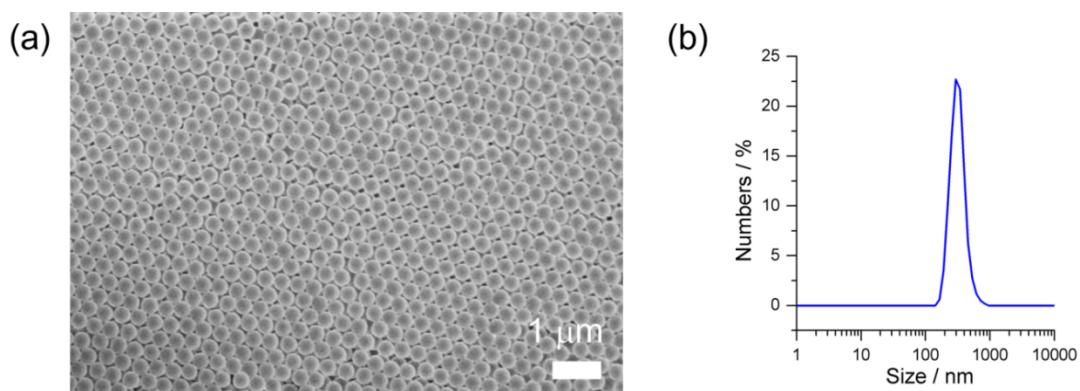


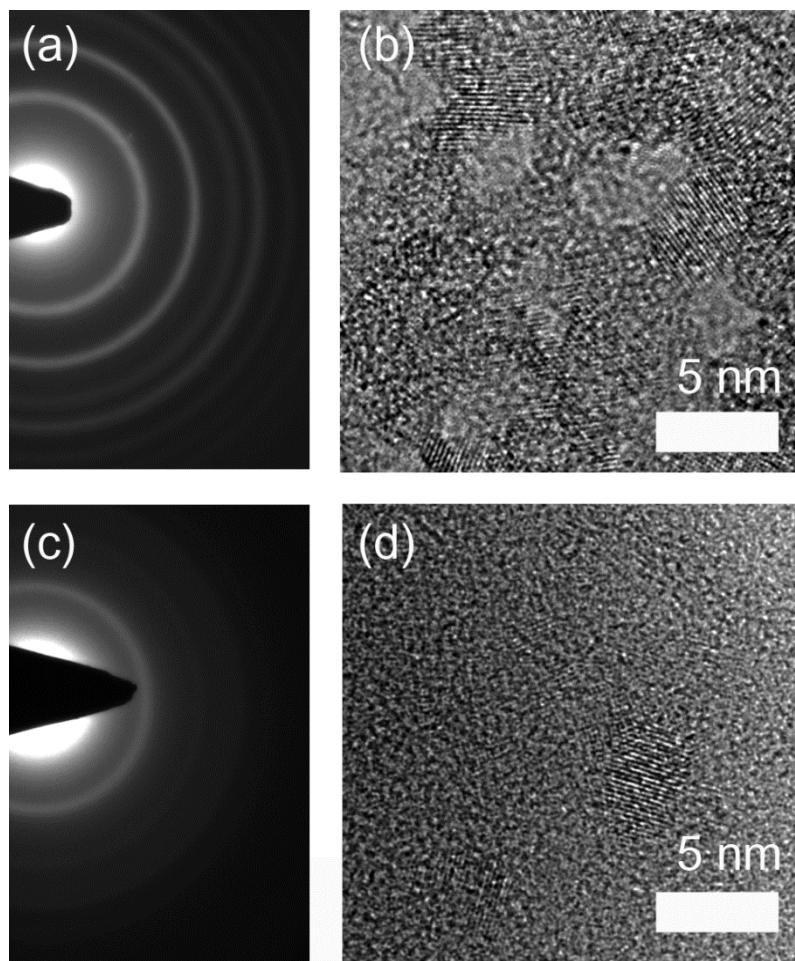
# Supporting Information

## Tuning the crystallinity parameters in macroporous titania films

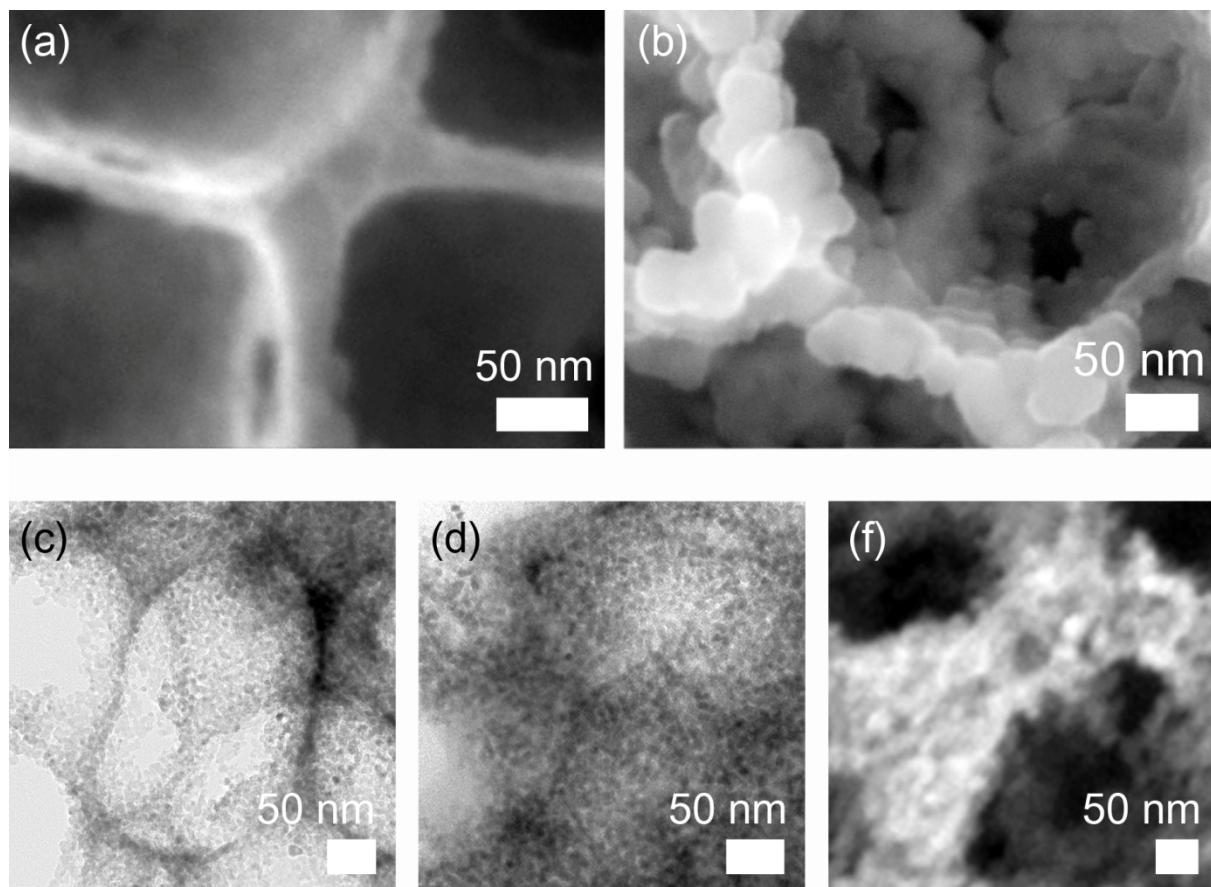
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