Elastocapillary Self-Assembly

Spring 2019

Sameh Tawfick

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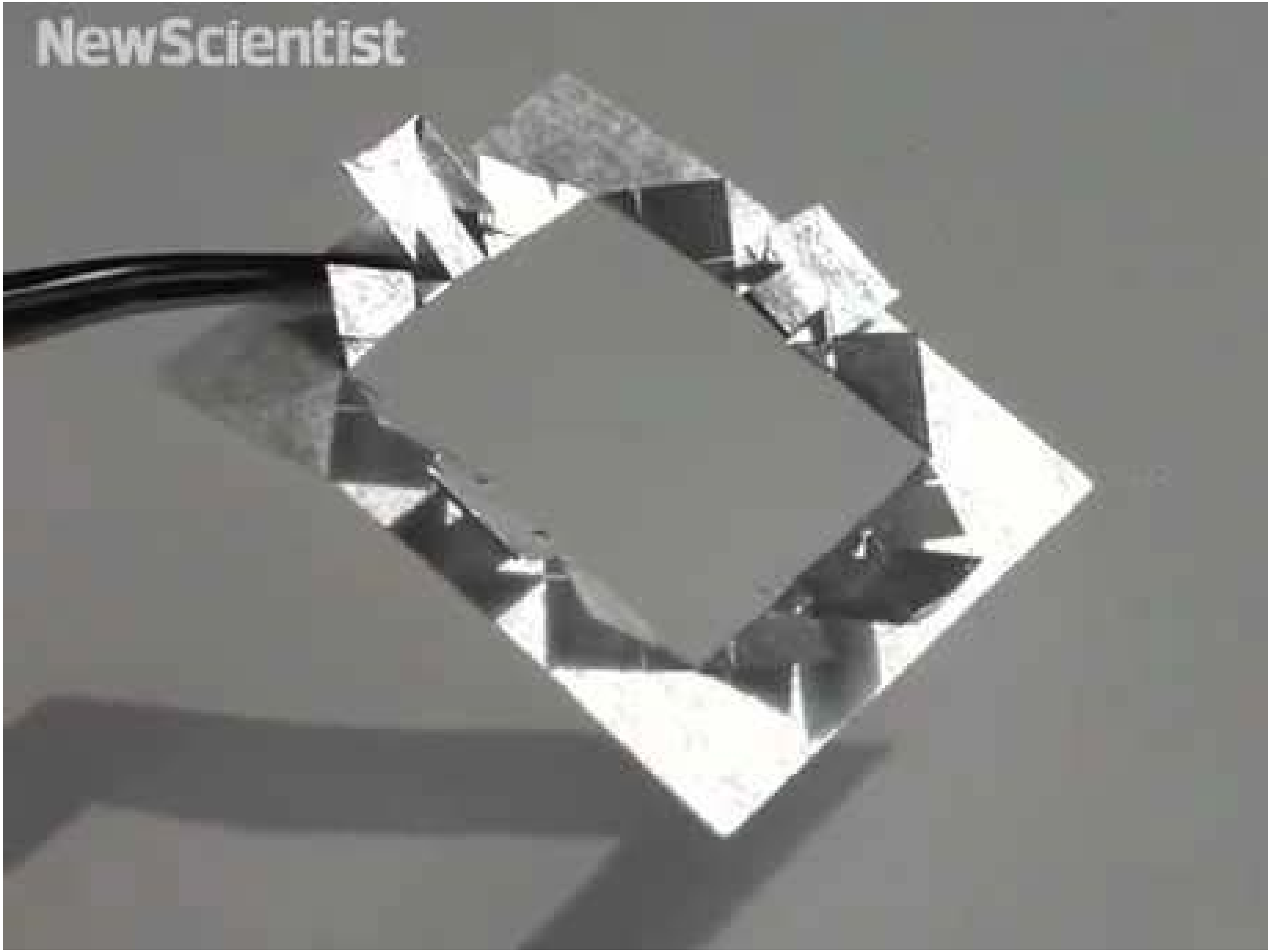
Capillary origami



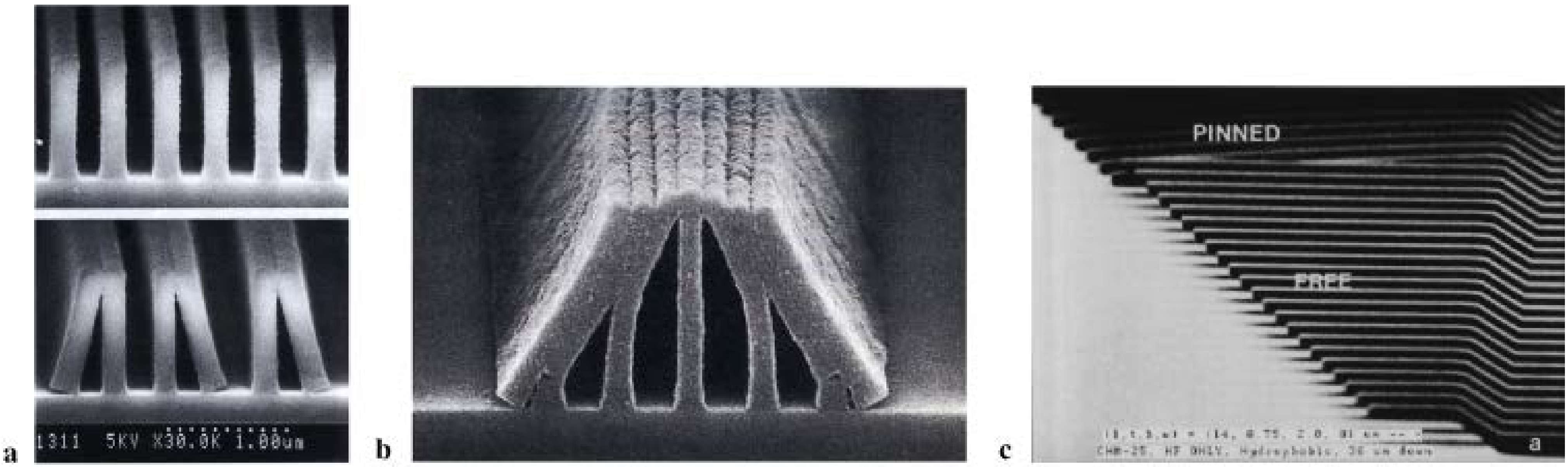
By Etienne Cliquet, Flotille

https://www.youtube.com/watch?v=n51Vi3rv_kA

First observations: Elastocapillary aggregation

First observations took place when high aspect ratio microfabricated pillars aggregated during drying.

Elasto-capillarity was considered nuisance to microfabrication of thin beams and pillars.

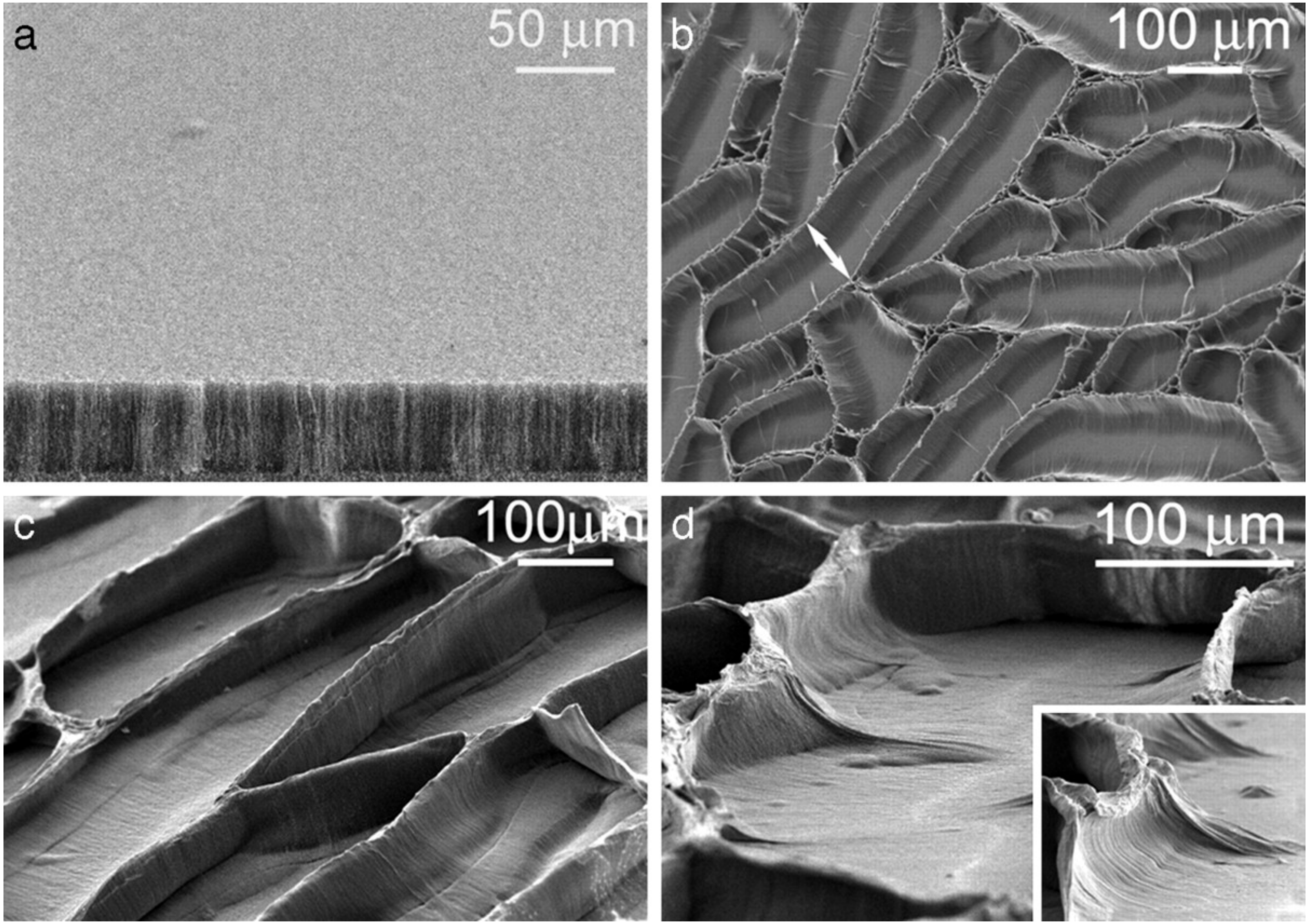


Tanaka T, Morigami M and Atoda N 1993 Mechanism of resist pattern collapse during development process *Japan. J. Appl. Phys.* 1 **32** 6059-64

Mastrangelo C H and Hsu C H 1993 Mechanical stability and adhesion of microstructures under capillary forces-part ii: experiments *J. Microelectromech. Syst.* **2** 33-43

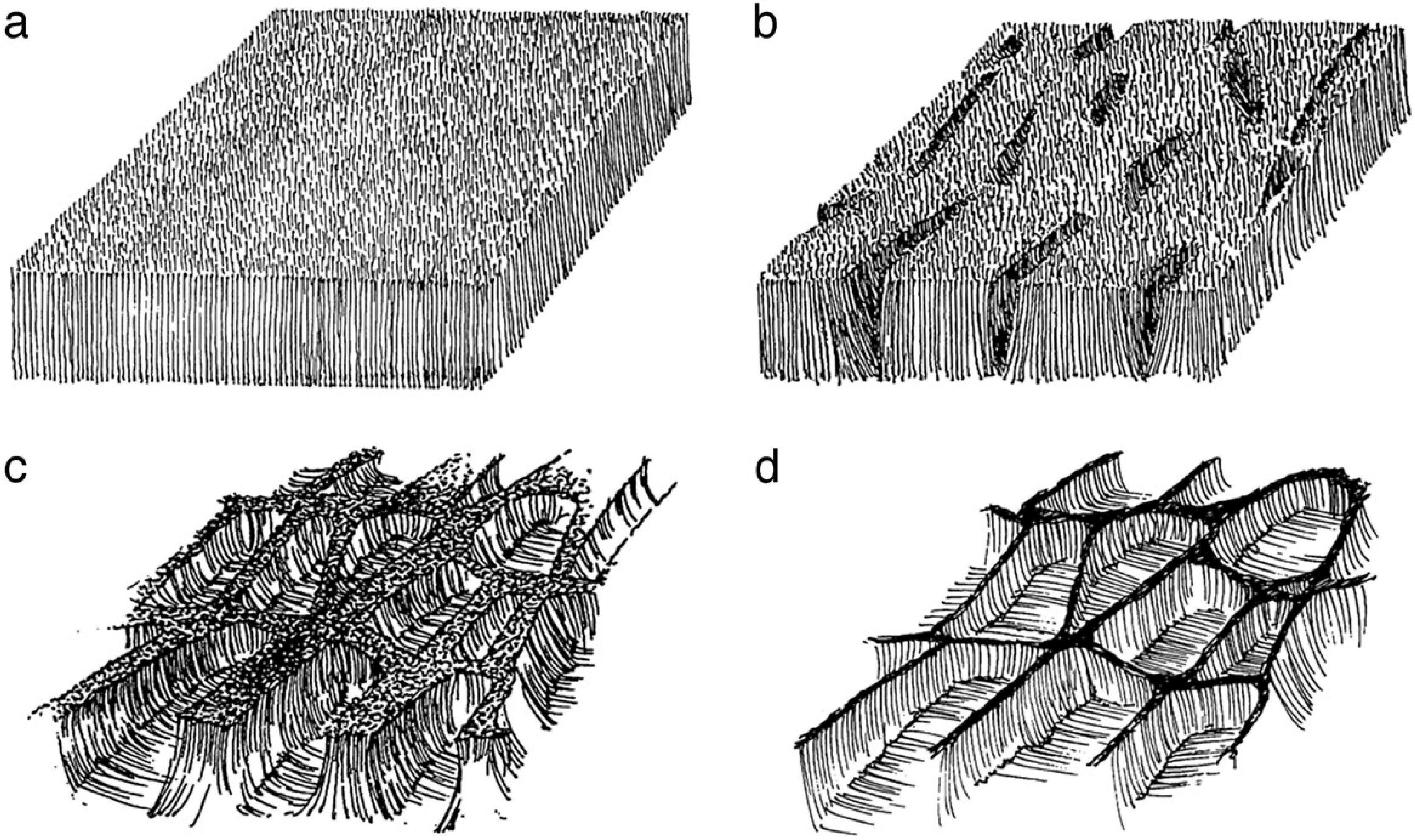
Elastocapillary nuisance
→ Interesting patterns

