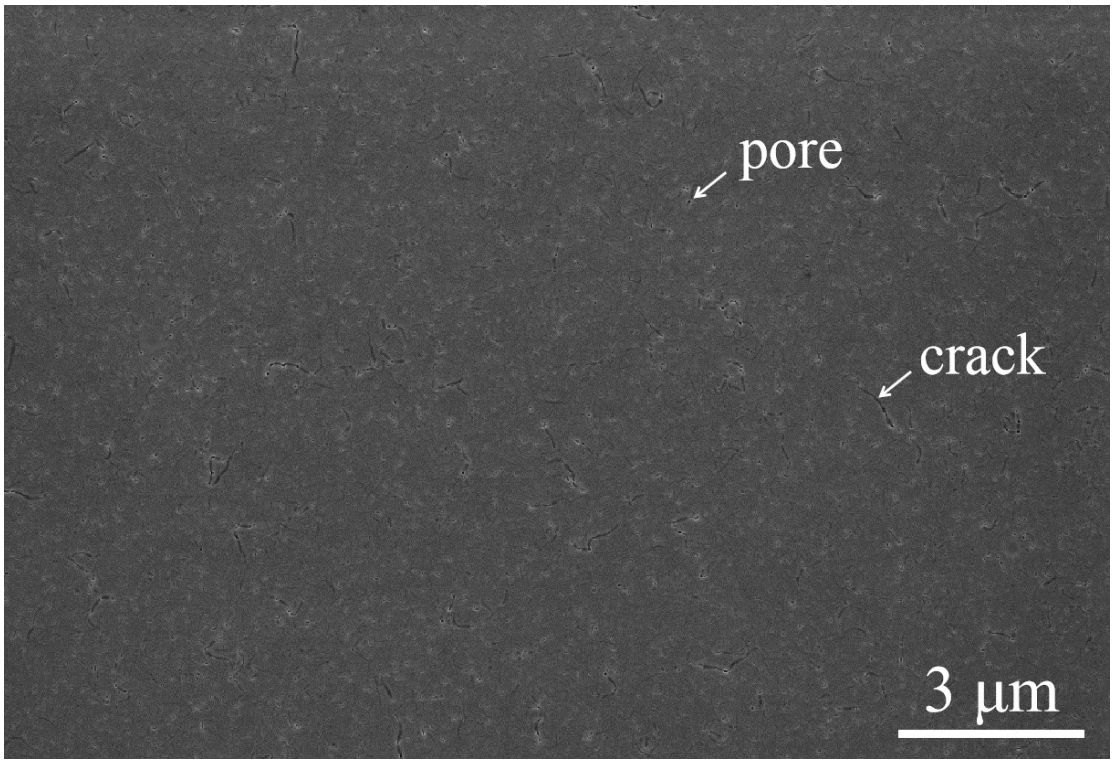


**Figure S1.** SEM image in secondary electrons mode of a film without Sr, showing the presence of cracks and pores.

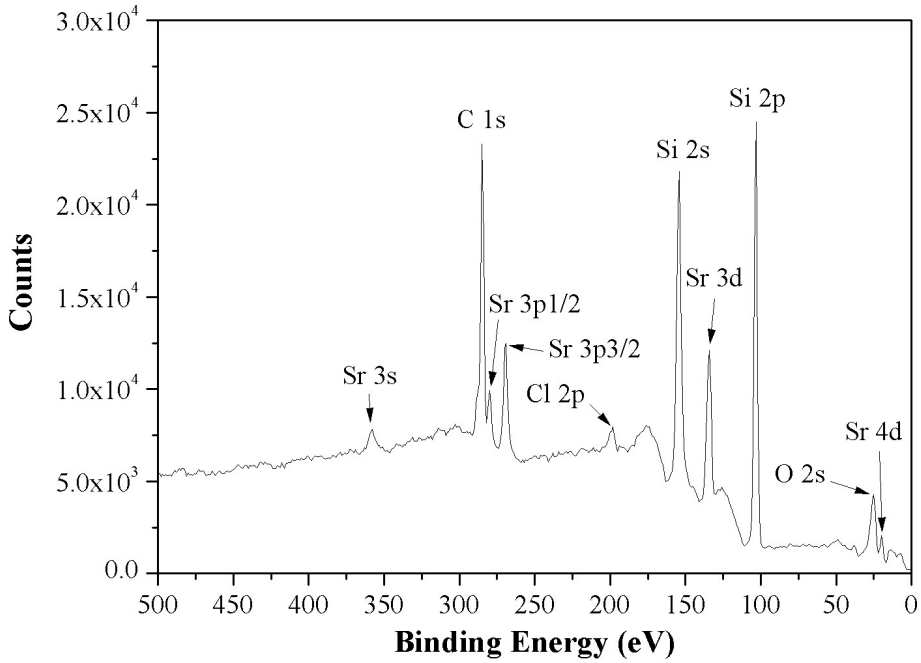


**Figure S2.** XPS overview of $\boldsymbol{R}_{Sr}$ = 0.05 film to show the present of Sr components

inside film.