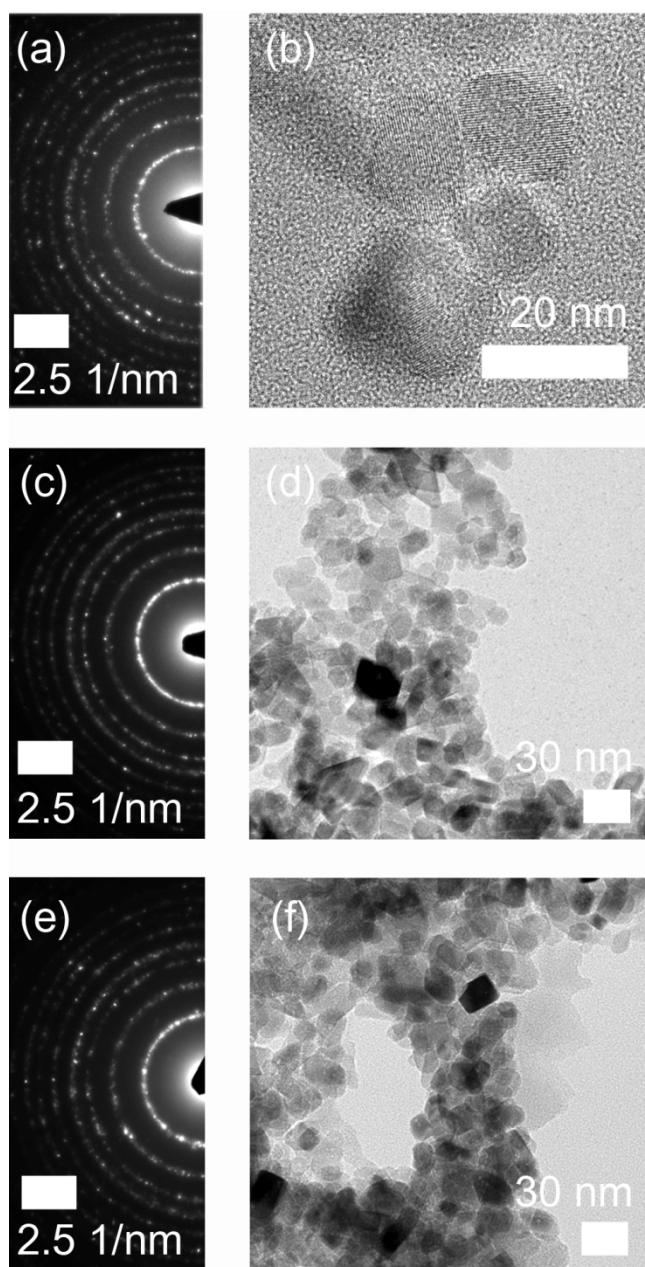
**Figure S1.** SEM top-view image of washed and loosely packed PMMA particles dried on a silicon wafer. Particle diameters are in the range of 320 nm (a, SEM) and a peak maximum of 324 nm was estimated by DLS (b).