Interesting patterns were observed after wetting and drying of CNT forests



Chakrapani N, Wei B, Carrillo A, Ajayan P M and Kane R S 2004 Capillarity-driven assembly of two-dimensional cellular carbon nanotube foams *Proc. Natl Acad. Sci. USA* **101** 4009-12

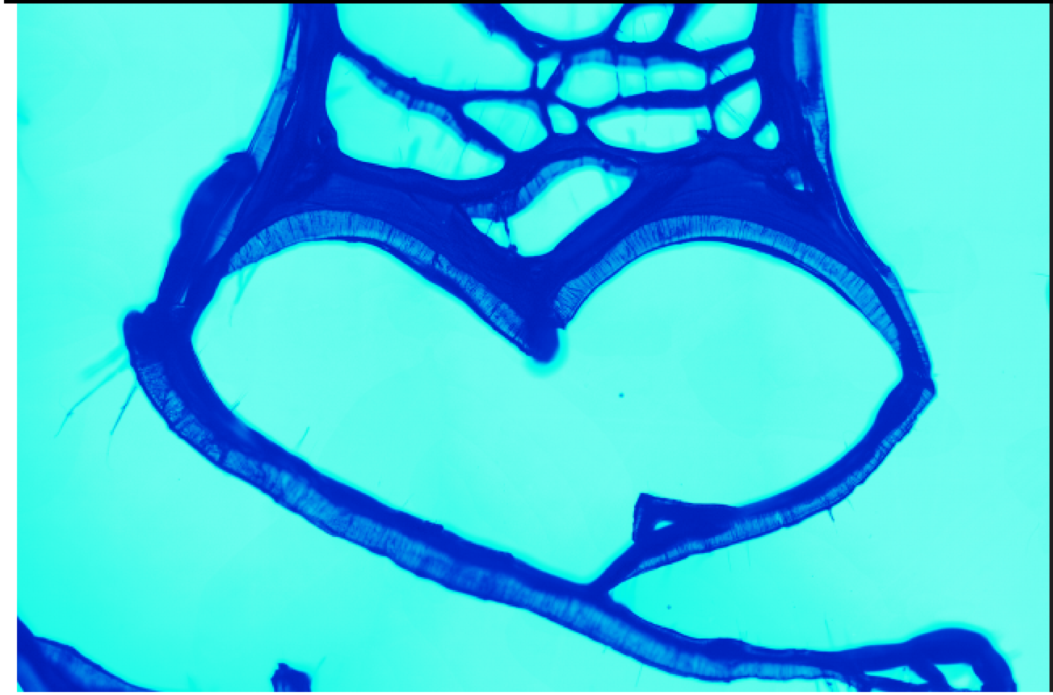
Mechanism of formation of interesting patterns



Mud cracks

MIT News
ON CAMPUS AND AROUND THE WORLD

Browse or Search



FULL SCREEN

A recolored optical image obtained by MIT researchers shows a heart-shaped carbon nanotube cell. A version of the image is featured on the cover of the Feb. 14 print edition of Physical Chemistry Chemical Physics.

Image: Ashley Kaiser and Itai Stein/MIT

Getting to the heart of carbon nanotube clusters

MIT researchers create predictable patterns from unpredictable carbon nanotubes.

Denis Paiste | Materials Research Laboratory
February 14, 2018

Press Inquiries

Integrating nanoscale fibers such as carbon nanotubes (CNTs) into commercial applications, from coatings for aircraft wings to heat sinks for mobile computing, requires them to be produced in large scale and at low cost. Chemical vapor deposition (CVD) is a promising approach to manufacture CNTs in the needed scales, but it produces CNTs that are too sparse and compliant for most applications.

Applying and evaporating a few drops of a liquid such as acetone to the CNTs is an easy, cost-effective method to more tightly pack them together and increase their stiffness, but until

RELATED

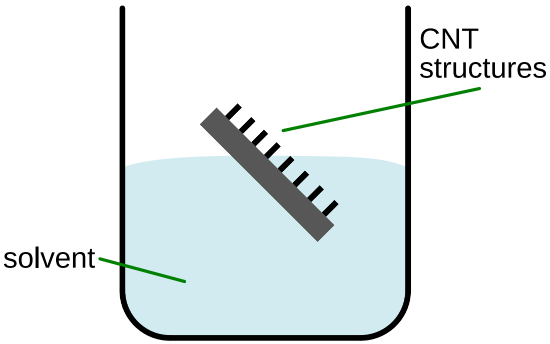
Paper: "Process-morphology scaling relations quantify self-organization in capillary densified nanofiber arrays"

Ashley Kaiser

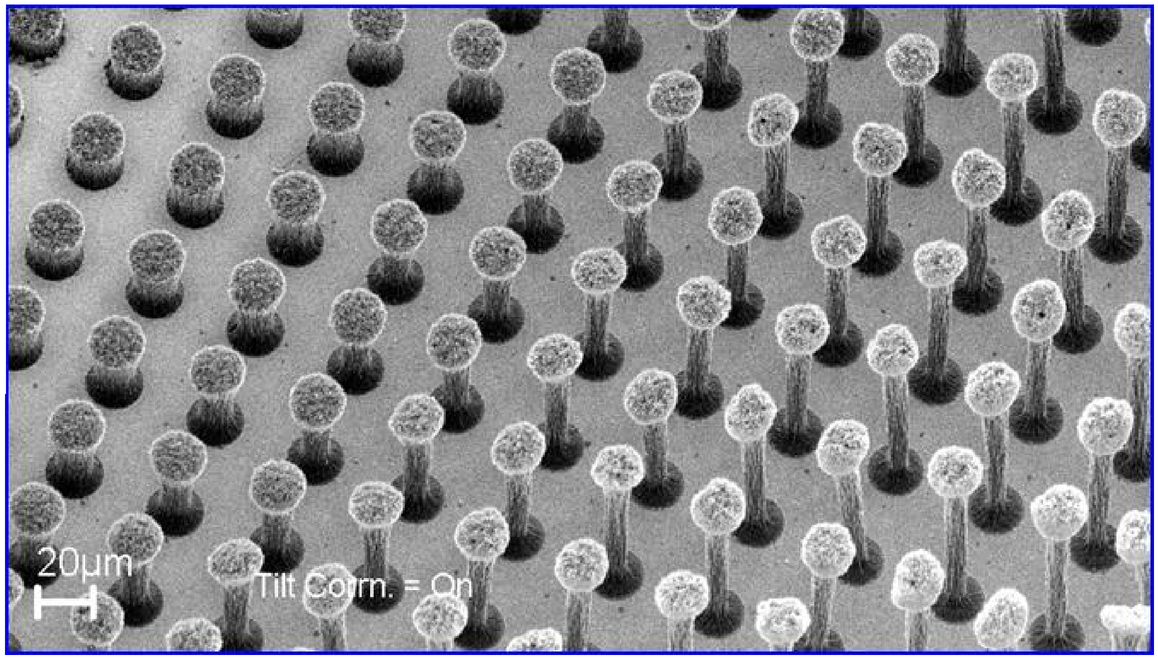
Itai Stein

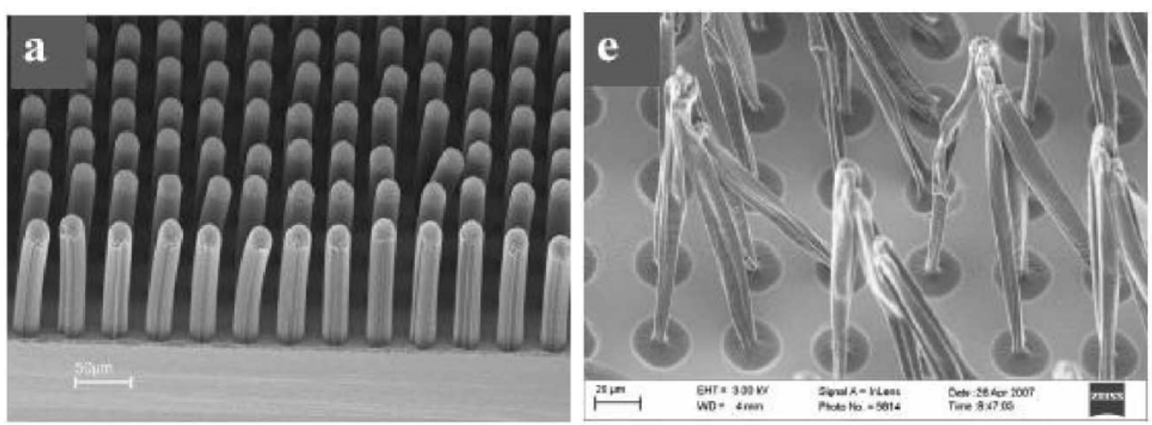
Opportunity: CNT densification by immersion

Low density vertical CNT pillars: high density, strong and more electrically conductive pillars for interconnects



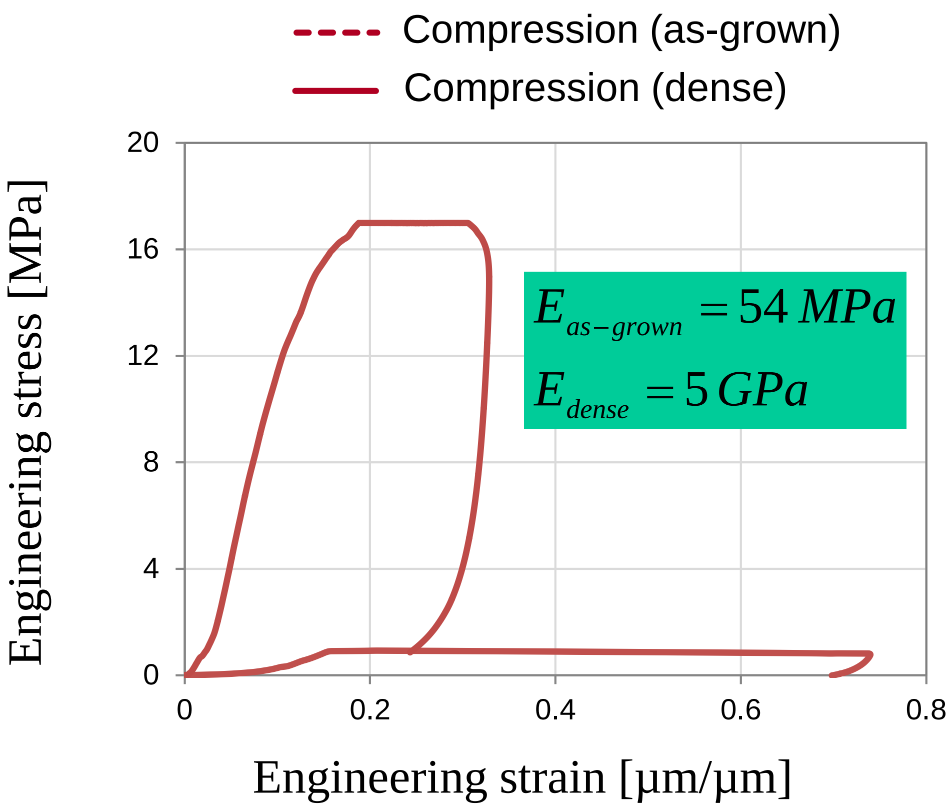
immersion





Liu et al. *IEEE Trans. Nanotech* 8(2):196-203, 2009.

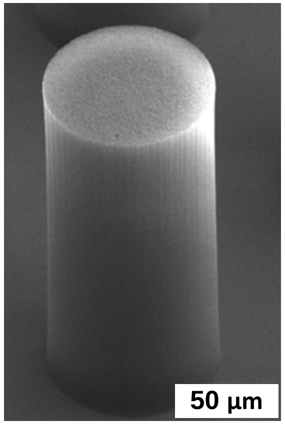
CNT forest as a (stronger) mechanical material

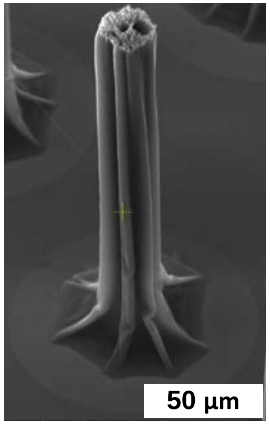


$$E_{as-grown} = 54\,MPa$$
$$E_{dense} = 5\,GPa$$

How is scales?

$$\frac{E}{E_d} = \left(\frac{\rho}{\rho_d}\right)^2 = \left(\frac{A}{A_d}\right)^2 = \left(\frac{R}{R_d}\right)^4 = 81$$





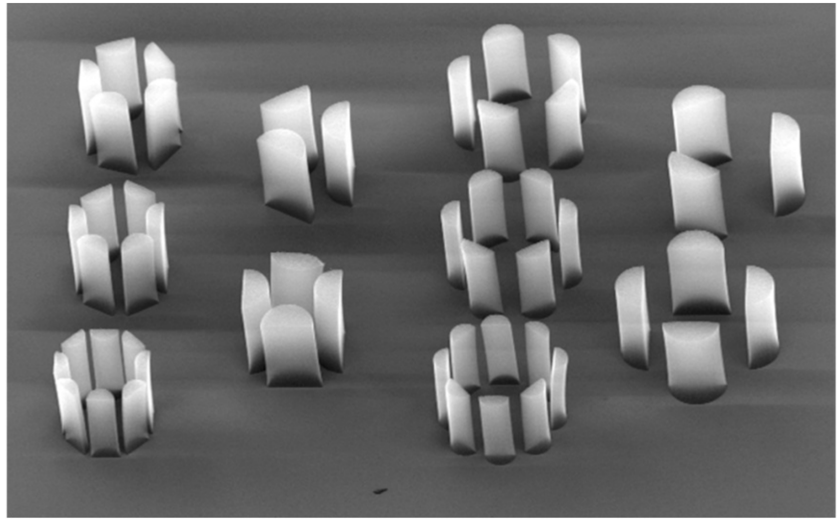
And by coating

$$E_{SU8} = 18\,GPa$$
$$E_{PMMA} = 25\,GPa$$

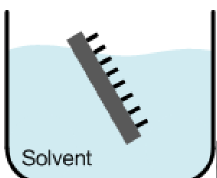
Process control: immersion vs. condensation

Condensation can be used to densify, shape and form very delicate nanostructures.