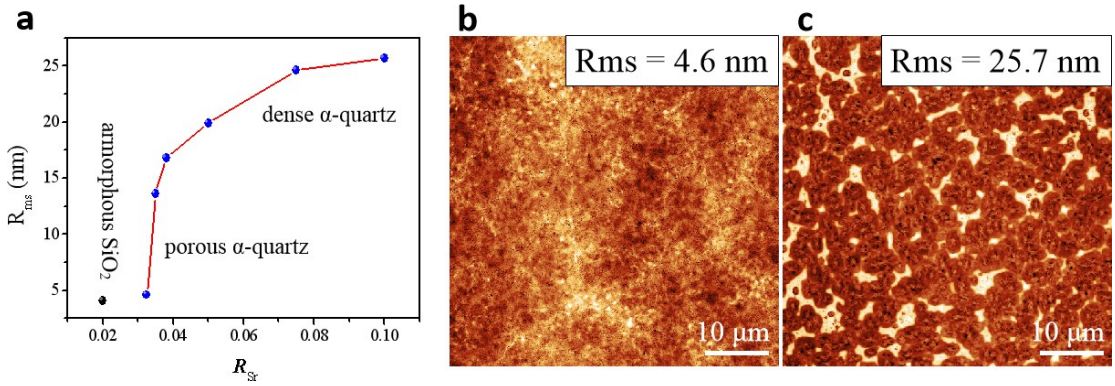


**Figure S3.** Topography study for different $\boldsymbol{R}_{Sr}$: **(a)** The relationship between with $\boldsymbol{R}_{Sr}$ and film roughness ($R_{ms}$) **(b)** Topographic AFM image of the porous film ($\boldsymbol{R}_{Sr}$=0.033) with the lowest $R_{ms}$, **(c)** Topographic AFM image of a dense film (with $\boldsymbol{R}_{Sr}$=0.1) displaying the largest $R_{ms}$.

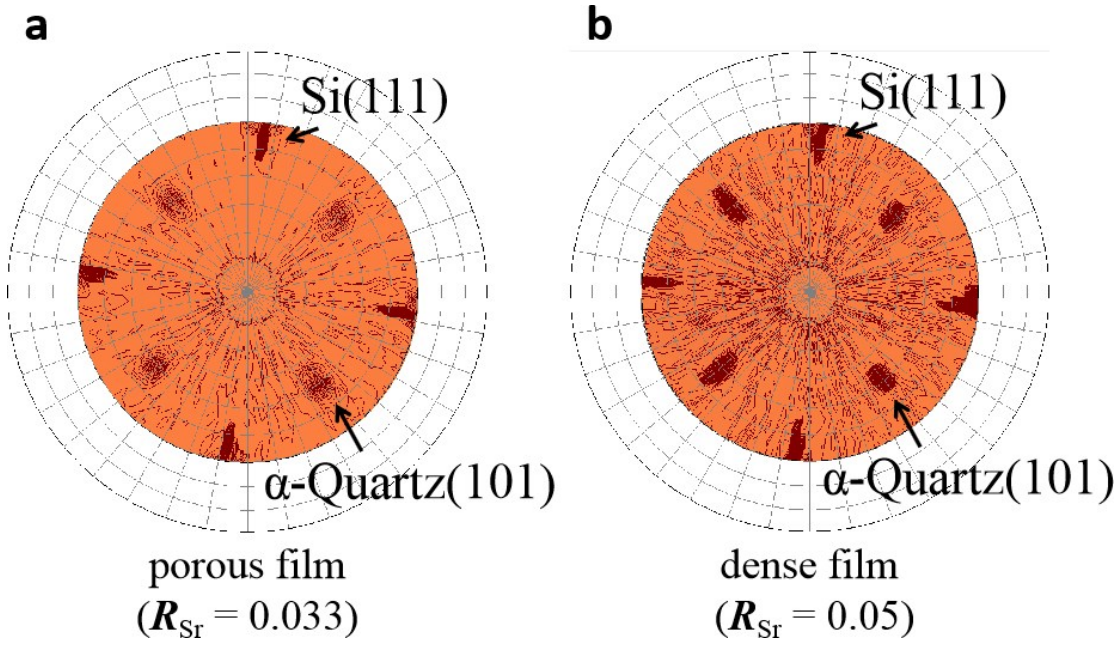


**Figure S4.** Pole figure of (101) α-quartz for two characteristic microstructures: **(a)** porous film ($\boldsymbol{R}_{Sr}$ = 0.033), **(b)** dense film ($\boldsymbol{R}_{Sr}$ = 0.05).

