

**Clay platelet orientation inside self-standing beidellite clay films:
effect of silica nanospheres and link with macroscopic mechanical
resistance**

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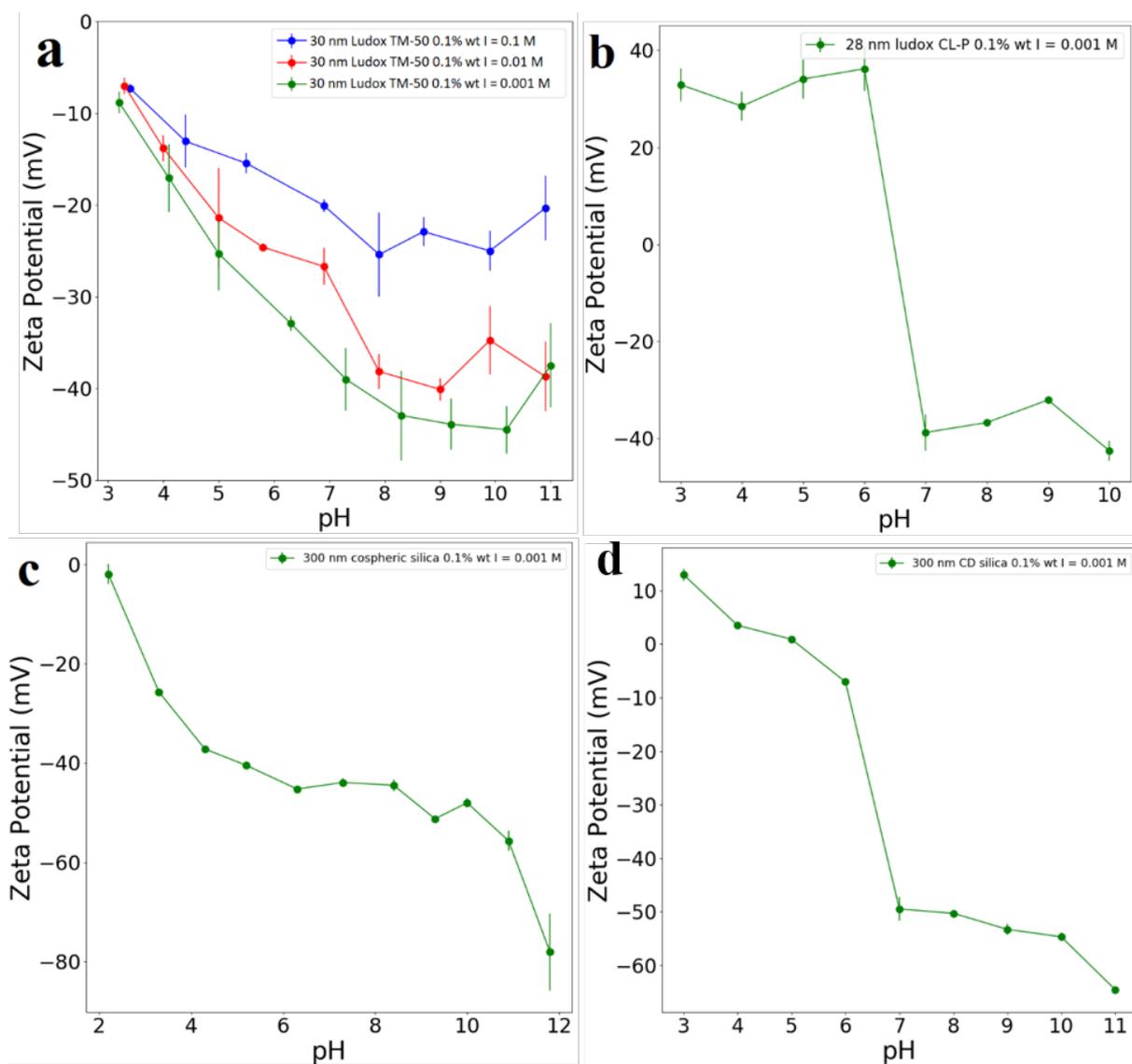


Figure. S1 : Evolution with pH of the zeta potential of silica spheres in deionized water. Solid concentration 0.1 % wt. (a) Ludox TM-50 (b) Ludox CL-P (c) Cospheric silica. (d) CD silica.