Forces encountered during “piercing” a liquid interface are avoided



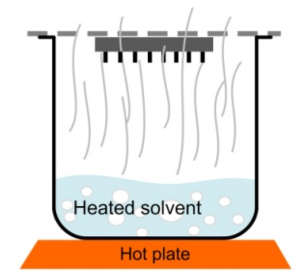
Immersion

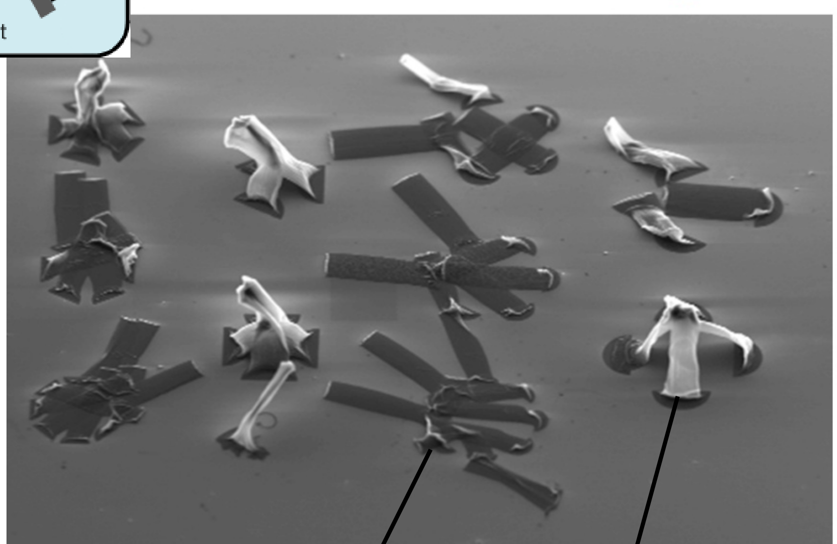


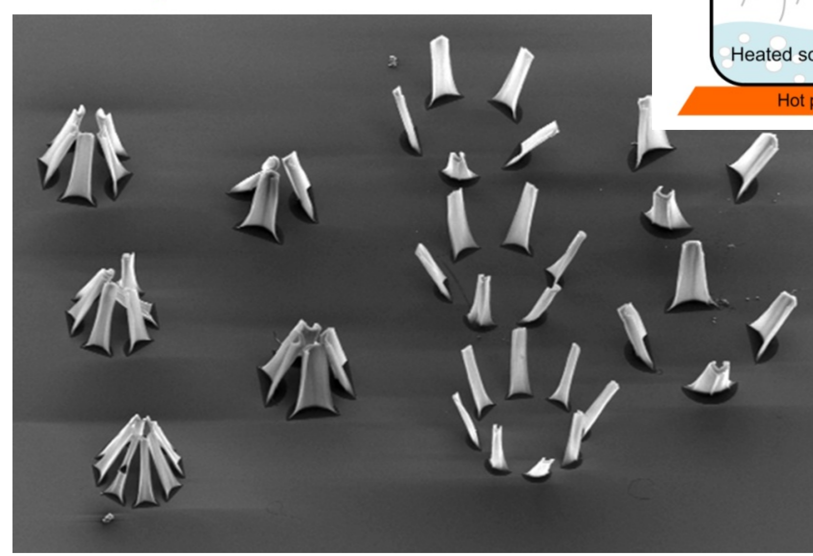
Immersion in acetone

Condensation of acetone

Condensation



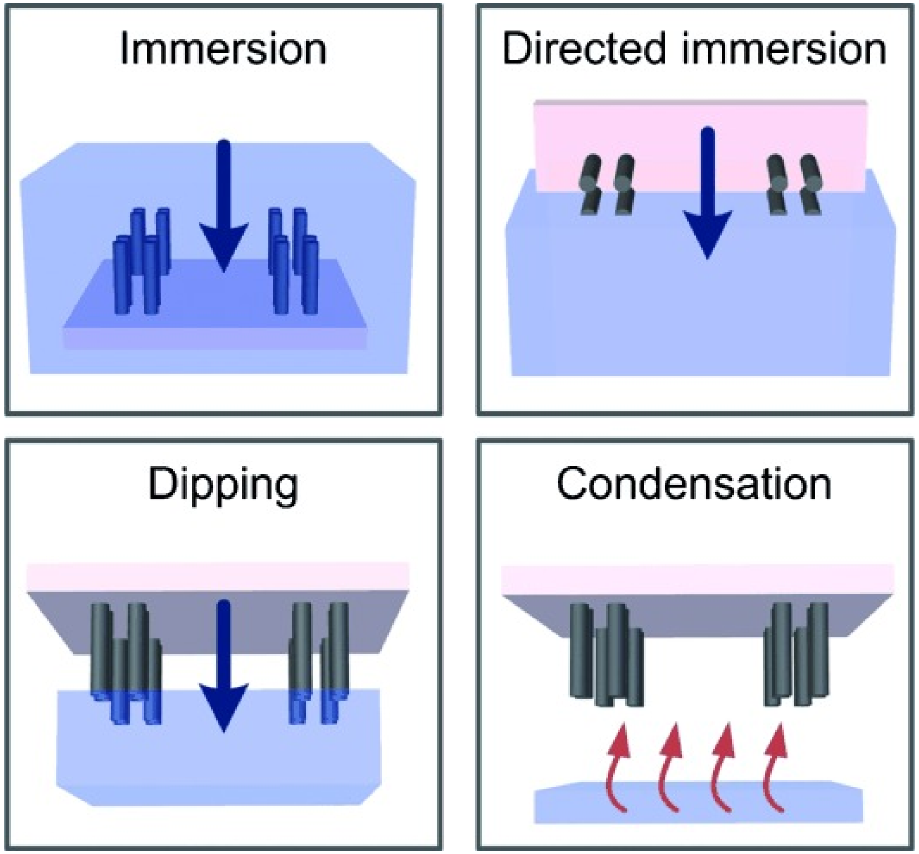




Collapse **Grouping**

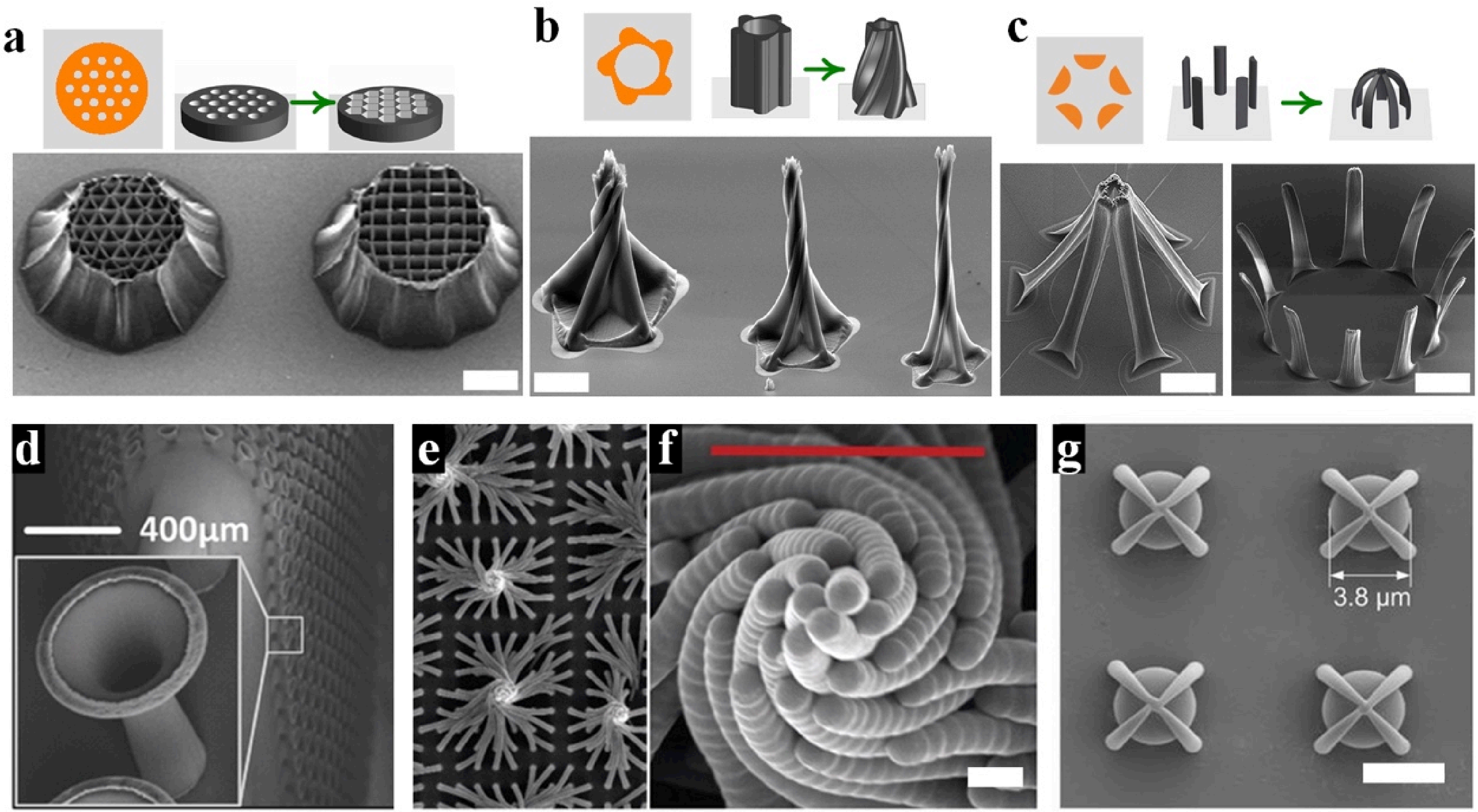
De Volder* and Tawfick* et al. *Advanced Materials*, 2010.

How to control elastocapillary self-assembly?



De Volder and Hart, Angewandte Chemie 2013

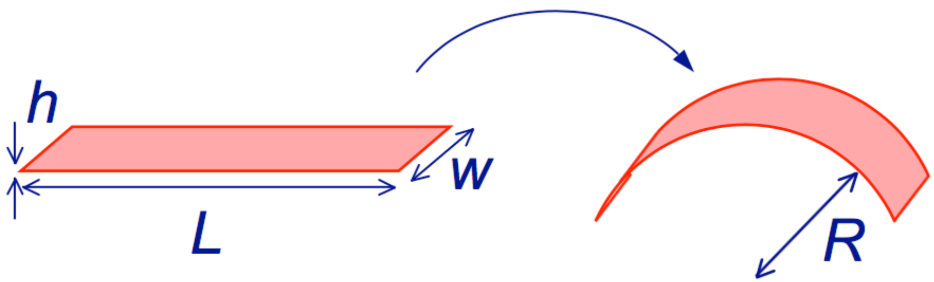
Capillary forming: Complex 3D nanostructures by elastocapillary self-assembly



Tawfick et al. *MRS Bulletin 2015.*

Elasticity *vs.* Capillarity

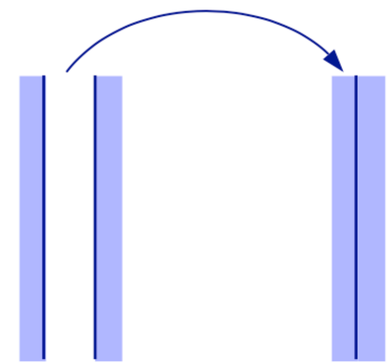
Bending energy



$$E_b \sim \frac{Eh^3}{R^2} wL$$

E Young modulus

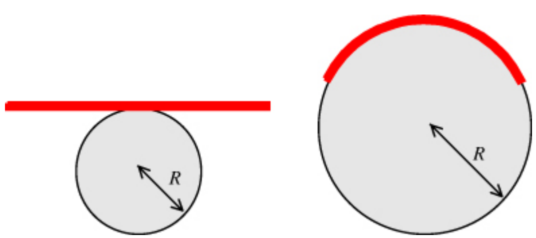
Surface energy



$$E_s \sim \gamma S \sim \gamma wL$$

$$E_b \sim E_s \leftrightarrow R \sim L_{EC} = \sqrt{\frac{Eh^3}{\gamma}}$$

Elastocapillary length



$$R > \sqrt{B / \gamma}$$

Capillary origami

Py et al. PRL 98, (2007)

https://www.youtube.com/watch?v=6gYb2fOnMvM

Capillary origami

Py et al. PRL 98, (2007)

