

Supplementary information: AC-assisted deposition of aggregate free silica films with vertical pore structure

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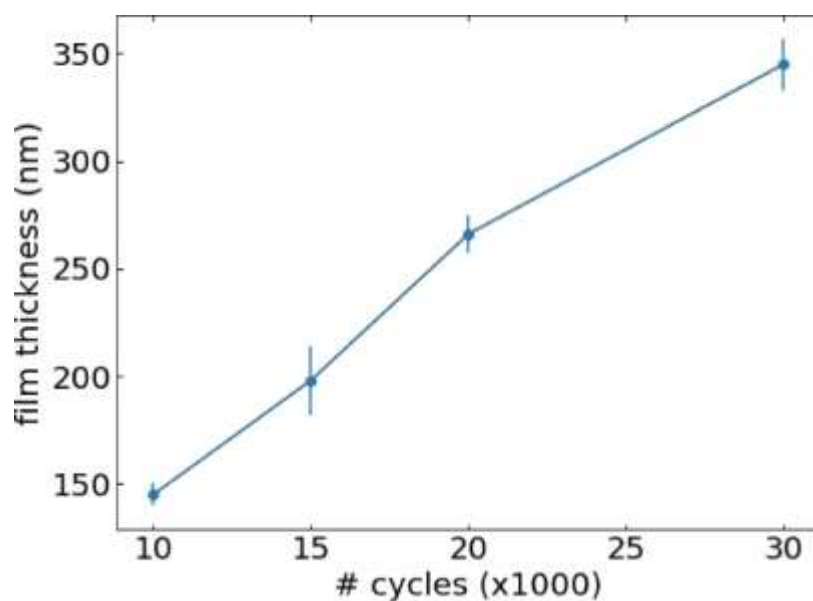


Figure S1: Film thickness versus number of periods extracted from the images shown in Figure 1 using the software ImageJ.¹

Table S1: Diameters of spherical particles from films deposited at increasing number of periods starting from 10,000.

#periods	Particle diameter (nm)	Standard variance (nm)
10,000	385	7
20,000	586	43
30,000	617	120
40,000	828	163

Table S2: Lateral peak positions of features shown in Figure 3 a-c) as well as their ratios showing the hexagonal ordering (1:√3:2).

#periods	$q_0 \pm 0.15$ (nm^{-1})	$q_1 \pm 0.15$ (nm^{-1})	$q_2 \pm 0.15$ (nm^{-1})	q_1/q_0
10000	1.70	2.95	3.42	1.74
20000	1.69	2.93	3.37	1.73
30000	1.68	2.92	3.37	1.74

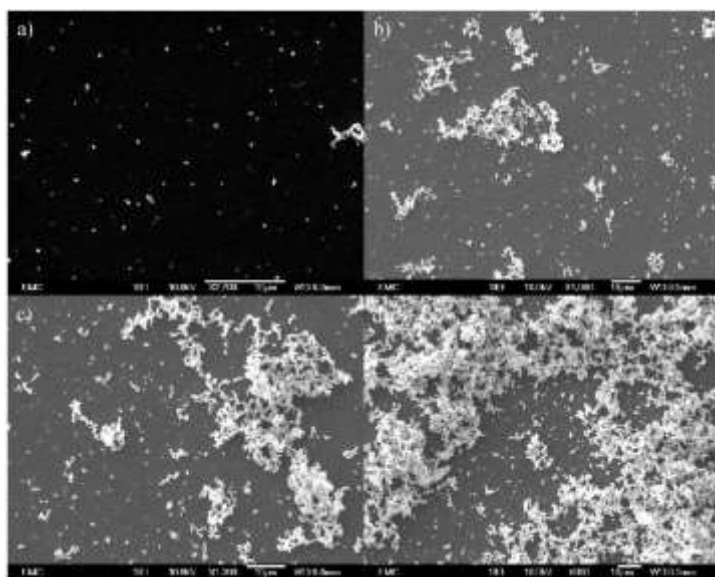


Figure S2: SEM images of control samples made by applying a sinusoidal potential to a TiN electrode immersed in a regular silica sol. $f=100$ Hz, potential, -1 V to -2 V, a) 10k, b) 15k, c) 20k, d) 30k cycles.