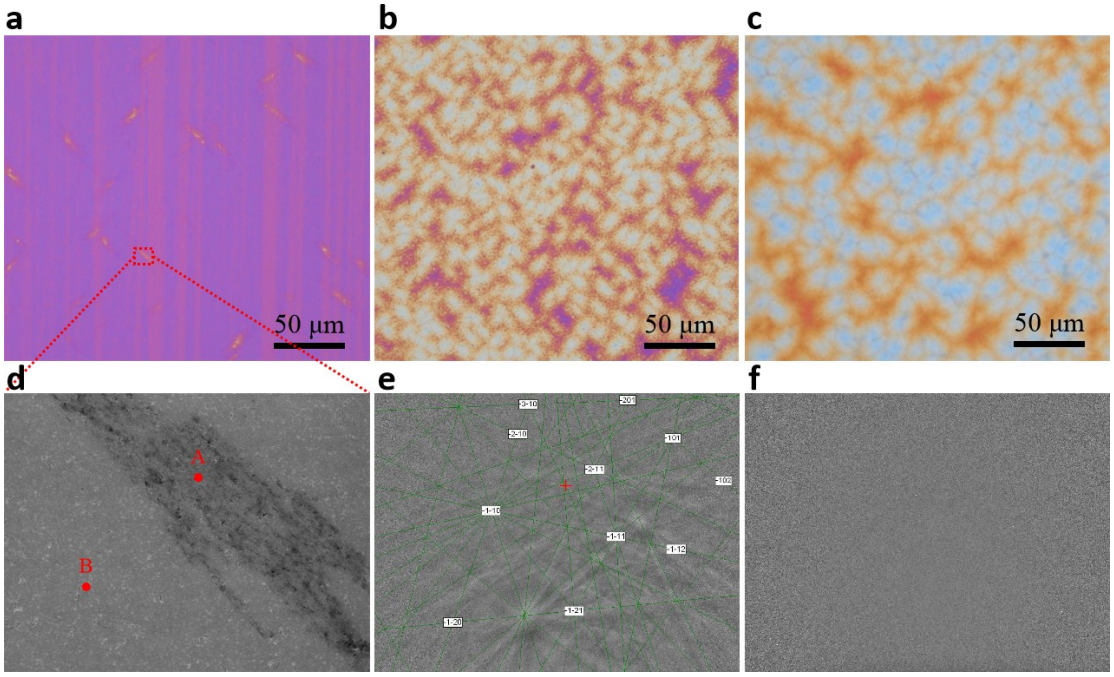


**Figure S5.** Optical images of partially crystallized films when $\boldsymbol{R}_{Sr}$ is **(a)** 0.02, **(b)** 0.025, **(c)** 0.03. In these images, only the light color areas are crystallized to α-quartz phase, the remaining violet and brown areas are still amorphous. **(d)** is the SEM image of one selected zone of $\boldsymbol{R}_{Sr}$ = 0.02 condition film which containing crystalline structure (dark area) and amorphous surface (light area) for EBSD test. **(e)** is the EBSD result of point A in **(d)** with α-quartz phase signals. **(f)** is the EBSD result of point B in **(d)** without any signal.