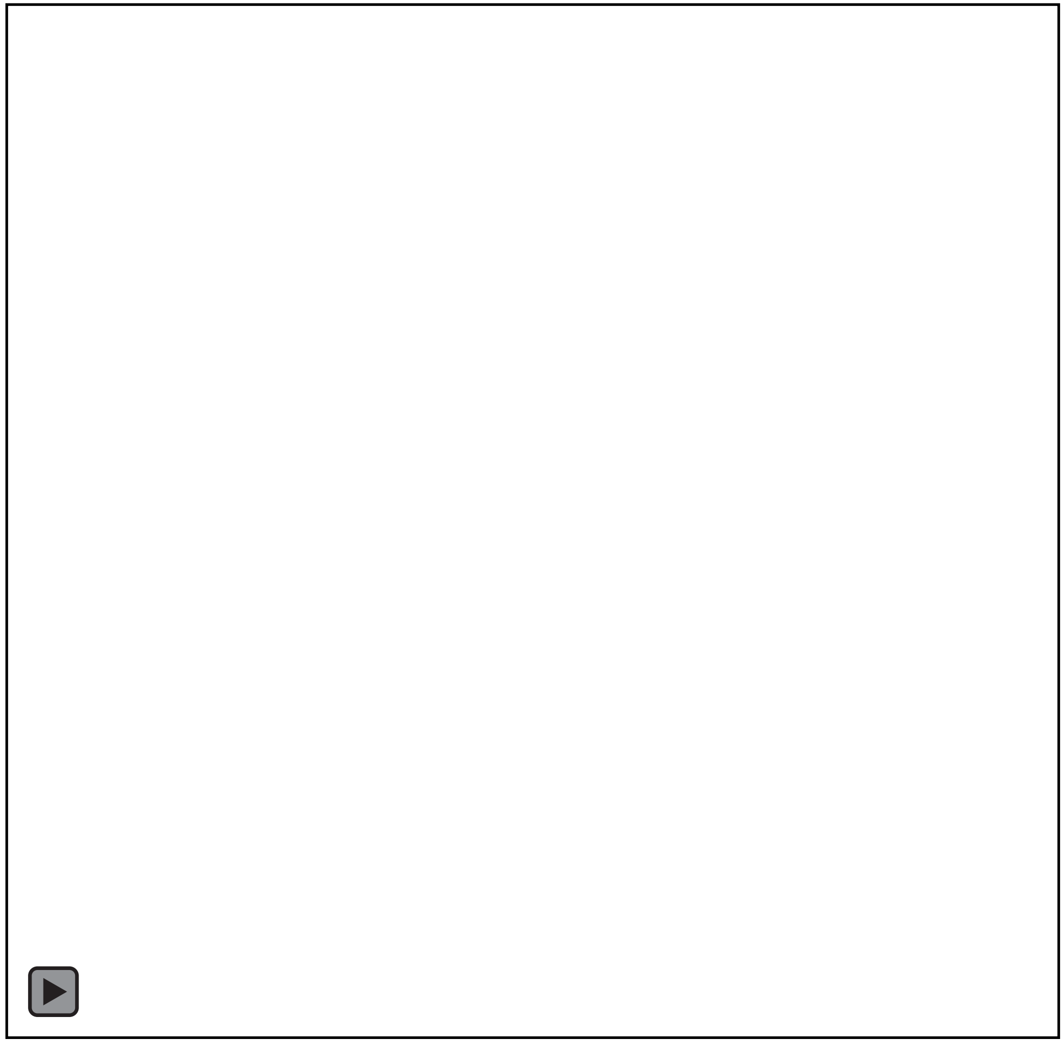
Grabbing an air bubble

https://www.dropbox.com/s/gpkig98vb87kxtk/reis_Supplementary_Movie_S1.mov?dl=0

Reis et al. Soft Matter (2010) https://pubs.rsc.org/en/content/articlelanding/2010/sm/c0sm00895h#!divAbstract

Grabbing a liquid droplet

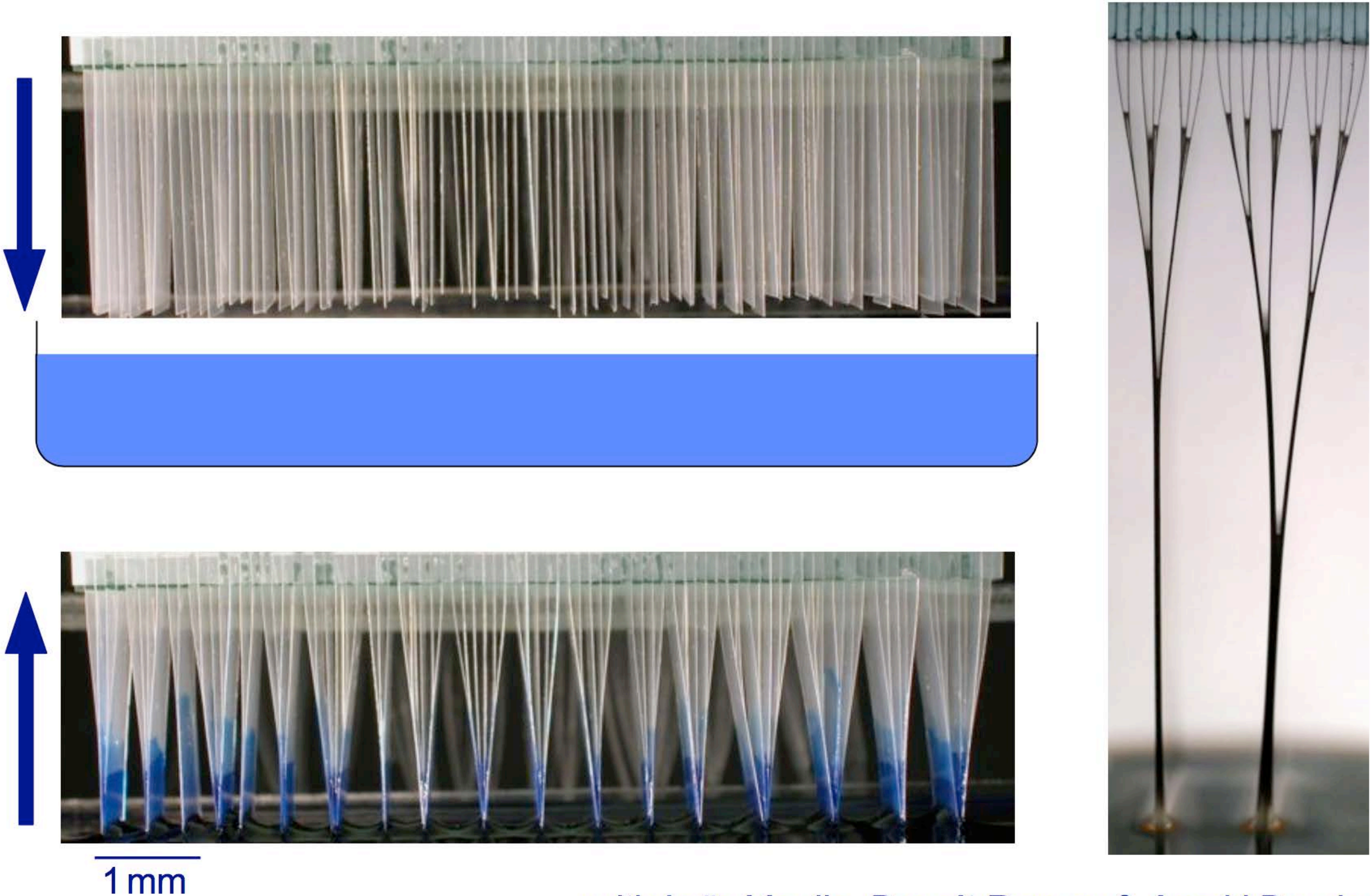
https://www.dropbox.com/s/yy3jyjxhtgmkzmb/reis_Supplementary_Movie_S2.mov?dl=0



Reis et al. Soft Matter (2010) https://pubs.rsc.org/en/content/articlelanding/2010/sm/c0sm00895h#!divAbstract

Slide by J. Bico

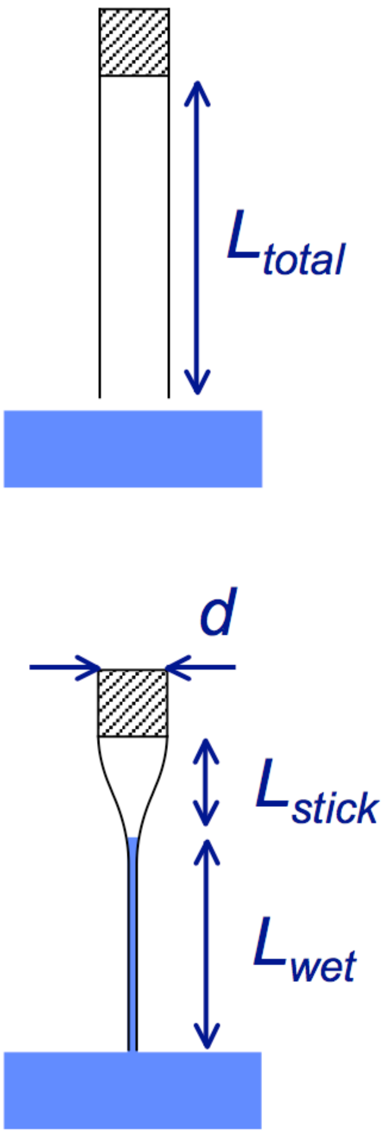
Wet hairs

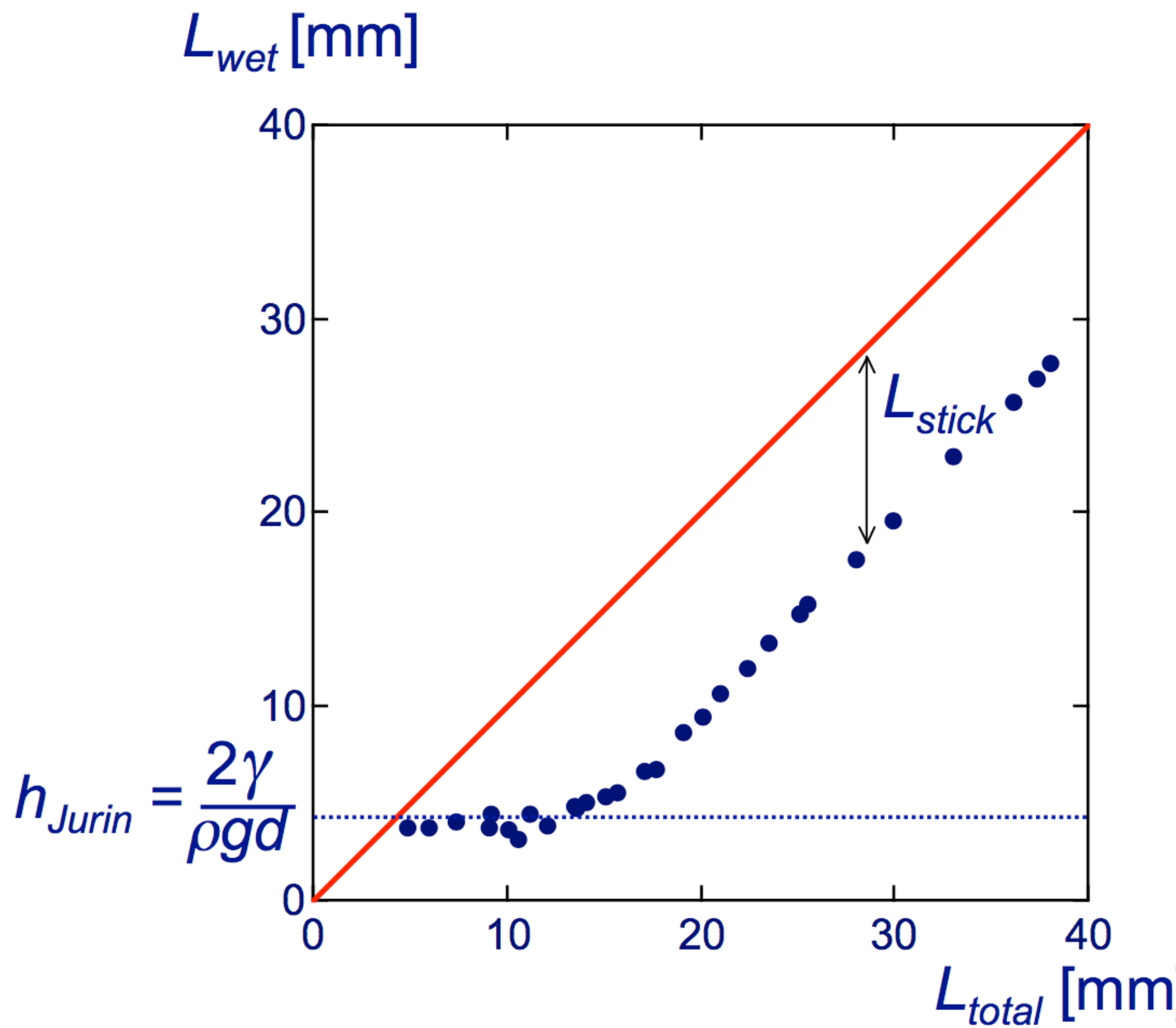


with Loïc Moulin, Benoît Roman & Arezki Boudaoud
Nature 2004

Slide by J. Bico

2 flexible lamellae





transparency, d = 1 mm, h = 100 μm
silicon oil

more detailed: Kim & Mahadevan JFM 2006

Sticking length ↔ Fracture

Griffith's criterion (1921):