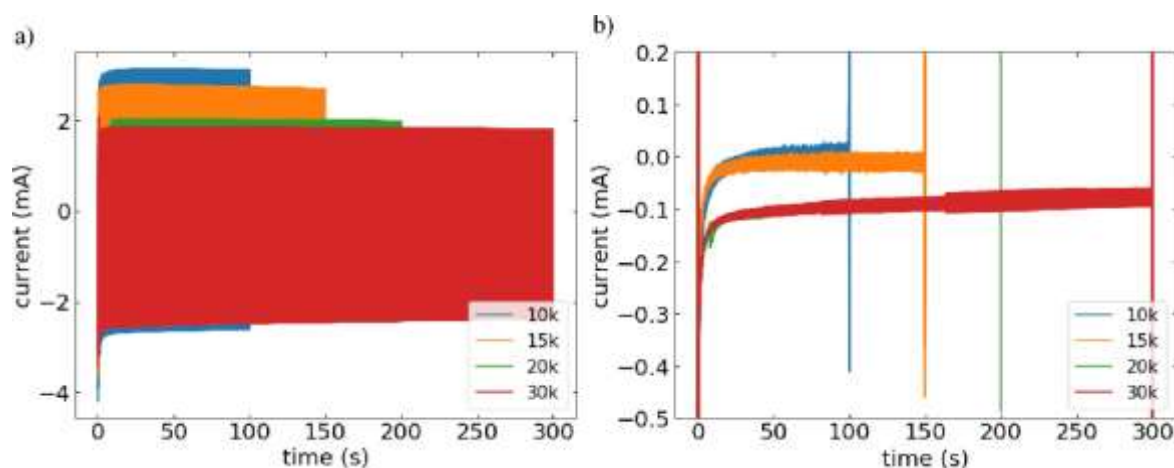


Figure S3: Filtered current response during silica deposition a regular silica sol using a sinusoidal signal of $f=100$ Hz, potential, -1 V to -2 V, (a) 10k, 15k, 20k and 30k periods. (b) A custom Python script was used to employ a bandstop filter at 100 Hz, $Q=30$ to the raw data.