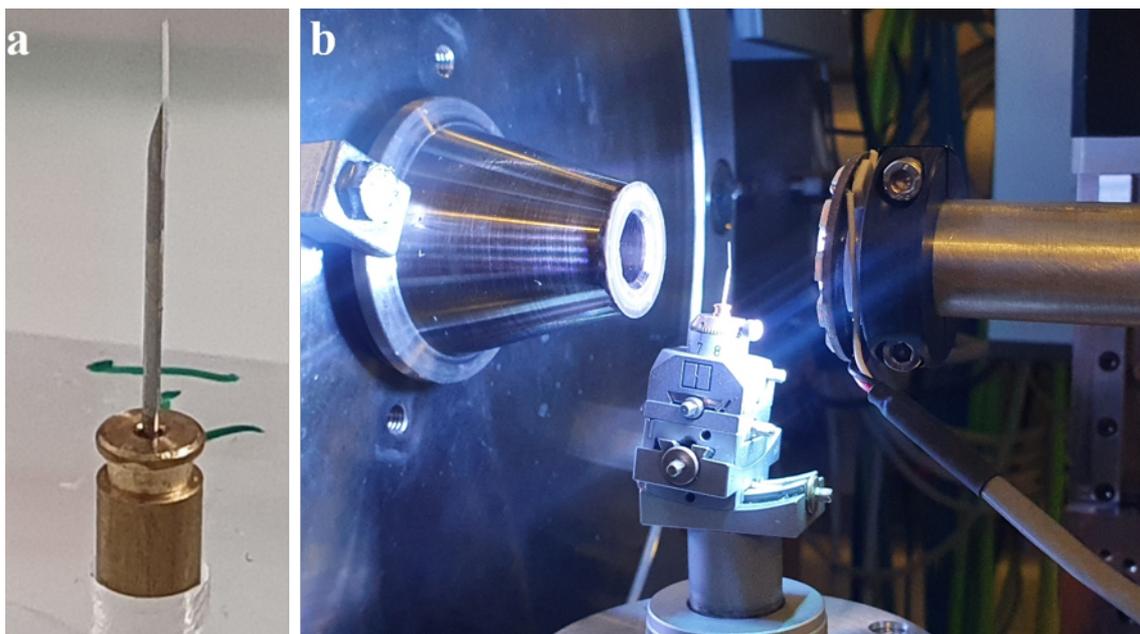


Figure. S2 : Illustration procedure for SAXS measurements. (a) Self-standing film of few tenths of microns cut into rectangular slate and stuck to a needle's top. (b) Setup with goniometer head for SAXS measurements at different rotation angles of the films.

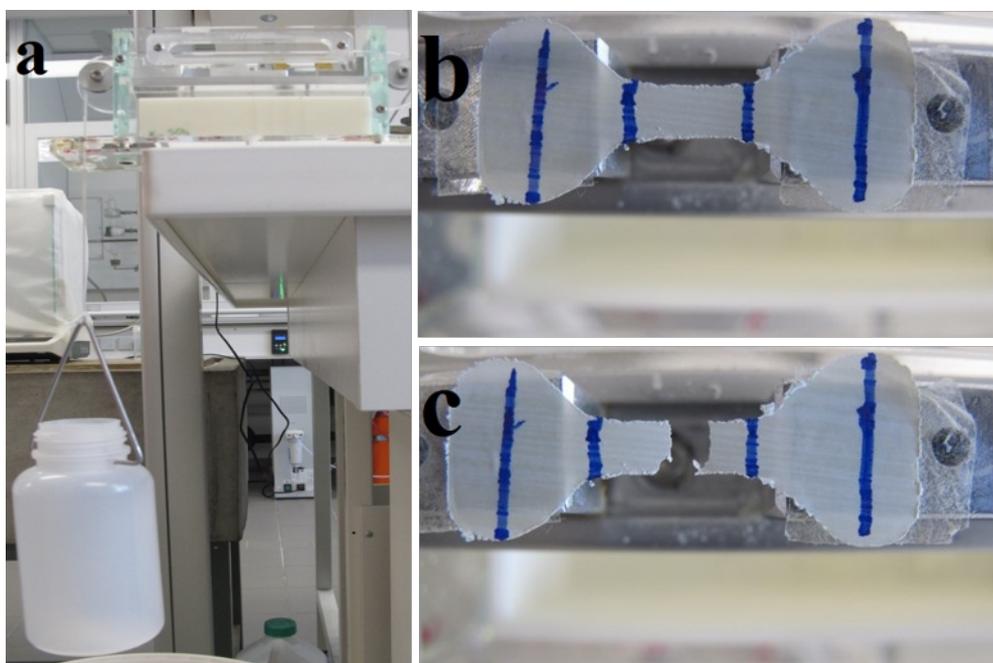


Figure. S3 : Tensile strength experiments. (a) Setup used to estimate the breaking stress of self-standing. (b) Positioning of films on the setup. (c) Films after breakup.