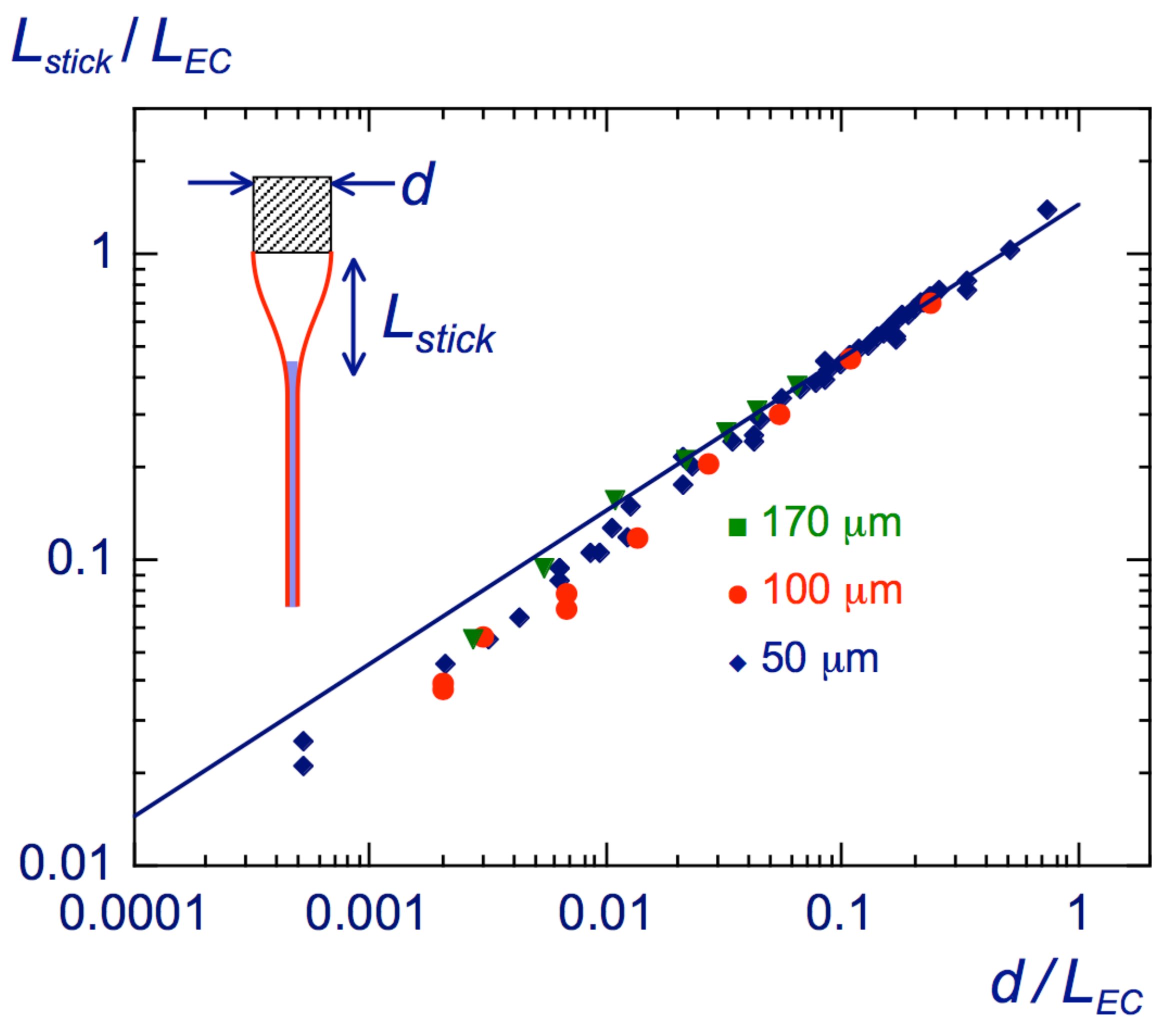
$$\gamma L_{stick} \sim E h^3 \left(\frac{d}{L_{stick}^2} \right)^2 L_{stick}$$

$$L_{stick} \sim \left(\frac{Eh^3}{\gamma} \right)^{1/4} d^{1/2} \sim L_{EC}^{1/2} d^{1/2}$$

Elastocapillary length

$$L_{EC} = \sqrt{\frac{Eh^3}{\gamma}}$$

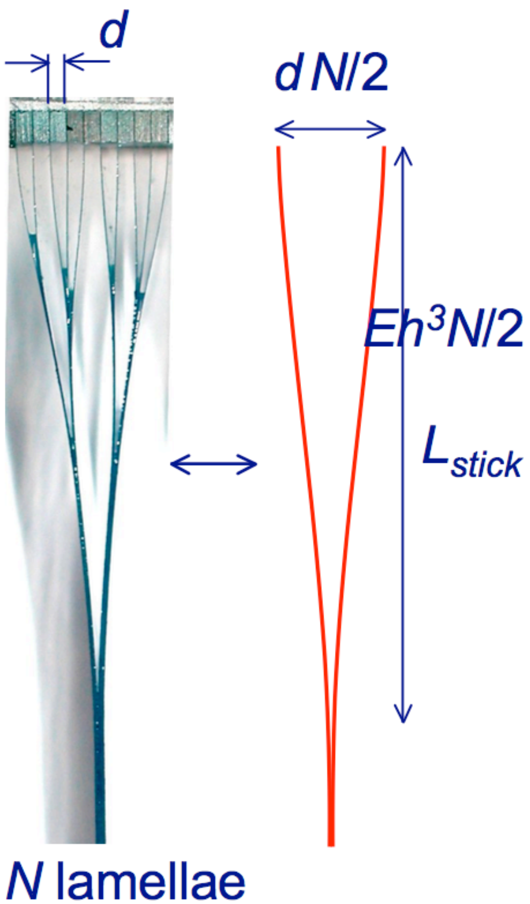
also Mastrangelo 1992

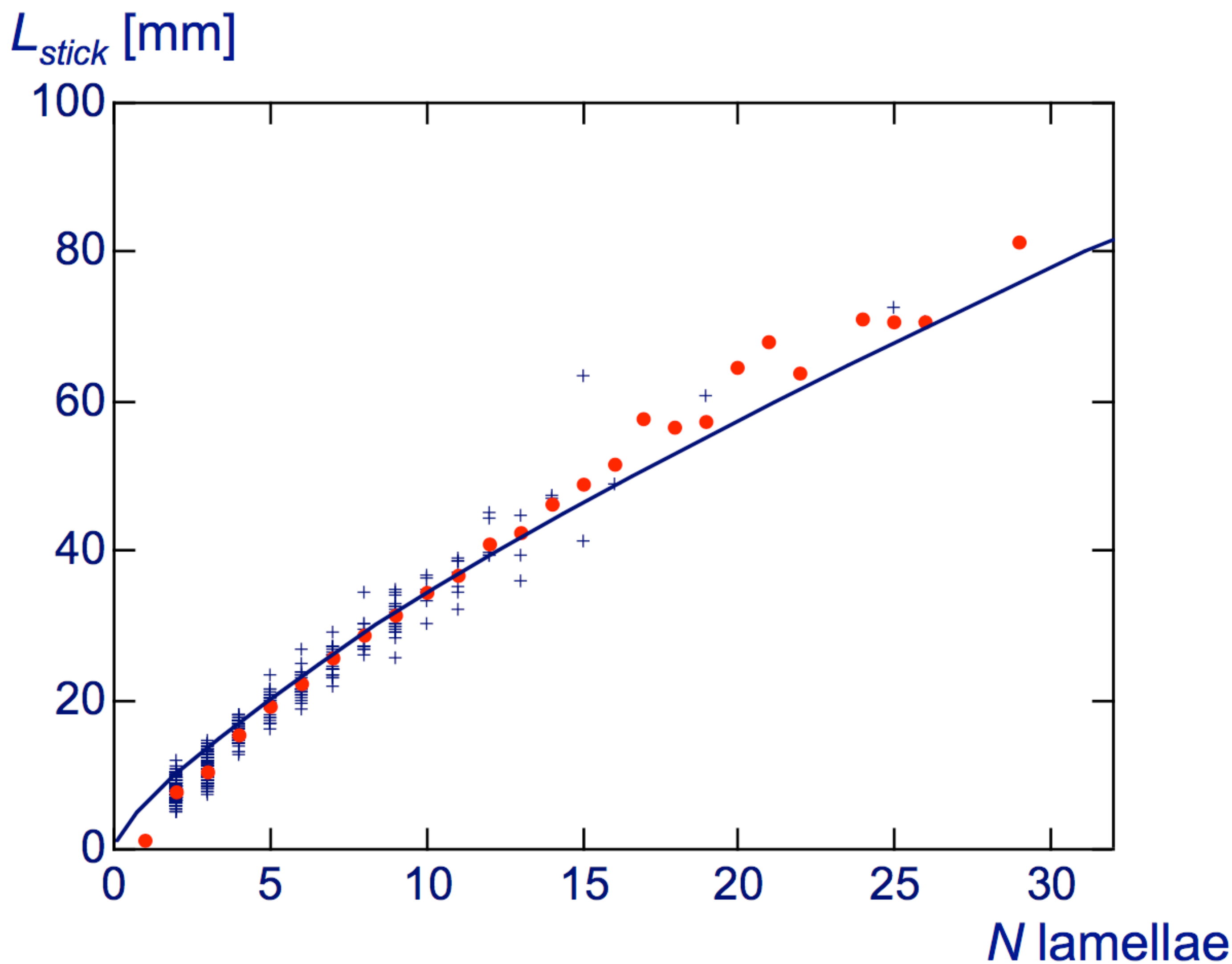


Slide by J. Bico

Multilamellae

$$L_{stick} \sim N^{3/4} L_2$$



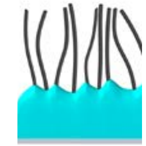


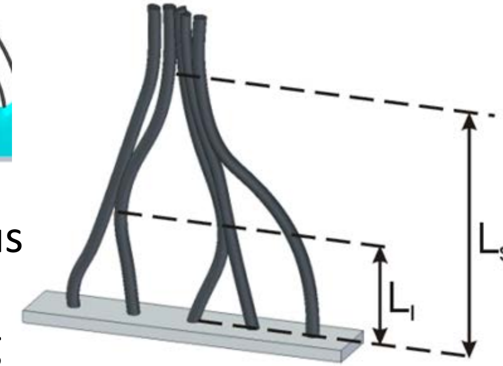
Size Limits For Void-Free Structures

$$L_I = C_1\sqrt{d\sqrt{\frac{E\pi R^3}{4\gamma}}}$$

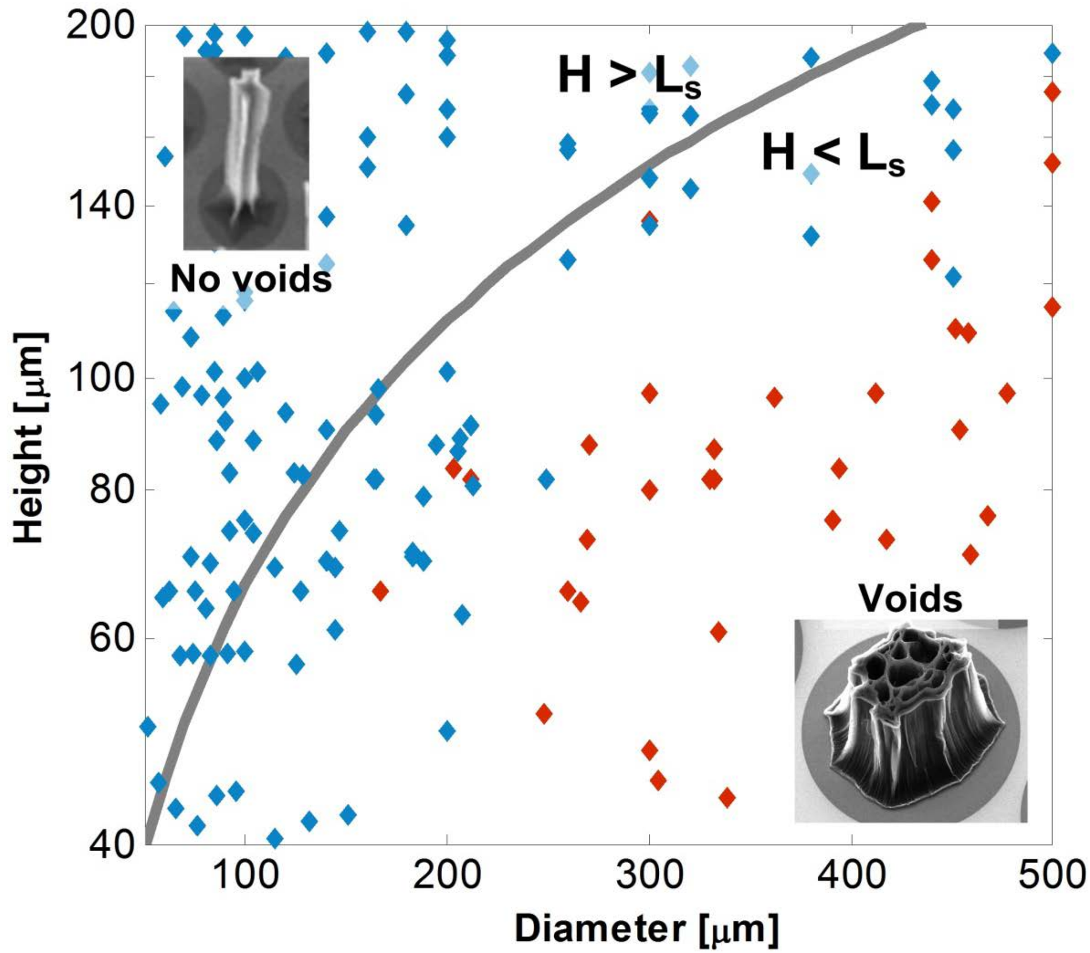
$$L_S = C_2 L_I N^{3/8}\sqrt{\beta}$$

Py et al., *EPL 77(4):44005, 2007.*



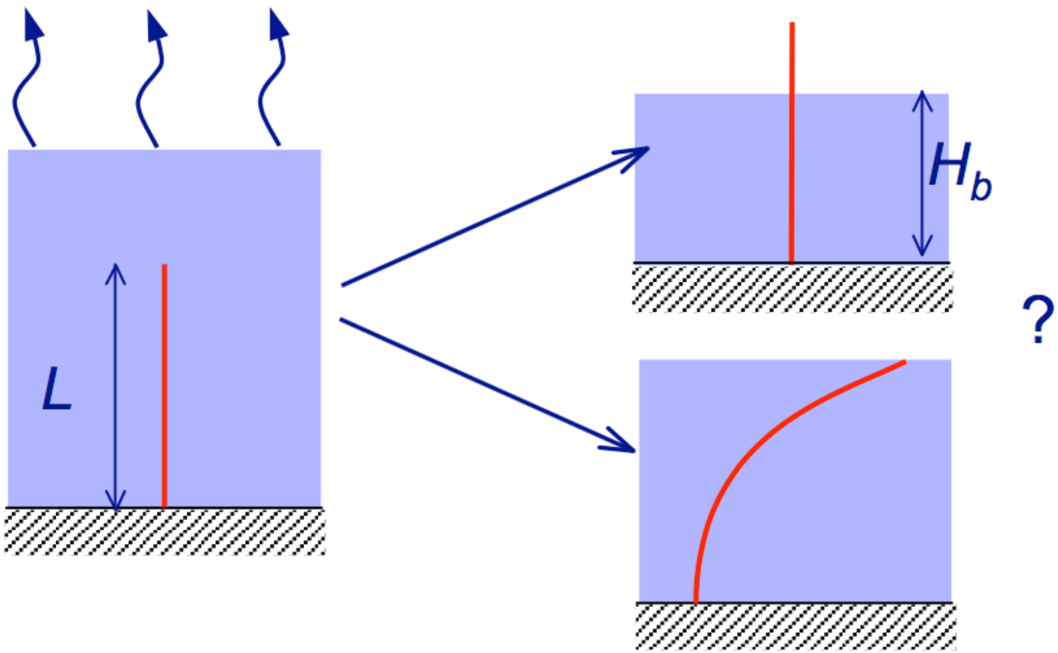


E = modulus
R = radius
d = spacing
γ = surface tension
N = number of filaments in the cluster