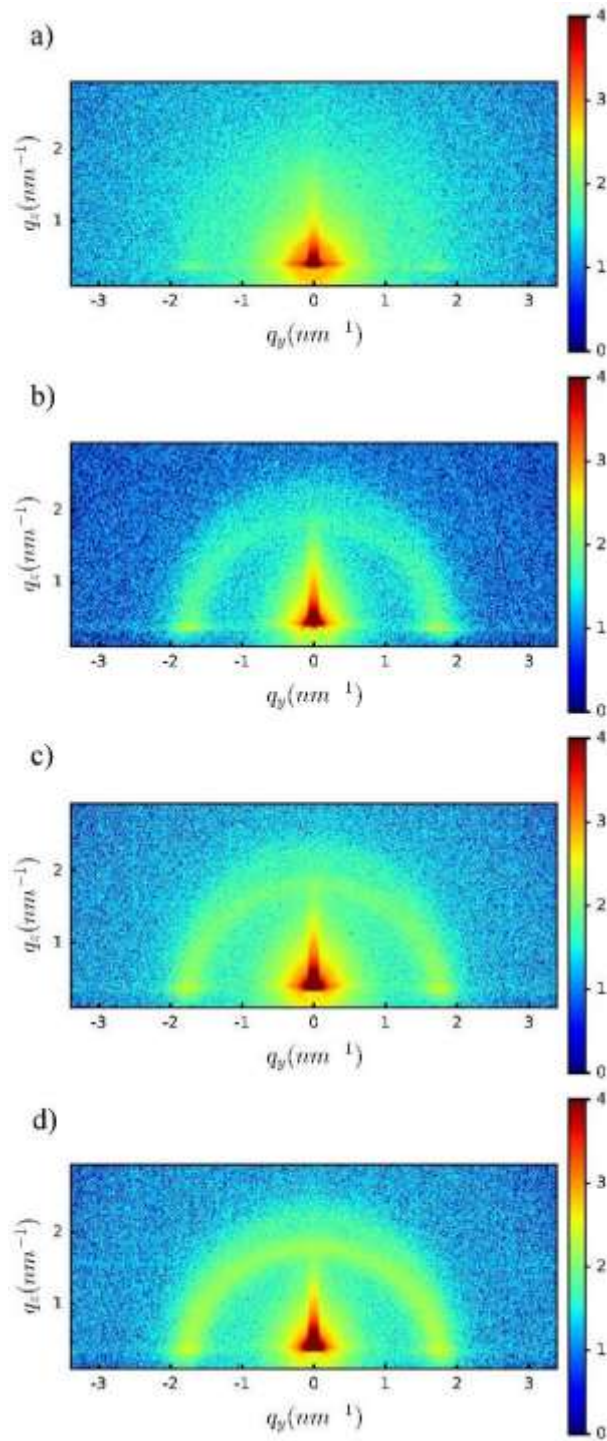


Figure S4: 2D GISAXS scattering images of the control samples taken on our Rigaku Smartlab at an angle of 0.3° for a duration of 30 min for the samples deposited with a) 10k, b) 15k, c) 20k, and d) 30k periods.