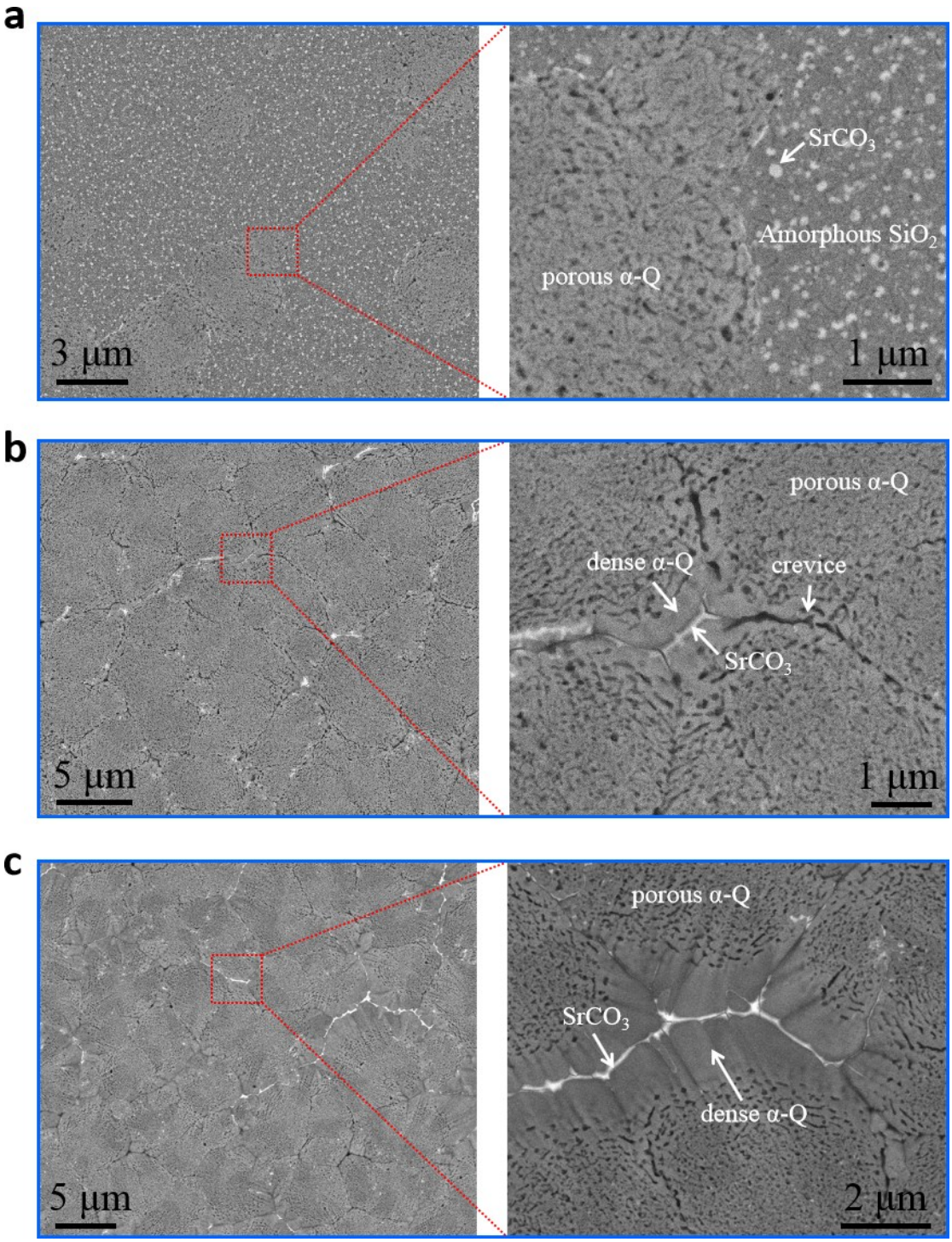


**Figure S6.** SEM images in secondary electrons mode of $R_{Sr}$ = 0.035 films crystallized with different times at 1000ºC: **(a)** 40 min. **(b)** 50 min. **(c)** 60 min.