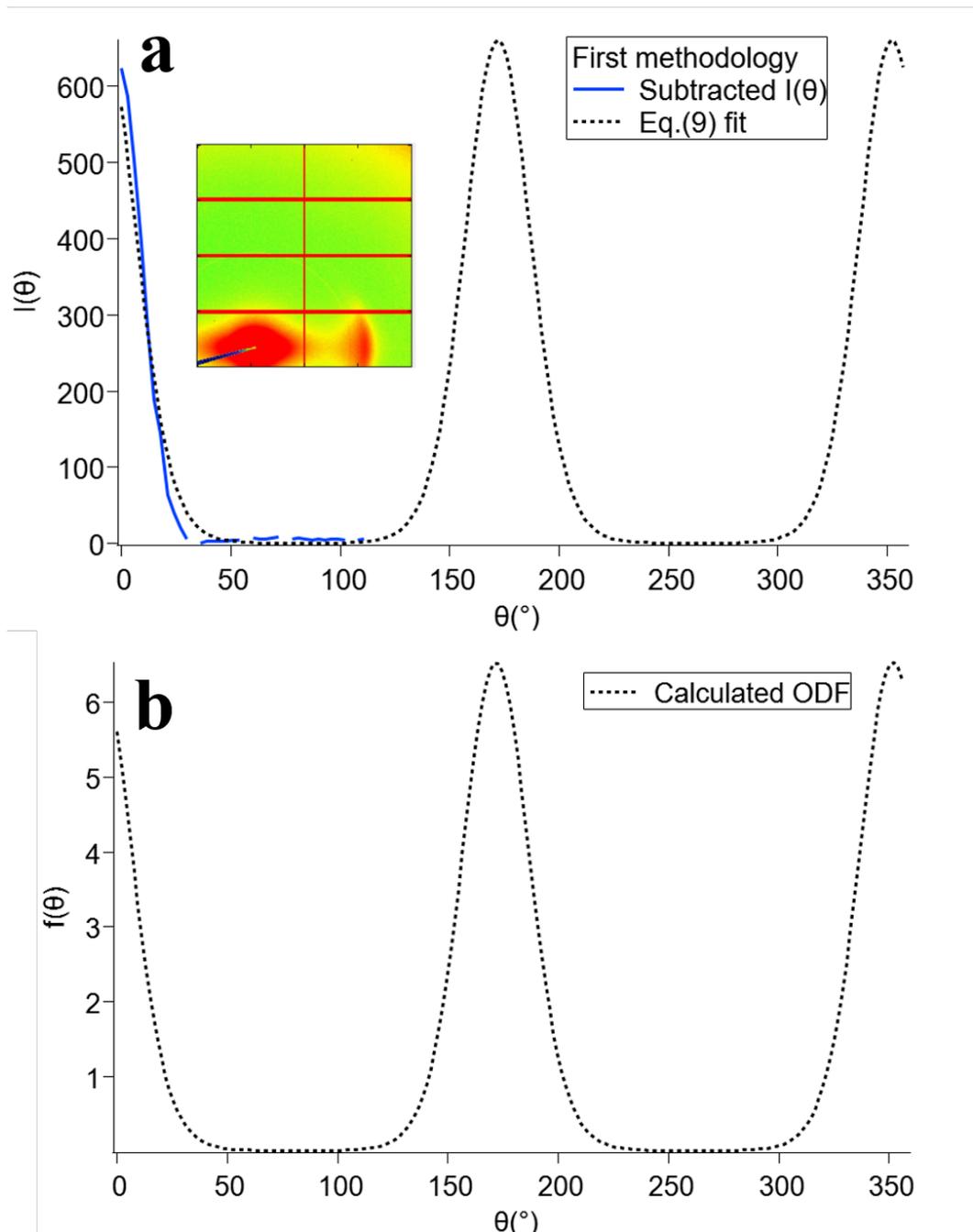


Figure. S4 : SAXS measurements for deposits on mylar and methodology. (a) 2D SAXS pattern: the entire angular range (0° - 360°) was not available for this detector position. Subtracted intensity modulation $I(\theta)$ obtained by the first methodology had many missing points. Thereby, it was fitted by eq.(9) to get the full $I(\theta)$ angle range. (b) Fitted $I(\theta)$ allowed deriving ODF $f(\theta)$ from eq.(7) and estimating $\langle P_2 \rangle$ according to eq.(8).



Figure. S5 : Deposited self-supported films prepared for tensile strength measurements. Numerous deposits had a broken structure after drying at room temperature.

Tableau. S6 : Number of trials for the different samples in tensile strength measurements at similar temperature and relative humidity with R_n the number ratio of sphere/platelet.