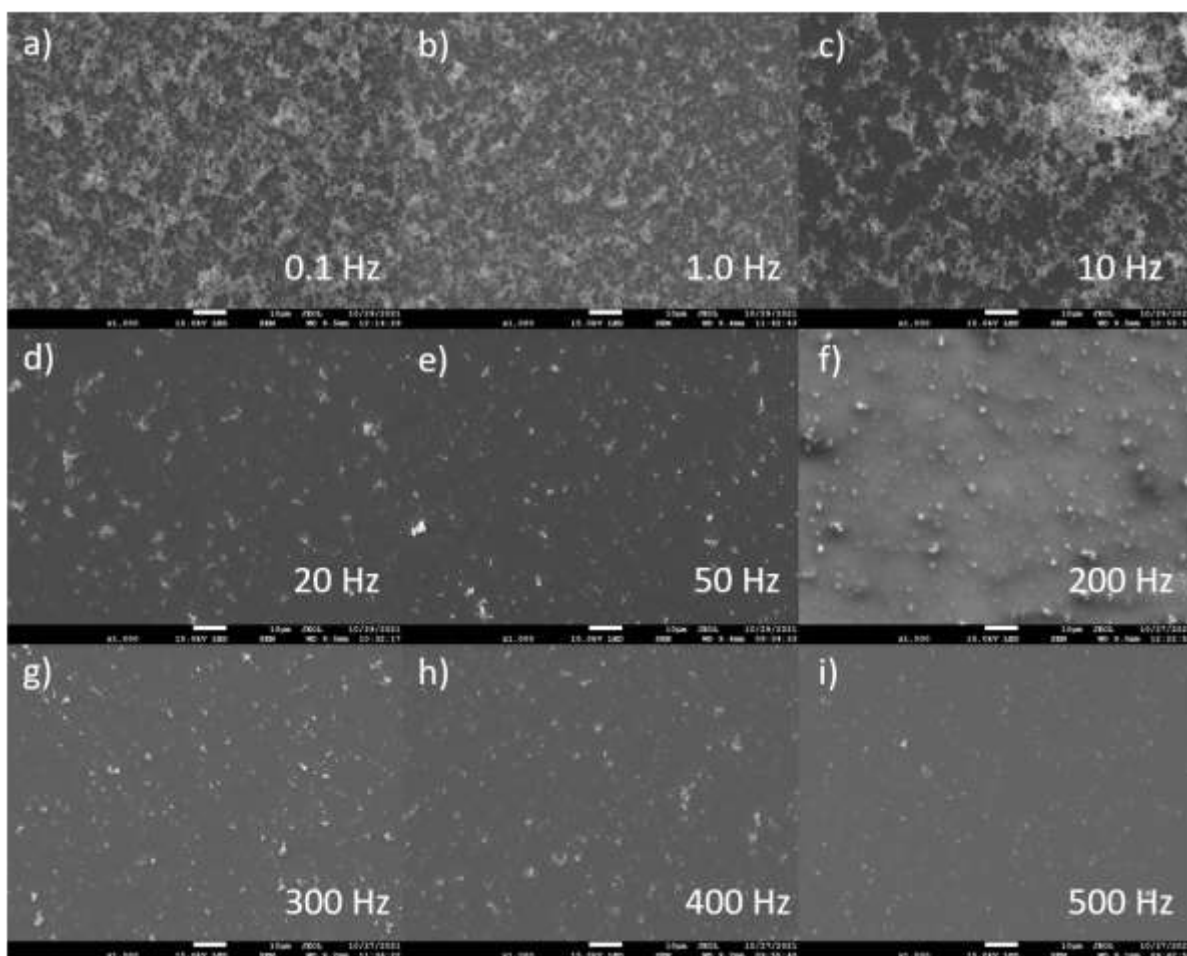


Figure S5: SEM images taken of samples deposited using a sinusoidal potential at frequencies of a) 0.1, b) 1, c) 10, d) 20, e) 50, f) 200, g) 300, h) 400 and i) 500 Hz in a nitrate free silica sol.

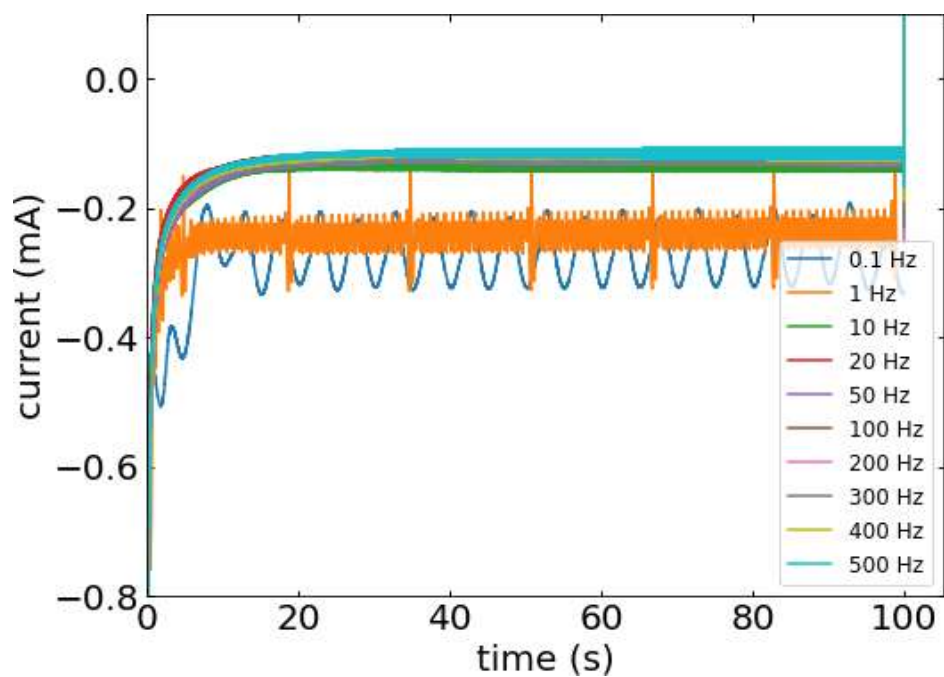


Figure S6: Filtered chronoamperometry data for samples deposited at various frequencies for 100 s each.