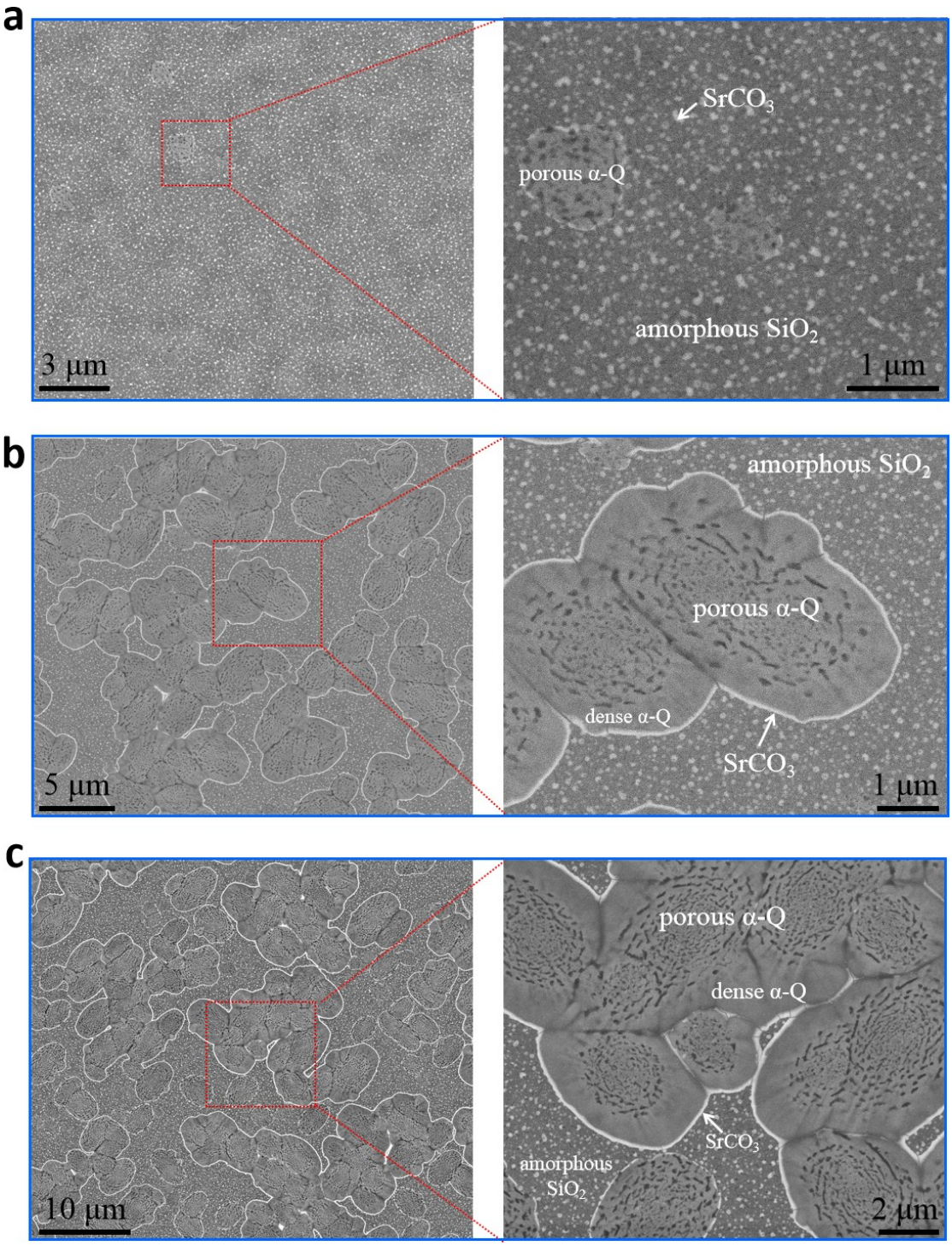


**Figure S7.** SEM images in secondary electrons mode of $R_{Sr}$ = 0.05 films crystallized with different annealing times at 1000ºC: **(a)** 20 min. **(b)** 30 min. **(c)** 60 min.

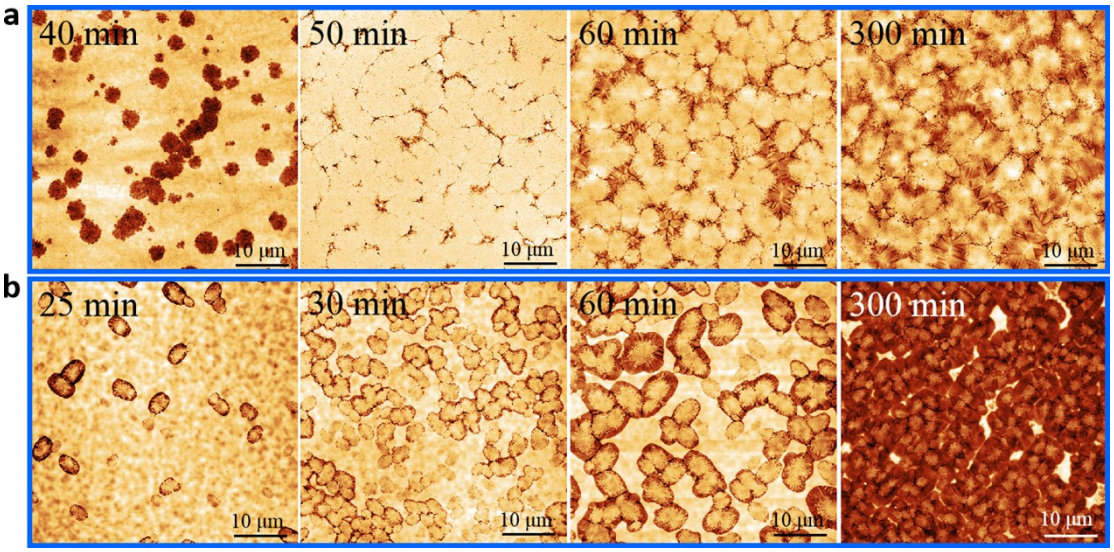


**Figure S8.** Topographic AFM images showing the influence of annealing time at 1000ºC for **(a)** porous microstructure (obtained with $\boldsymbol{R}_{Sr}$ = 0.035), **(b)** dense microstructrure (obtained with $\boldsymbol{R}_{Sr}$ = 0.05).