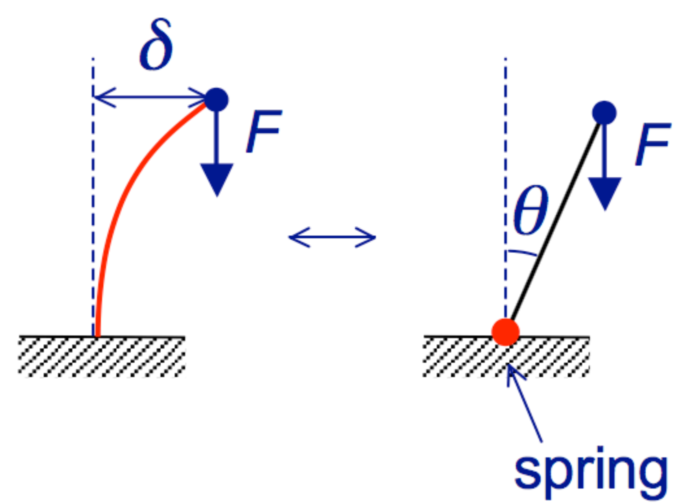


Bico J. *Nature* **432**, 690 (2004)

De Volder M., Tawfick S. et al., J. *Micromech Microeng.*, (2011)



Euler buckling



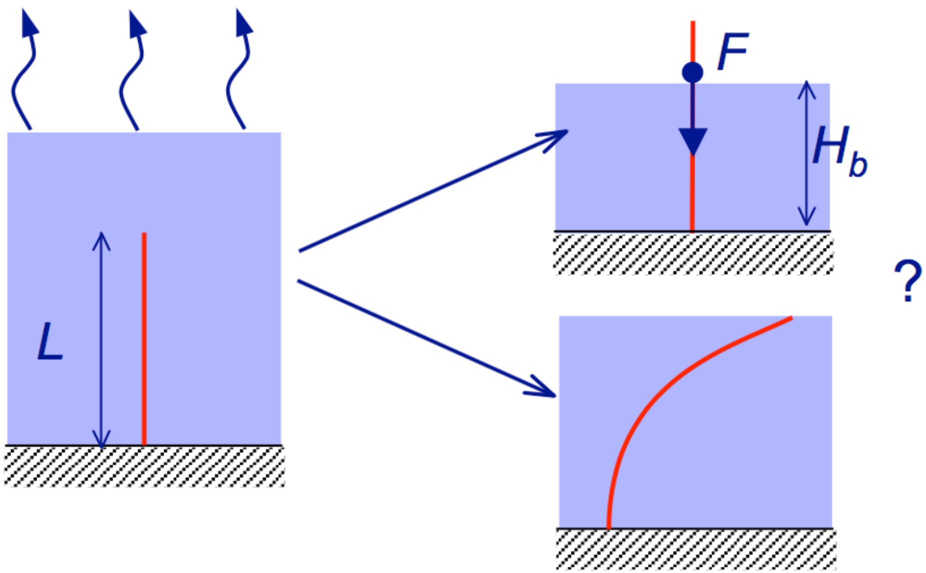
$$\delta/L \longleftrightarrow \theta$$

$$M = B\kappa \sim B\,\delta/L^2 \sim F L\,\delta/L$$

$\Rightarrow$ critical force: $F \sim B/L^2$

full calculation: $\boxed{F_{crit} = \dfrac{\pi^2}{4}\,\dfrac{B}{L^2}}$

Piercing hairs



capillary force

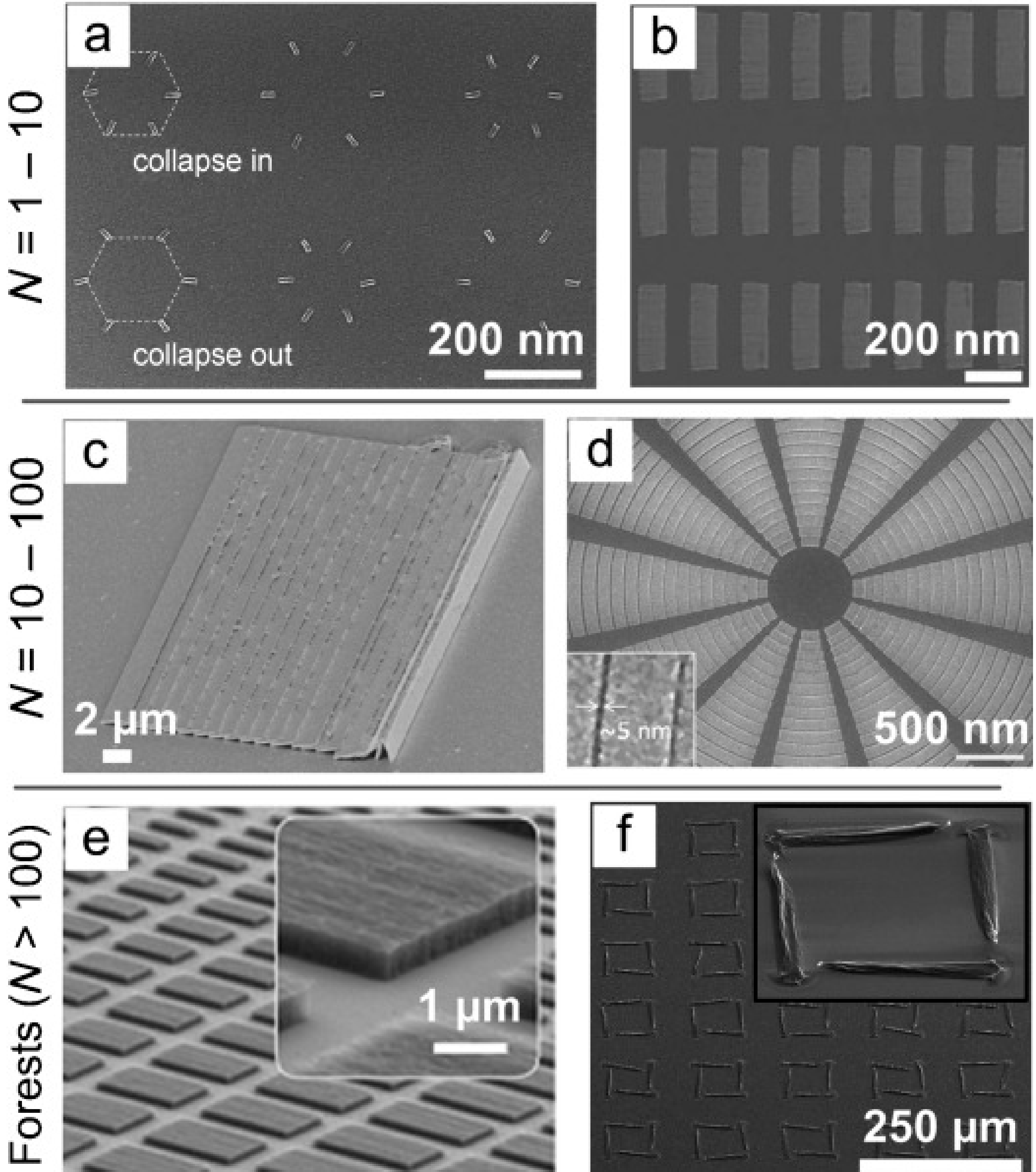
$$F = 2\pi b\gamma$$

$$L_{crit} = \frac{\pi}{2}\sqrt{\frac{B}{F}} \sim \sqrt{\frac{Eb^3}{\gamma}} \sim L_{EC}$$

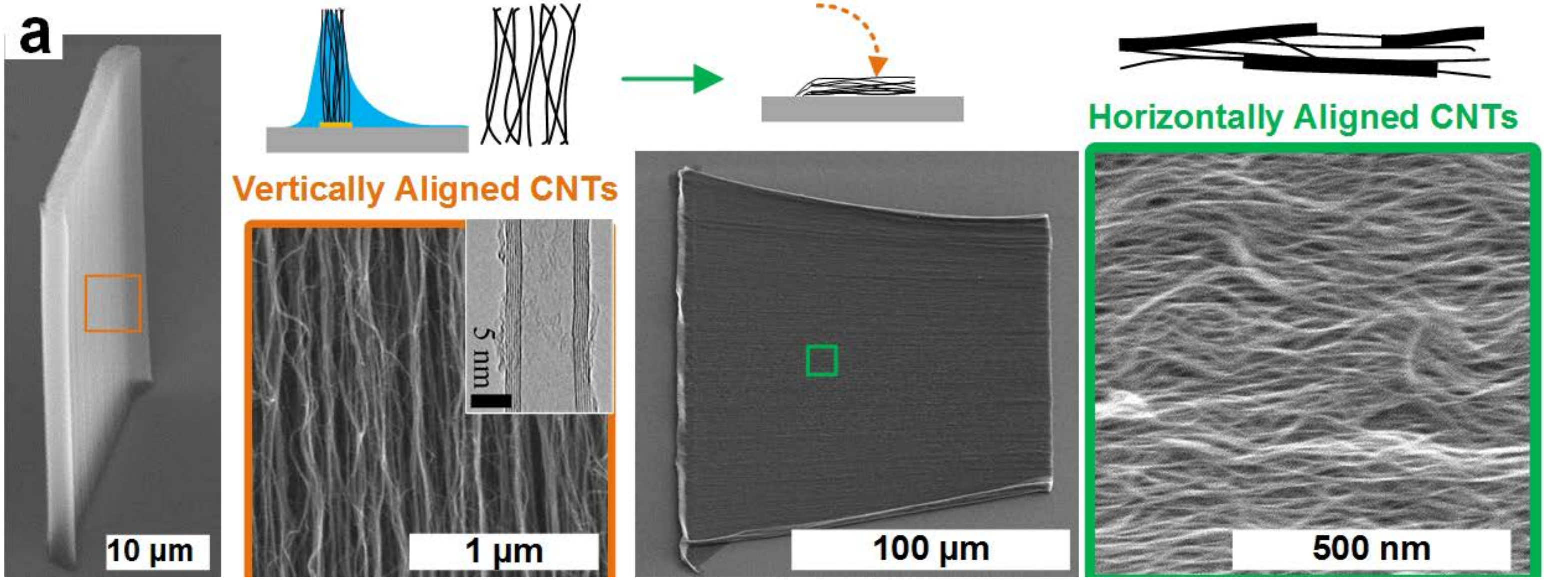
post-buckling: complex!

with Benoît Roman & Sébastien Neukirch

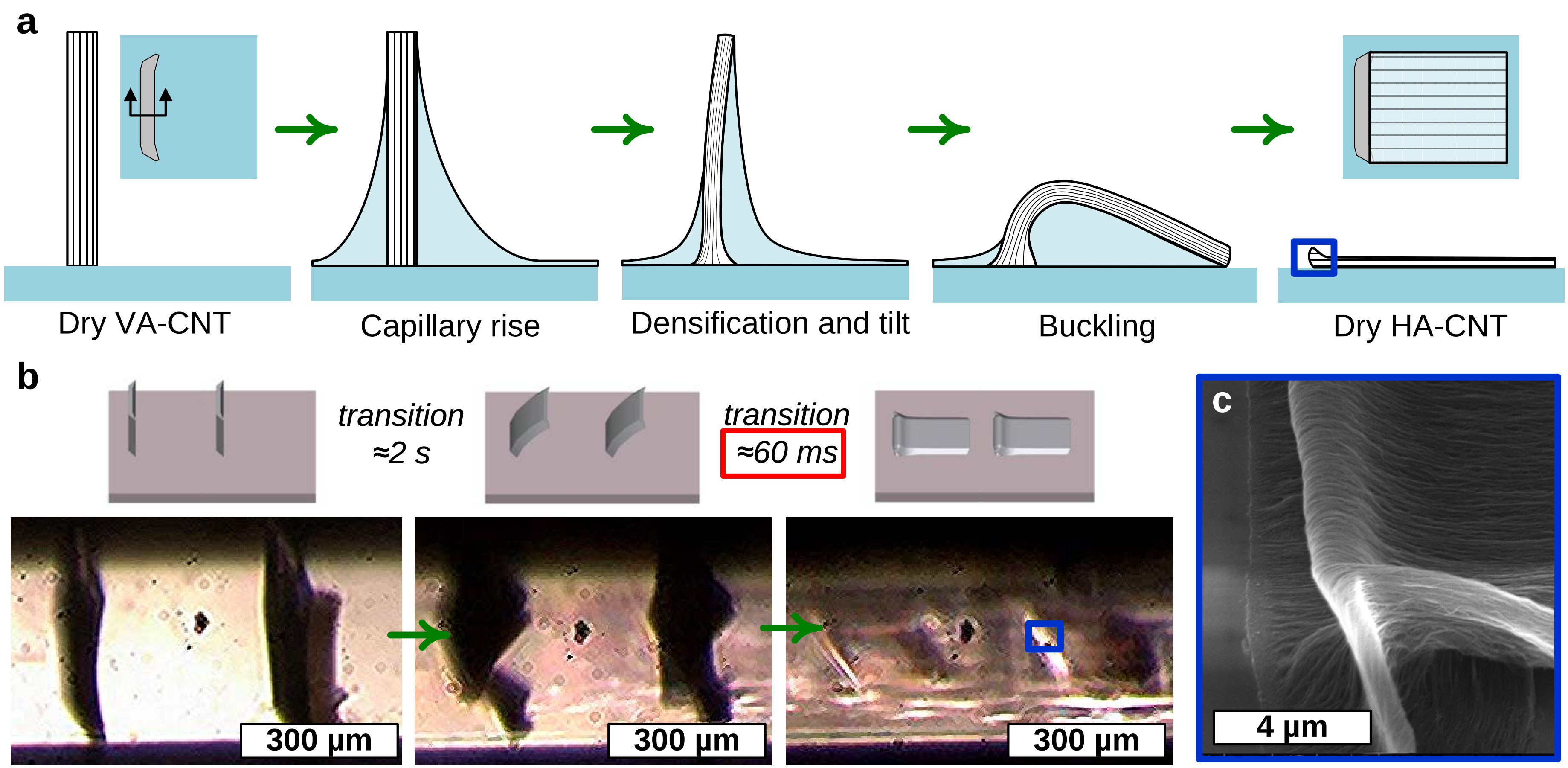
JMPS 2007


N = 1 – 10
a
collapse in
collapse out
200 nm
b
200 nm
N = 10 – 100
c
2 µm
d
~5 nm
500 nm
Forests (N > 100)
e
1 µm
f
250 µm