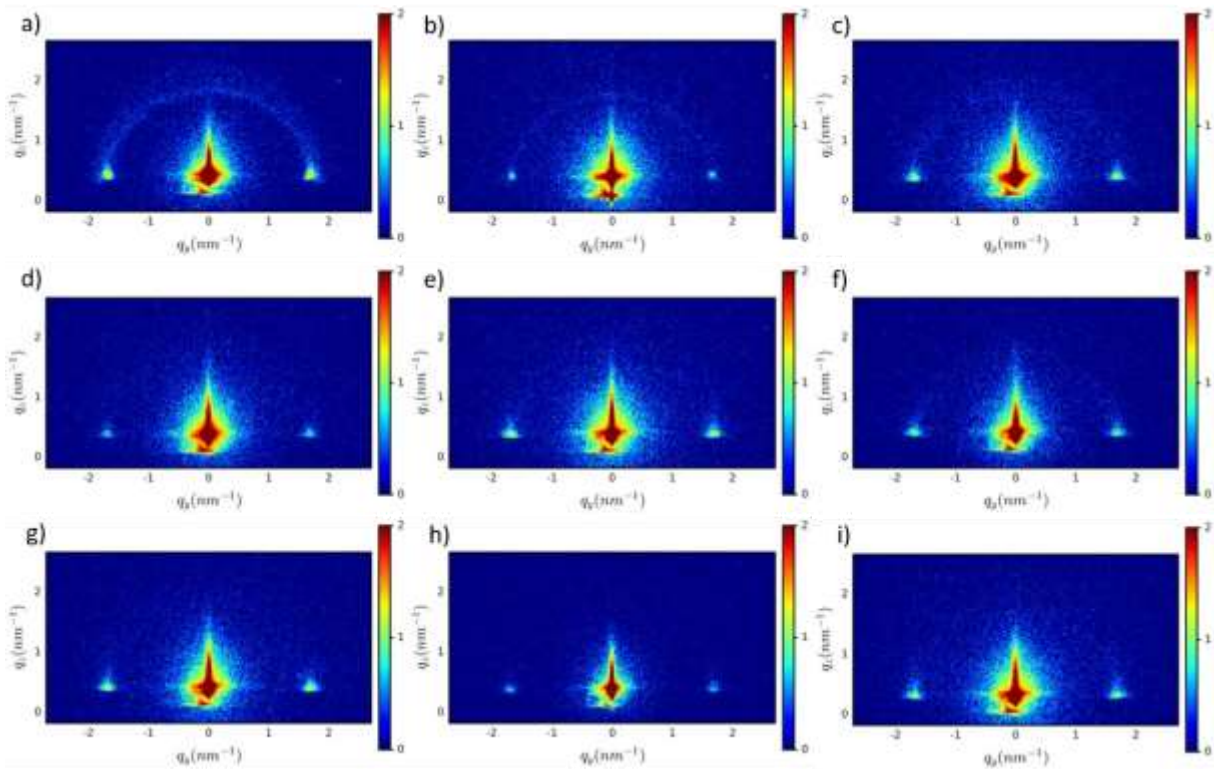


Figure S7: Two-dimensional GISAXS data of samples deposited using a sinusoidal potential at frequencies of a) 0.1, b) 1, c) 10, d) 20, e) 50, f) 200, g) 300, h) 400 and i) 500 Hz in a nitride free silica sol. Incident angle 0.3° , exposure time 15 min.