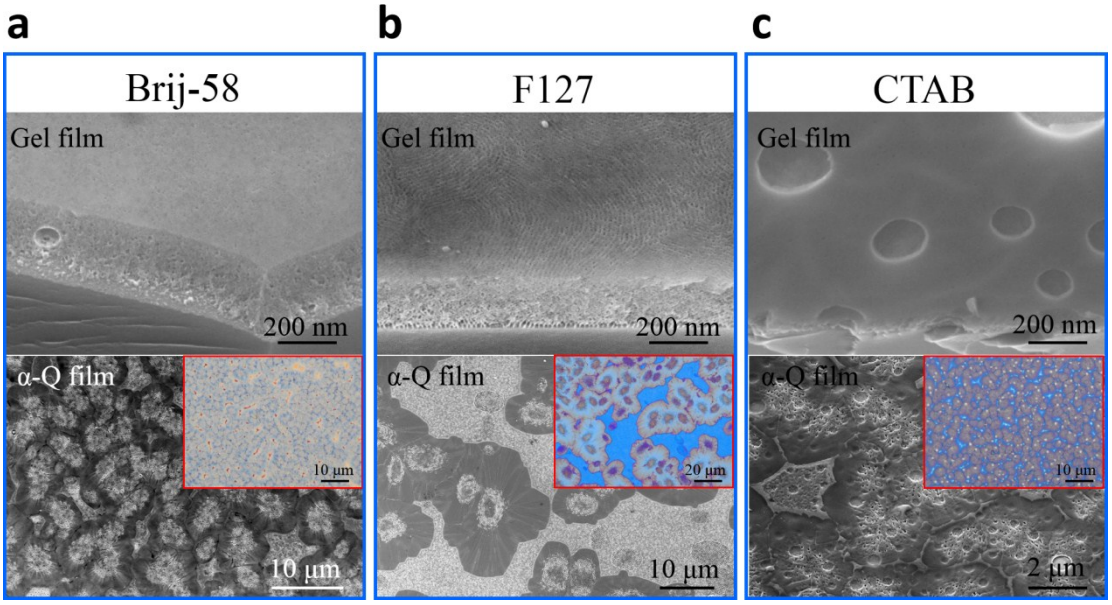


**Figure S9.** SEM pictures of gel films with $\boldsymbol{R}_{Sr}$ = 0.05 (upper panel) and the corresponding annealed films for 300 min at 1000ºC (lower panel) with optical images in as an inset using three different three surfactants: **(a)** Brij-58, **(b)** F127, **(c)** CTAB.

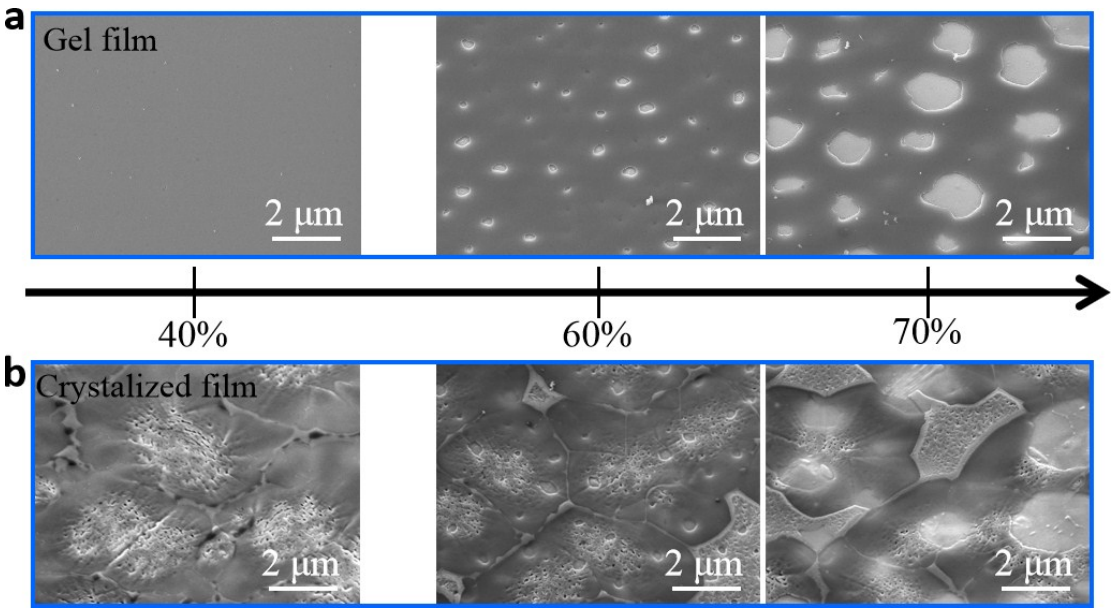


**Figure S10.** SEM pictures of films with $R_{Sr}$ = 0.05 prepared at three different relative humilities indicated in the axis next to the images: **(a)** gel films **(b)** after annealing for 300 min at 1000ºC.