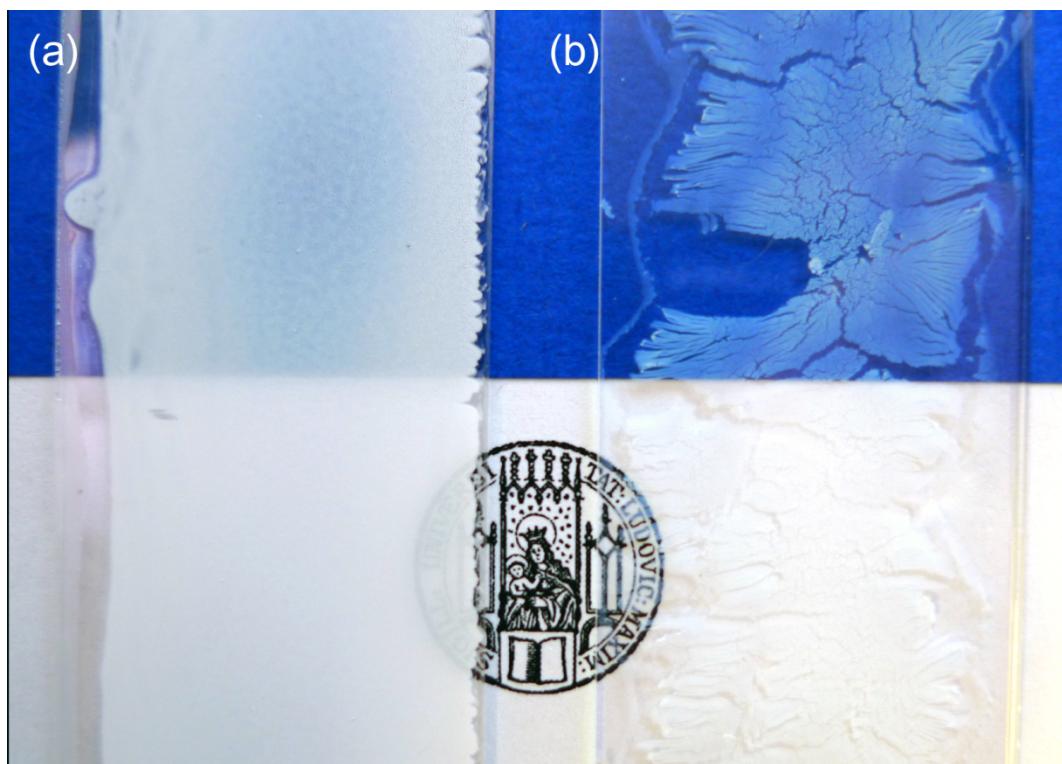
**Figure S2.** SAED and TEM images of small titania nanoparticles NP-6 (a, b) and NP-4 (c, d) obtained after synthesis. Particles were transferred to the TEM grids from solution.