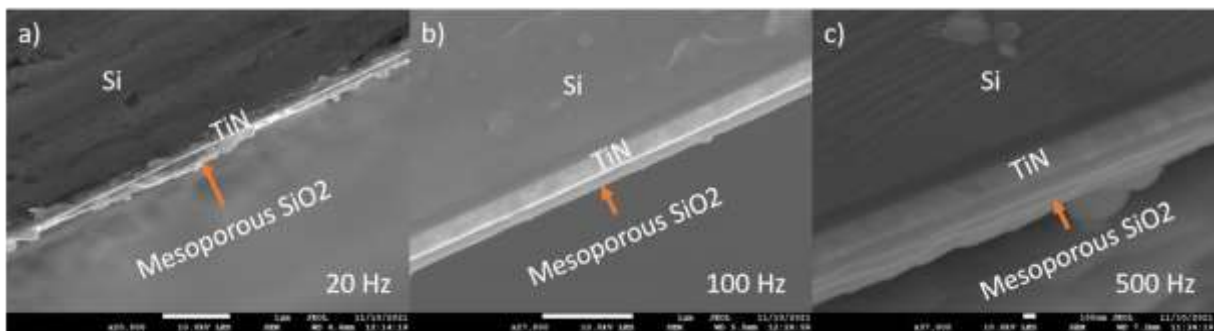


Figure S8: SEM cross-sections from silica samples deposited at a) 20, b) 100 and c) 500 Hz of sinusoidal potential for 100 s each.

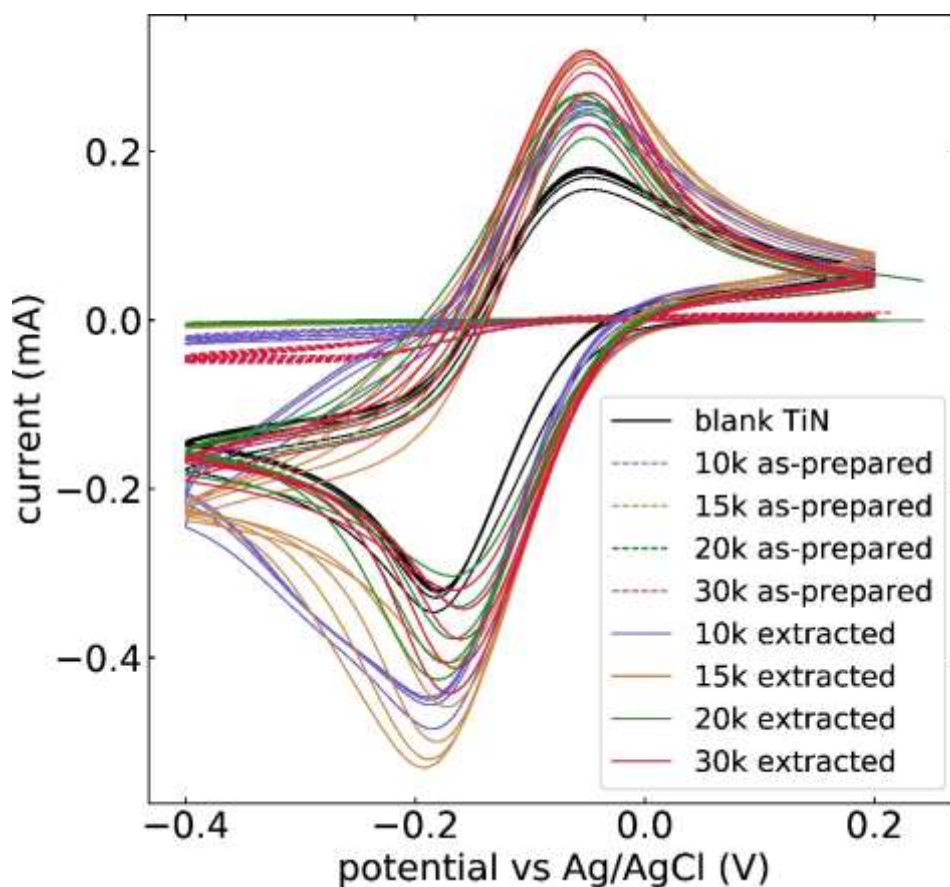


Figure S9: Cyclic voltammetry of mesoporous silica films prepared at 100 Hz on TiN in 5 mM $[Ru(NH_3)_6]Cl_3$ with 0.1M $NaNO_3$ between -0.4 V and 0.2 V vs Ag/AgCl at a scan rate of 20 mV/s, five cycles each. Solid black line shows response of a blank TiN substrate with an electrode area of 5 mm x 15 mm. Coloured dashed lines show the current response before, solid ones after, surfactant removal.

1 M. D. Abràmoff, P. J. Magalhães and S. J. Ram, *Biophotonics Int.*, 2004, **11**, 36–41.