Sample	Number of trials
T1 beidellite	10
T1 beidellite + ludox CL-P $R_n = 0.333$	7
T1 beidellite + ludox CL-P $R_n = 1.000$	6
T1 beidellite + ludox CL-P $R_n = 3.000$	0
T1 beidellite + ludox TM-50 $R_n = 0.333$	7
T1 beidellite + ludox TM-50 $R_n = 1.000$	5
T1 beidellite + ludox TM-50 $R_n = 3.000$	0
T1 beidellite + CD silica $R_n = 0.002$	4
T1 beidellite + CD silica $R_n = 0.005$	3
T1 beidellite + CD silica $R_n = 0.016$	0
T1 beidellite + cospheric silica $R_n = 0.002$	8
T1 beidellite + cospheric silica $R_n = 0.005$	6
T1 beidellite + cospheric silica $R_n = 0.016$	6
T3 beidellite	8
T3 beidellite + ludox CL-P $R_n = 0.333$	5
T3 beidellite + ludox CL-P $R_n = 1.000$	5
T3 beidellite + ludox CL-P $R_n = 3.000$	0
T3 beidellite + ludox TM-50 $R_n = 0.333$	4
T3 beidellite + ludox TM-50 $R_n = 1.000$	7
T3 beidellite + ludox TM-50 $R_n = 3.000$	0
T3 beidellite + CD silica $R_n = 0.002$	0
T3 beidellite + CD silica $R_n = 0.005$	0
T3 beidellite + CD silica $R_n = 0.016$	0
T3 beidellite + cospheric silica $R_n = 0.002$	5
T3 beidellite + cospheric silica $R_n = 0.005$	5
T3 beidellite + cospheric silica $R_n = 0.016$	0

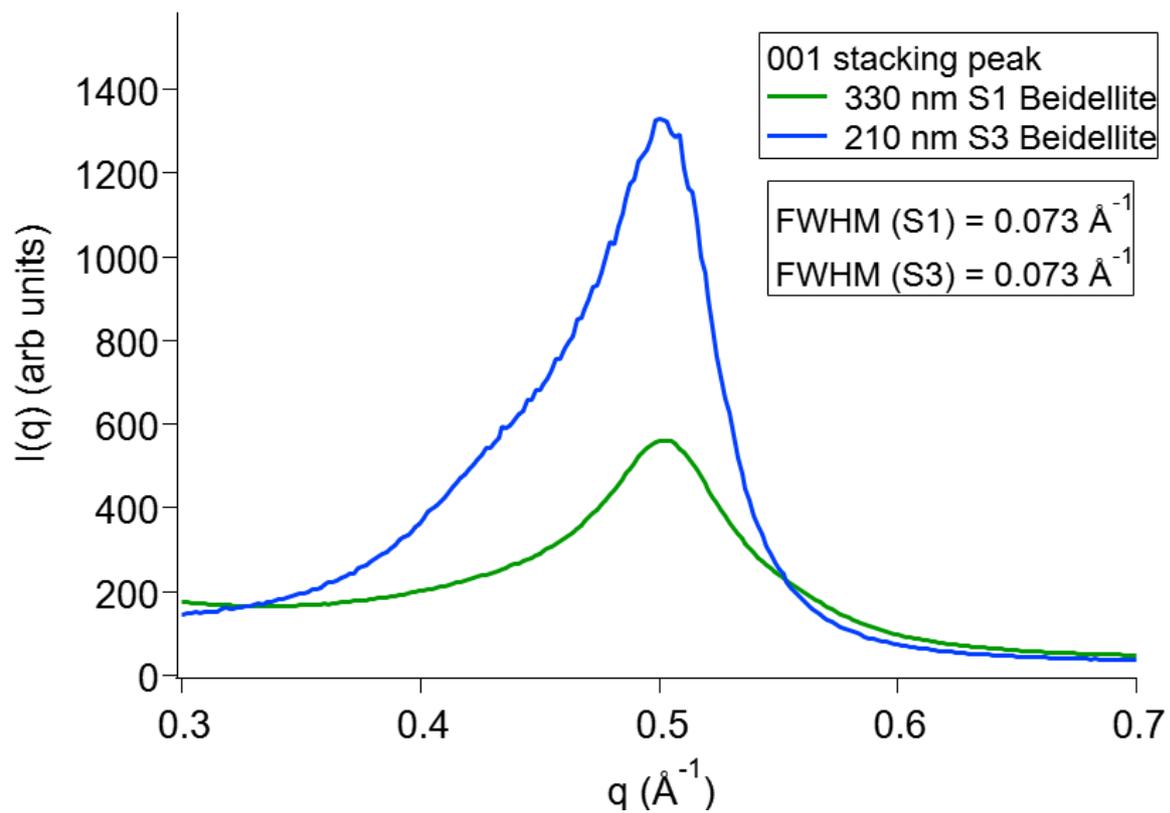


Figure. S7 : 001 stacking peak of S1 and S3 beidellite films without spheres. The full width half maximum (FWHM) is equal for the two films of different platelets sizes.