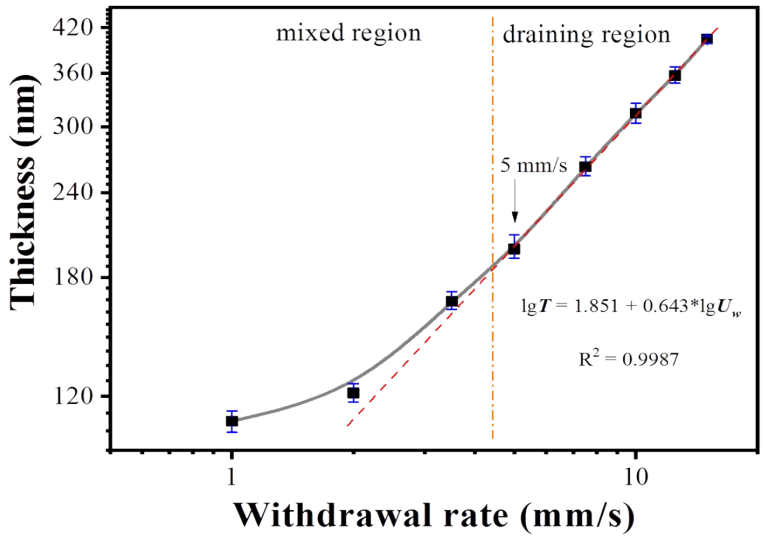


**Figure S11.** Thickness measurement of gel films with $R_{Sr}$ = 0.05 and prepared using different withdrawal speeds showing the increasing gel film thickness with $U_w$: and the different regimes: the viscous drag regime for $U_w$ > 4 mm/s and the mixed regime, for $U_w$ < 4 mm/s.

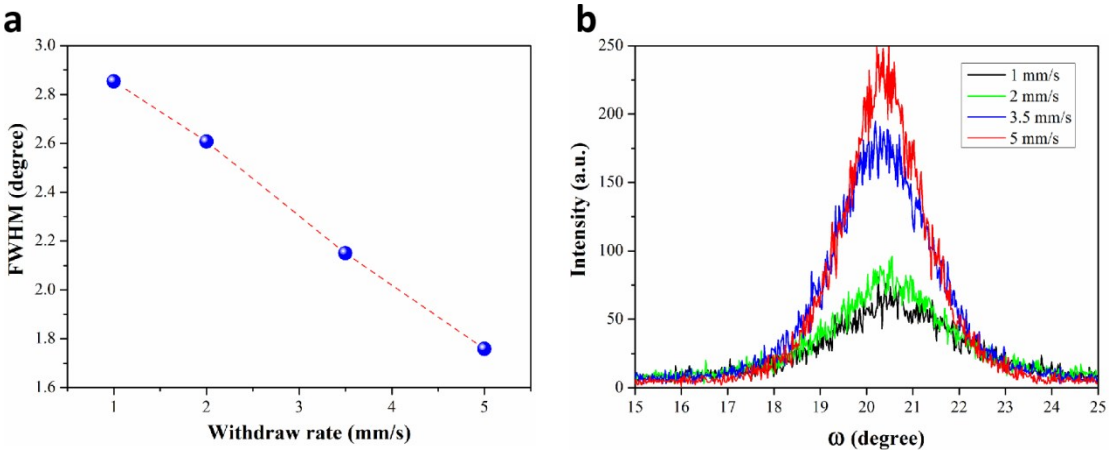


**Figure S12.** The FWHM analysis of $R_{Sr}$ = 0.05 films made by different withdraw rates is shown as **(a)**. Meanwhile, the raw data of rocking curves at α-quartz (100) peak are shown as **(b)**.