Capillary buckling of CNTs



Capillary buckling dynamics

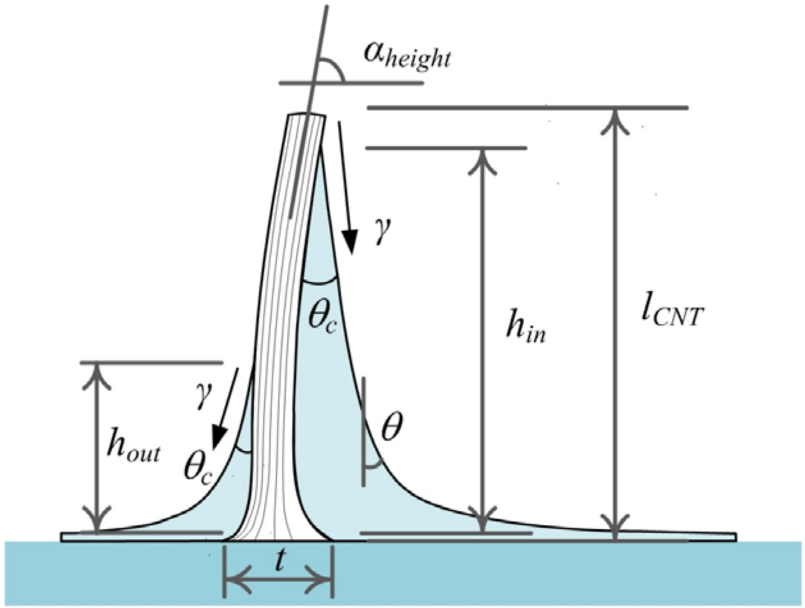


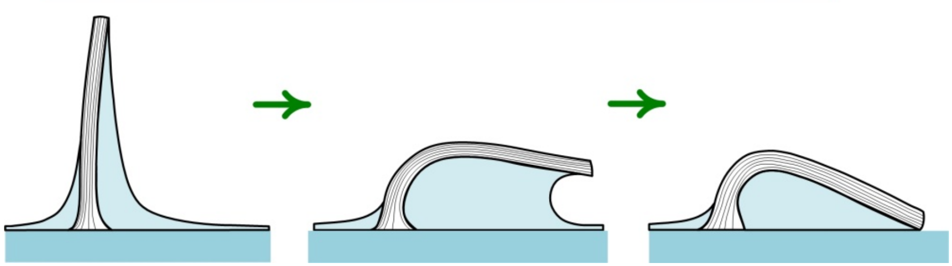
Tawfick S et al, *Langmuir* ***27*** (10) (2011)

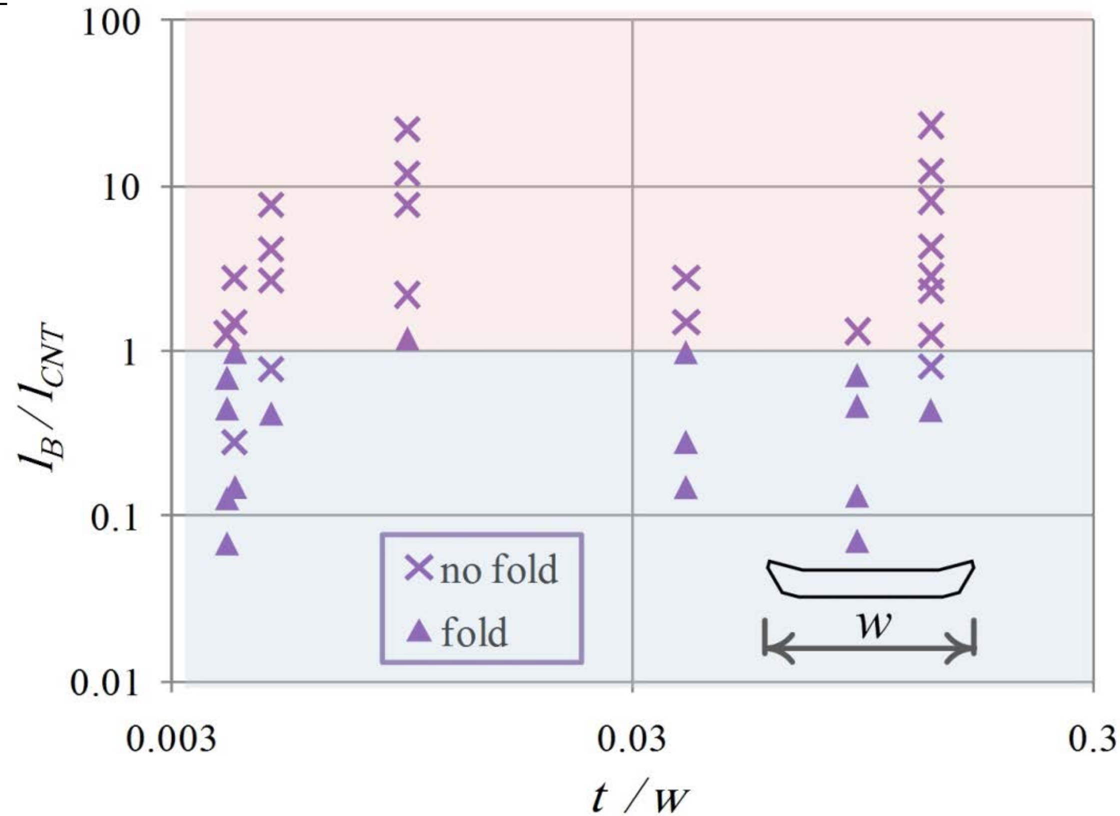
Design for Stable Structures

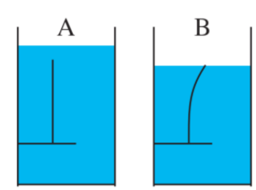
- Elastocapillary buckling limit

$$l_B = \sqrt{\left(\frac{E}{\gamma}\right)\frac{I}{w}} \qquad F_{Buckling} = \left(\frac{\pi}{2}\right)^2 \frac{EI}{l_{CNT}^2}$$





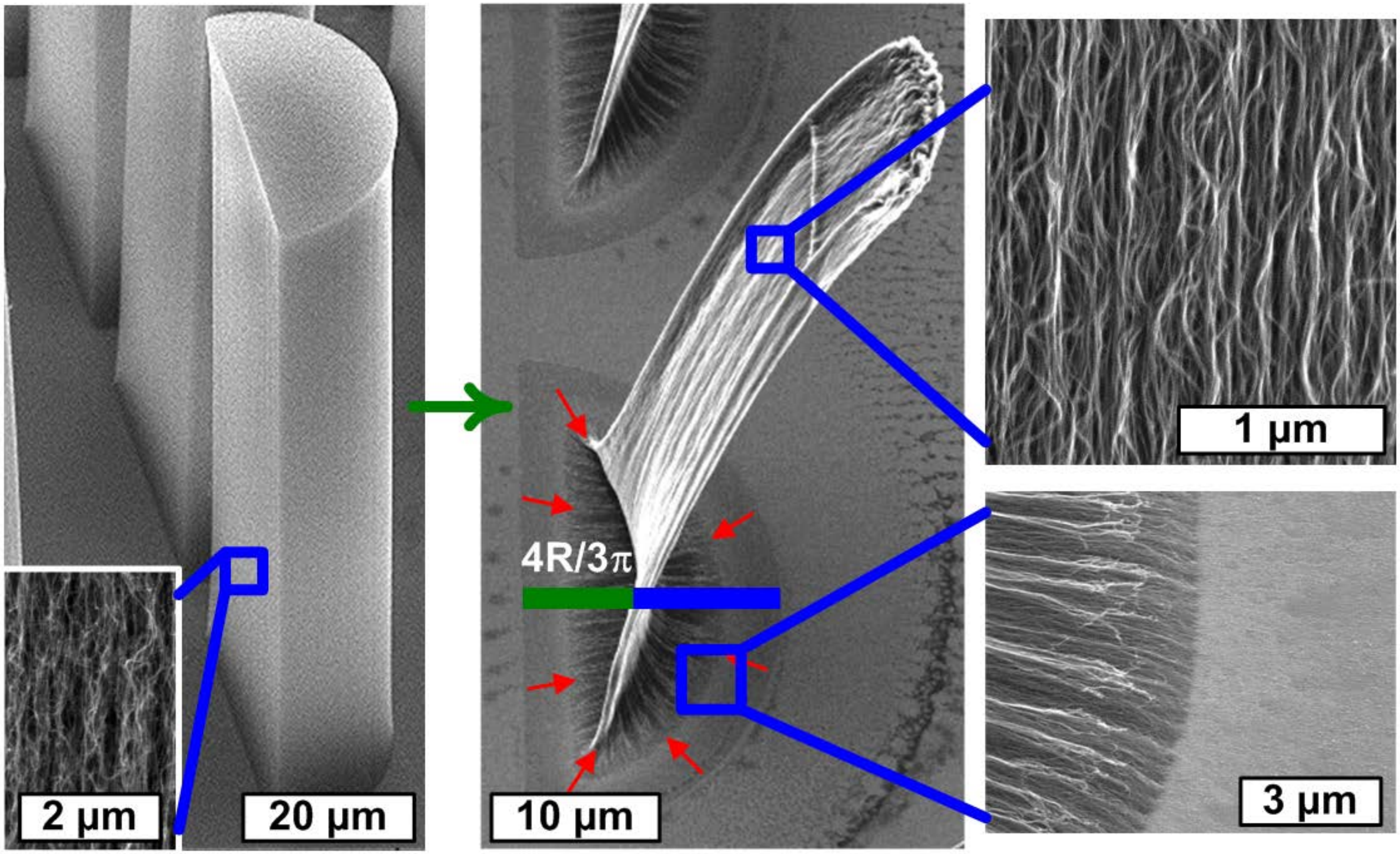




Neukirch S, *Journal of the Mechanics and Physics of Solids* 55 (6) 1212-1235 2007.

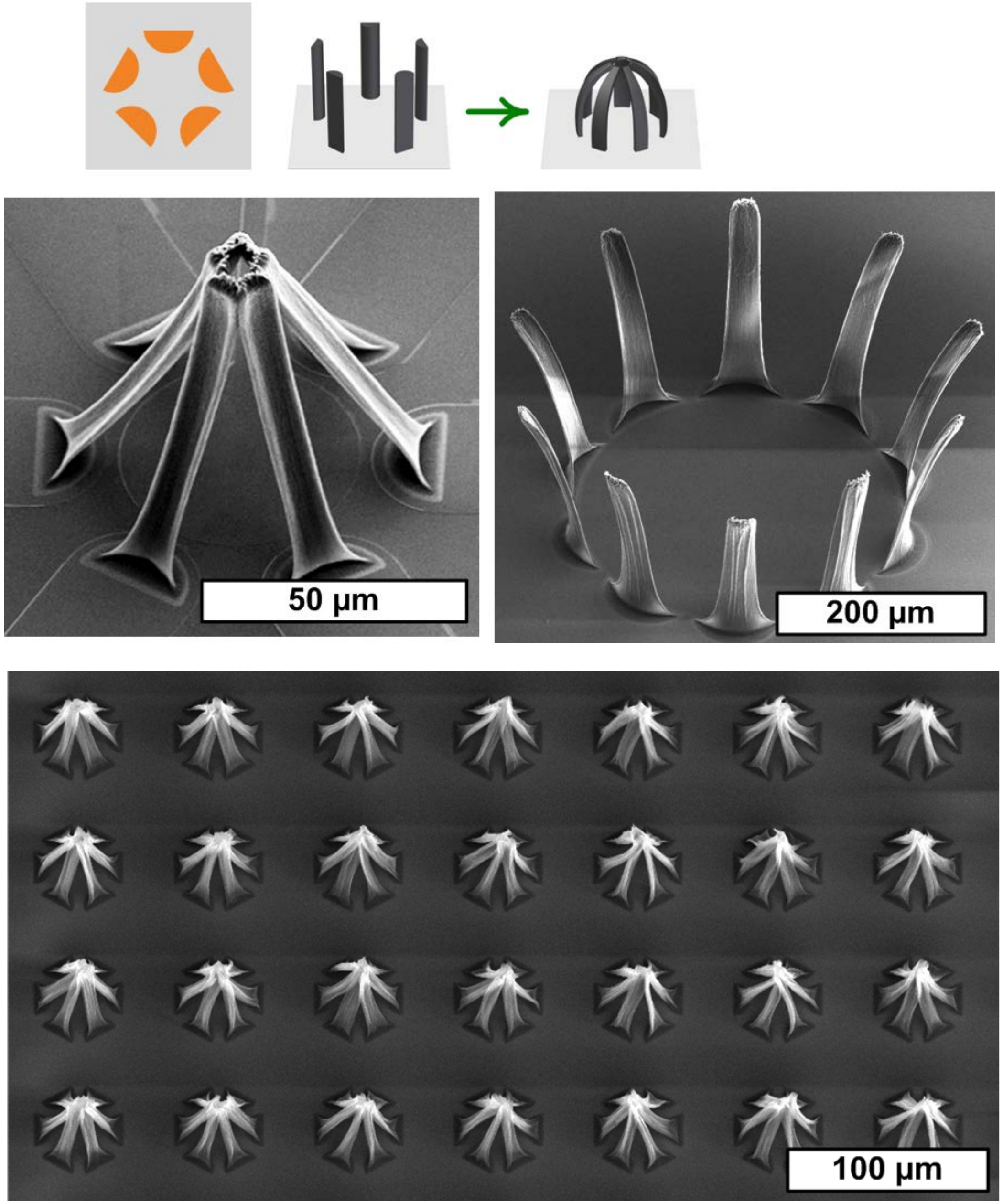
Tawfick et al, *Langmuir* ***27*** (10), 6389–6394 2011.