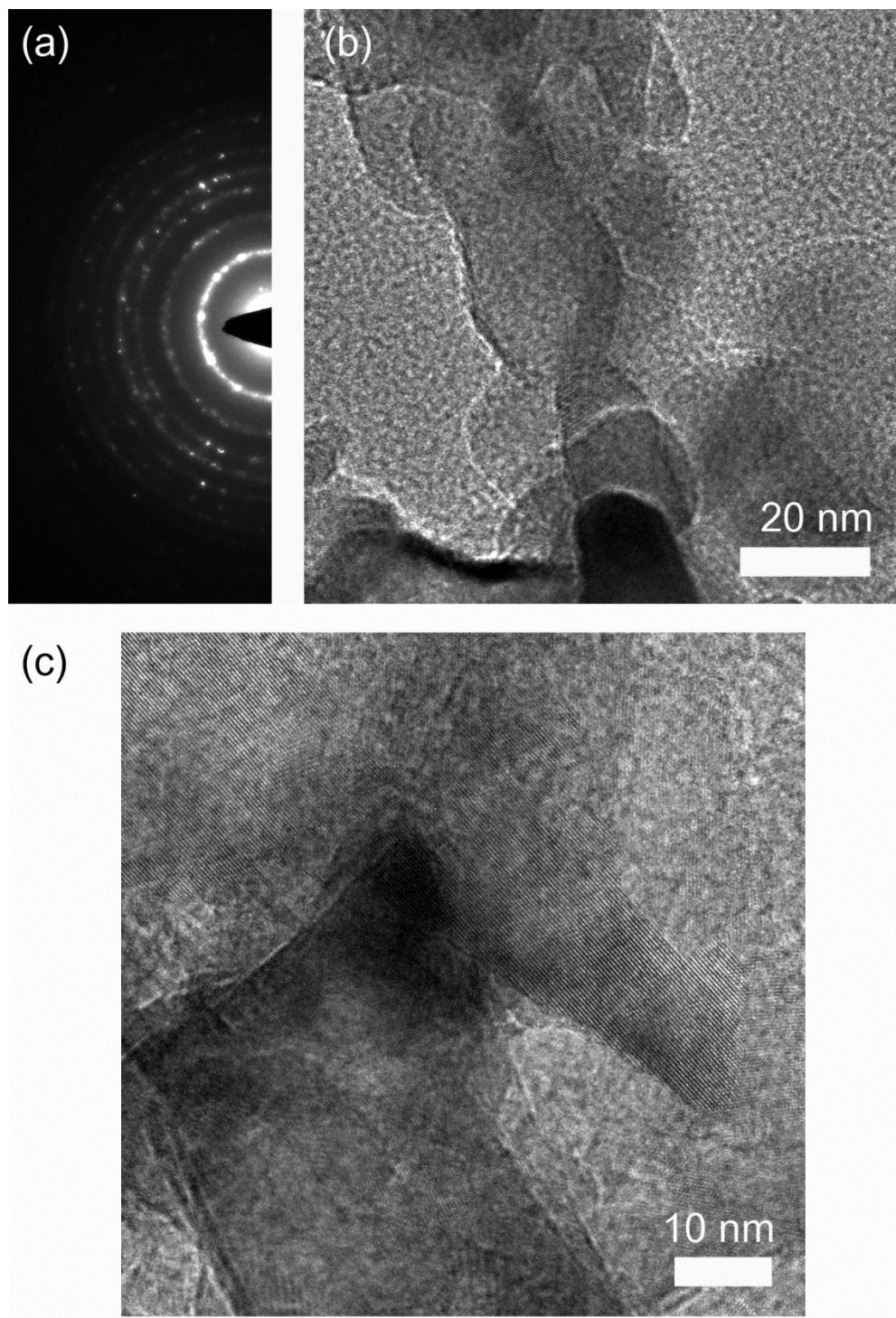
**Figure S3.** Morphology of macroporous titania samples calcined after 400 °C. FE-SEM top-view images of (a) Sol-gel (SG), (b) NP-20-SG and (f) NP-20. TEM images of films removed from the substrate: (c) NP-4 and (d) NP-6.



**Figure S4.** Detailed characterization of **NP-20** using SAED (a, c, e) and TEM (b, d, f). Images (a) and (b) refer to the applied building blocks as prepared after washing. Pictures (c) – (f) depict macroporous titania films that were calcined at 400 °C ((c), (d)) and 500 °C ((e), (f)), respectively, obtained by removal from the substrate. Titania powders were collected, redispersed in ethanol and drop wise transferred to the TEM grids.



**Figure S5.** (a) Stable and semi-transparent macroporous layers composed of **NP-20-SG** ('Brick and Mortar') and (b) delaminated and strongly cracked "film" created with **NP-20** precursor. Both samples were prepared by simple drop-casting on a microscope slide (one-step procedure) and identically calcined at 500°C for 1 h in air. The films are shown on the white and blue background for better contrast.



**Figure S6.** SAED of sol-gel derived macroporous anatase (a) and TEM images of dense polycrystalline anatase walls (b, c). The sample was collected from a **SG** film that was calcined at 500 °C in air.

Table S1: Estimation of particle size of anisotropic anatase depending on the calcination temperature.

Sample	Crystallite size (nm) after calcination at 400 °C and 500°C.						
	Estimation by		TEM		TEM		
	XRD	400 °C	500 °C	400 °C	length	width	
NP-4		8.9	13.4	10 ± 3	8 ± 2	21 ± 10	15 ± 4
NP-6		9.3	16.5	13 ± 2	10 ± 3	16 ± 4	14 ± 2
NP-20		20.3	20.7	20 ± 7	15 ± 6	21 ± 9	15 ± 9
NP-20-SG		16.3	23.1	n.a.		29 ± 8	21 ± 5