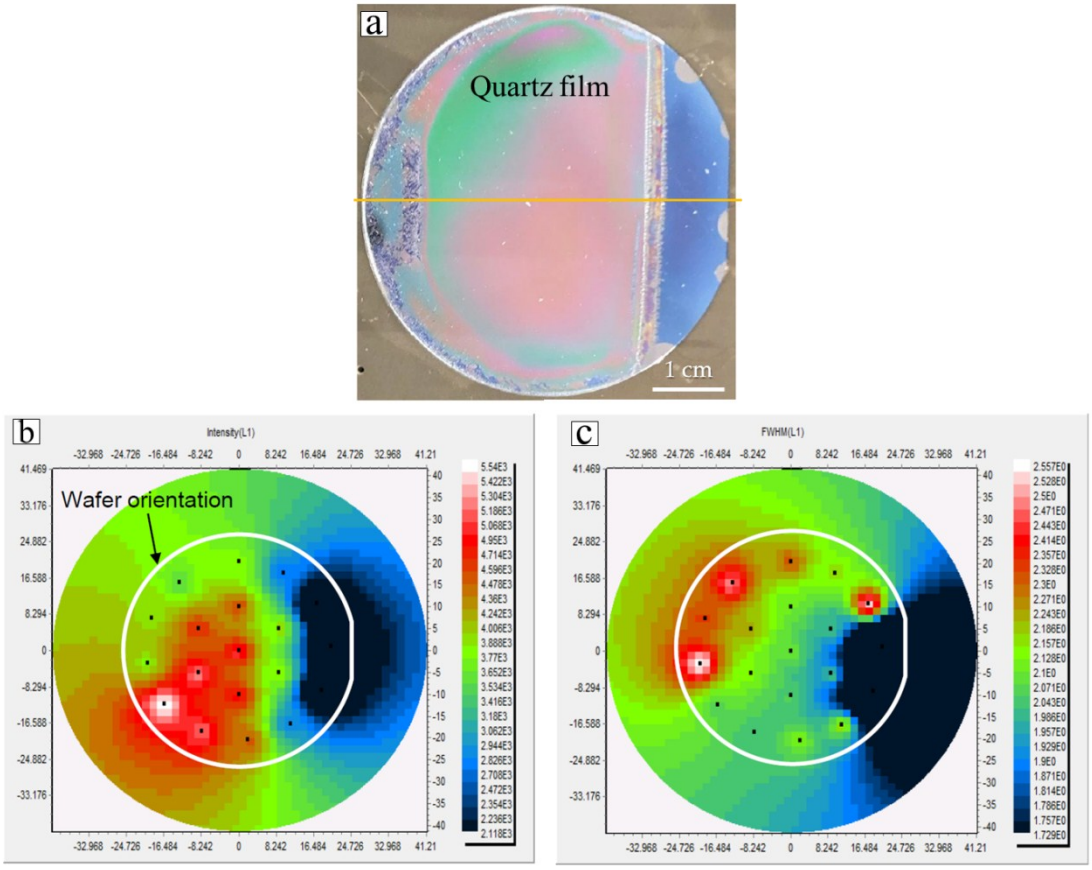


**Figure S13.** A 2 inch epitaxial quartz thin film on silicon substrate fabricated by 5 multilayer deposition, reaching a final thickness of 800 nm **(a)**. Heat map of peak intensity of (100) quartz reflection **(b)**. Heat map of evaluated peak width (FWHM) of rocking curves around quartz (100) reflection **(c)**.

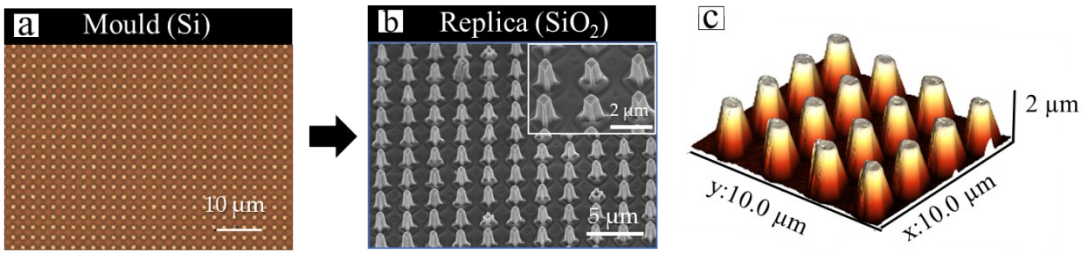


**Figure S14.** **(a)** Optical image of Si (100) masters used along this work obtained by using LIL lithography**. (b)** FEG-SEM image of printed Sr-doped silica columns with a diameter of 1 µm and height of 2 µm on silicon. The inset pictures shows FEG-SEM images at higher magnification of pillars. **(c)** 3D AFM images of the nanostructured silica films prepared by NIL lithography.