## Supplementary Information

# Impact of wormlike micelles on nano and macroscopic structure of TEMPO-oxidized cellulose nanofibrils hydrogels

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Figure ES1 – Surface tension dependence on CAPOX concentration in NaCl 1wt% solutions.



Table ESI 1A Fitting parameters for CAPB 20, 40 and 60 mM in NaCl 1wt% in 100 %

	20 mM	40 mM	60 mM
Surfactant	20 1110		
CAPB			
Scale (∝n 〖∆ρ〗 ^2)	1.22	2.48	3.75
R / Å	24±1	24 (fixed)	24 (fixed)
σ/%	15 ±1	15 (fixed)	15 (fixed)
	Interactions (h	nard-spheres)	
φ <sub>HS</sub> / %	0	1.3 ±0.1	2.4 ±0.1
R <sub>HS</sub> / Å	N/A	36 ± 1	32 ± 1

 $D_2O$ 

#### Table ESI 1B Fitting parameters for CAPOx 20, 40 and 60 mM in NaCl 1wt% in 100

% I	$D_2O$
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Concentration	20 mM	40 mM	60 mM	
Surfactant				
CAPOx				
Scale (∝n 〖∆ρ〗 ^2)	1.32	2.64	4.16	
R / Å	23±1	23 (fixed)	23 (fixed)	
σ/%	17 ±1	17 (fixed)	17 (fixed)	
Interactions (hard-spheres)				
<b>ф</b> нѕ / %	0	0	2.9 ±0.1	
R <sub>HS</sub> / Å	N/A	N/AN/A	31 ± 1	

Table ESI 1C Fitting parameters for SLS 20, 40 and 60 mM in NaCl 1wt% in 100 %

#### $D_2O$

Concentration	20 mM	40 mM	60 mM	
Surfactant				
SLS				
Scale (∝n 〖∆ρ〗 ^2)	2.24	1.78	1.62	
R / Å	19±1	19±1	19±1	
Interactions (Hayter – mean sphere approximation)				
Charge / e	54 (fixed)	54 (fixed)	54±2	
Vol. frac / %	0.23±0.01	0.62±0.01	1.05±0.01	
Dieletric constant	77.936 (fixed)	77.936 (fixed)	77.936 (fixed)	
Salt concentration /	0.173 (fixed)	0.173 (fixed)	0.173 (fixed)	
Μ				
Temperature / K	298.16 (fixed)	298.16 (fixed)	298.16 (fixed)	

Table ESI 2A Fitting parameters for CAPB/SDS 20/20, 40/40 and 60/60 mM in NaCl

Concentration	20 mM	40 mM	60 mM	
Surfactant				
CAPB/SDS				
Scale (∝n 〖∆ρ〗 ^2)	0.05	0.10	0.17	
R / Å	18±1	18 (fixed)	18 (fixed)	
σ/%	19 ±1	19 ±1 (fixed)	19 ±1 (fixed)	
Interactions (Random Phase Approximation)				
V <sub>RPA</sub>	16.2±1	45 ±1	82 ±0.1	
R <sub>c</sub> / Å	49 ±1	50 ± 1	46 ± 1	

1wt% in 100 % D<sub>2</sub>O

#### Table ESI 2B Fitting parameters for CAPOx/SDS 20/20, 40/40 and 60/60 mM in

NaCl	1wt%	in	100	%	$D_2O$
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Concentration	20 mM	40 mM	60 mM
Surfactant			
CAPOx/SDS			
Scale (∝n 〖∆ρ〗 ^2)	0.05	0.10	0.15
R / Å	18±1	18 (fixed)	18 ±1 (fixed)
σ/%	19 ±1	19 ±1 (fixed)	19 ±1 (fixed)
Interactions (Random Phase Approximation)			
v <sub>RPA</sub> /±1b	64 ±0.1	95 ±0.1	145 ±0.1
R₀ /±0.1 Å	32 ± 1	35 ± 1	37 ± 1

### Table ESI 2C Fitting parameters for CAPB/SLS 20/20, 40/40 and 60/60 mM in NaCl

#### 1wt% in 100 % D<sub>2</sub>O

Concentration	20 mM	40 mM	60 mM		
Surfactant					
CAPB/SLS					
Scale (∝n 〖∆ρ〗 ^2)	2.69	2.18	1.54		
R / Å	22 (fixed)	22 (fixed)	22 ±1		
Interactions (charged-spheres)					
Charge / e	76 (fixed)	76 (fixed)	76±1		
Vol. frac / %	0.8±0.1	1.5±0.1	2.2±0.1		
Dieletric constant	77.936 (fixed)	77.936 (fixed)	77.936 (fixed)		
Salt concentration /	0.173 (fixed)	0.173 (fixed)	0.173 (fixed)		
М					
Temperature / K	298.16 (fixed)	298.16 (fixed)	298.16 (fixed)		

Table ESI 3A Fitting parameters for the OCNF contribution in CAPB/SDS 20/20,40/40 and 60/60 mM in OCNF 1wt%/NaCl 1wt% in 15wt% D2O / 75wt% H2O

Concentration	20 mM	40 mM	60 mM	
Surfactant				
CAPB				
Scale (∝n 〖∆ρ〗 ^2)	0.0098	0.0085	0.0068	
R / Å	53±1	53 (fixed)	53 (fixed)	
3	0.24 ±0.02	0.24 (fixed)	0.24 (fixed)	
Interactions (Random Phase Approximation)				
V <sub>RPA</sub>	-1.9±0.1	-1.9 (fixed)	-1.9 (fixed)	
R <sub>c</sub> / Å	65±1	65 (fixed)	65 (fixed)	