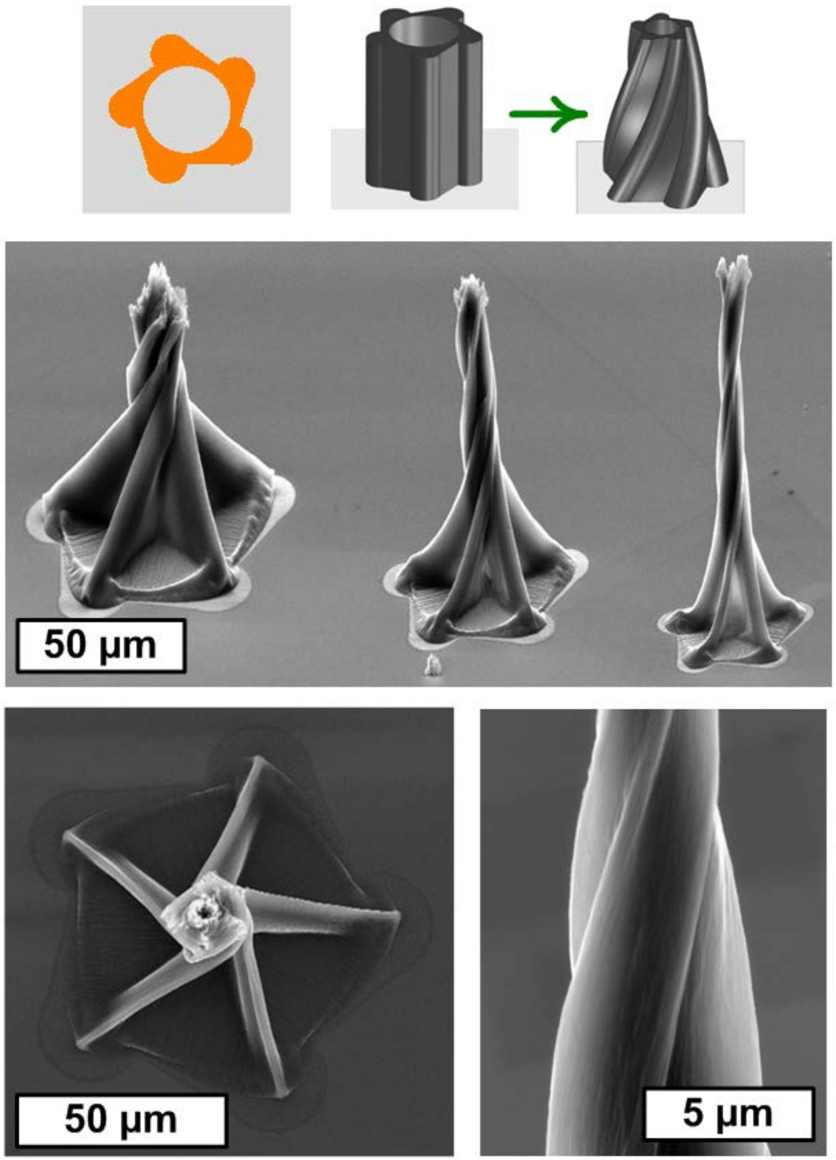
Capillary bending



Devolder *and Tawfick* et al. *Advanced Materials*, 2010.
Zhao, and Tawfick et al. *Physical Review E*, 2010.
Tawfick and Hart et al. *Nanoscale* 2012.

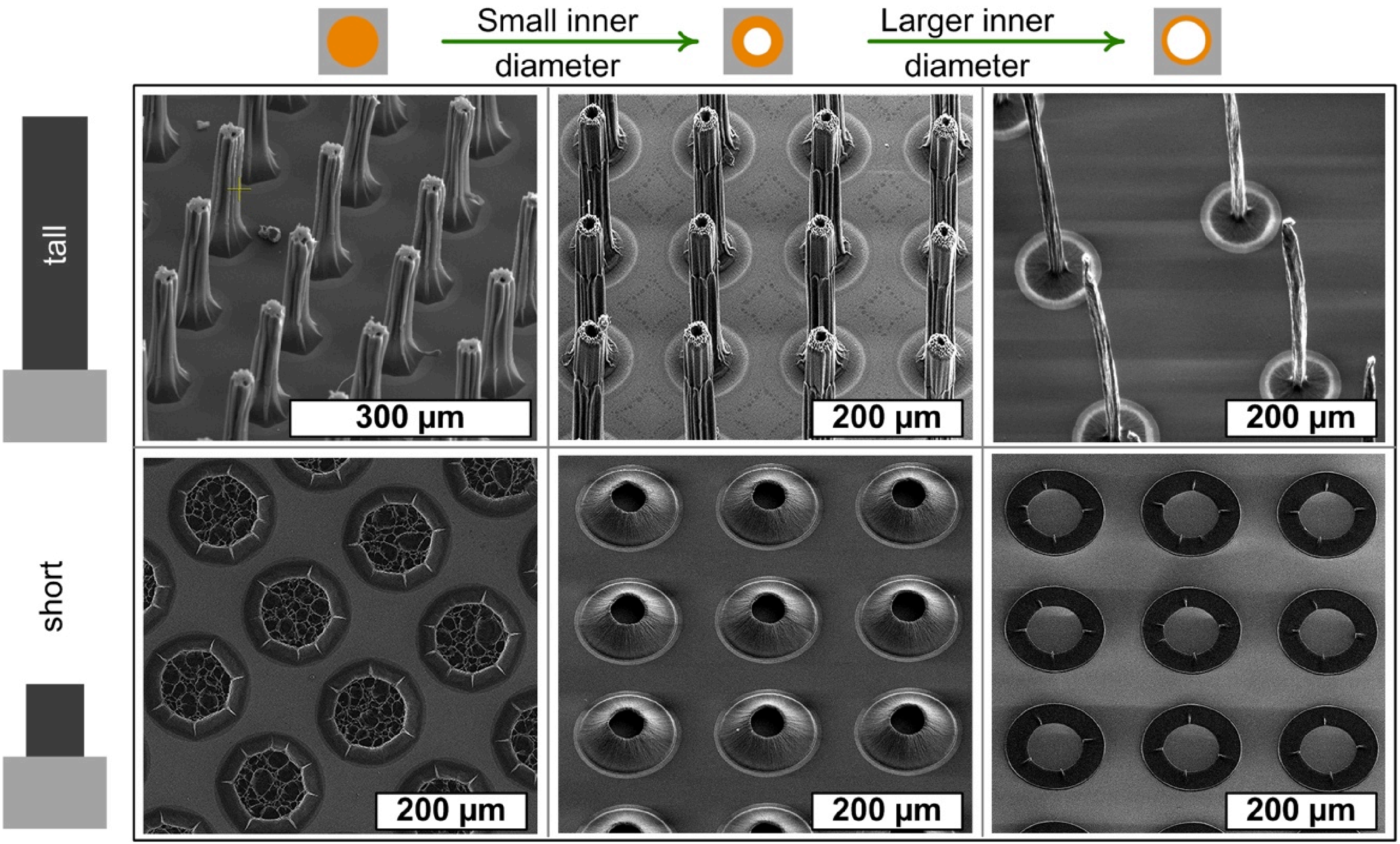
Multi-directional 3D architectures



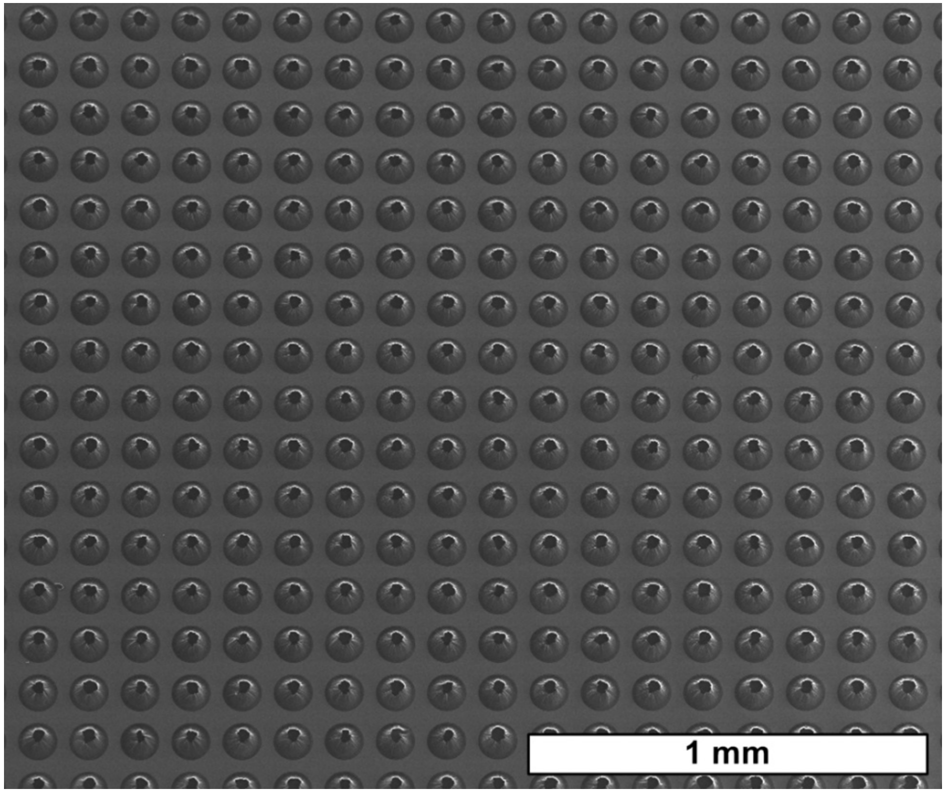


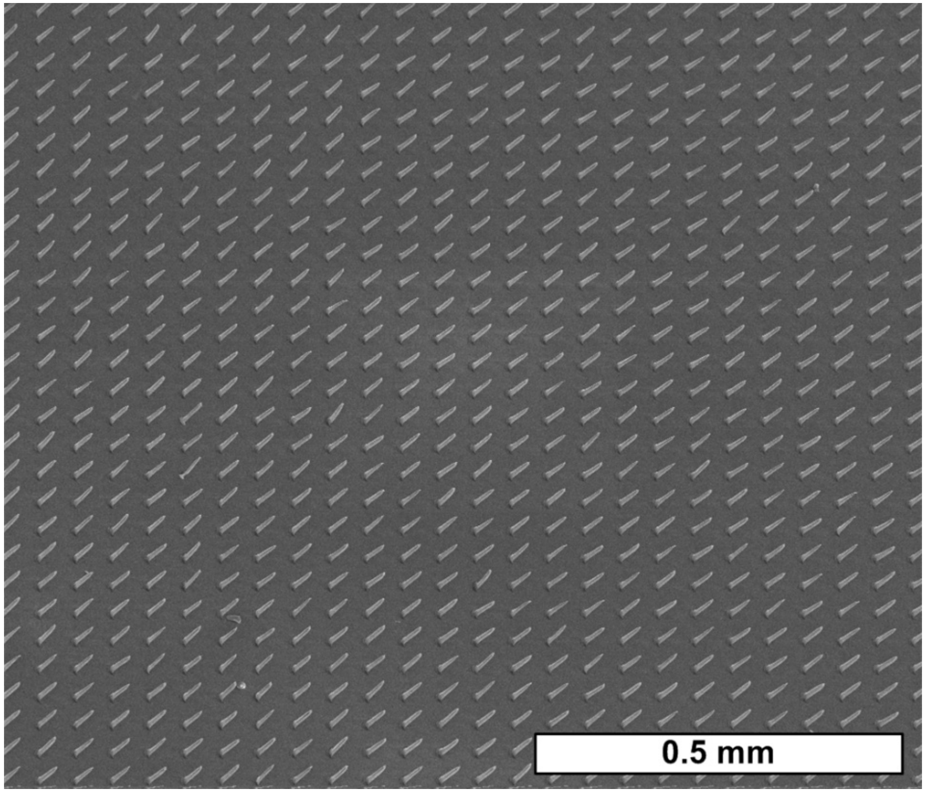
Devolder, Tawfick, et al. *Advanced Materials*, 2010.

Structural analysis of capillary forming transformations

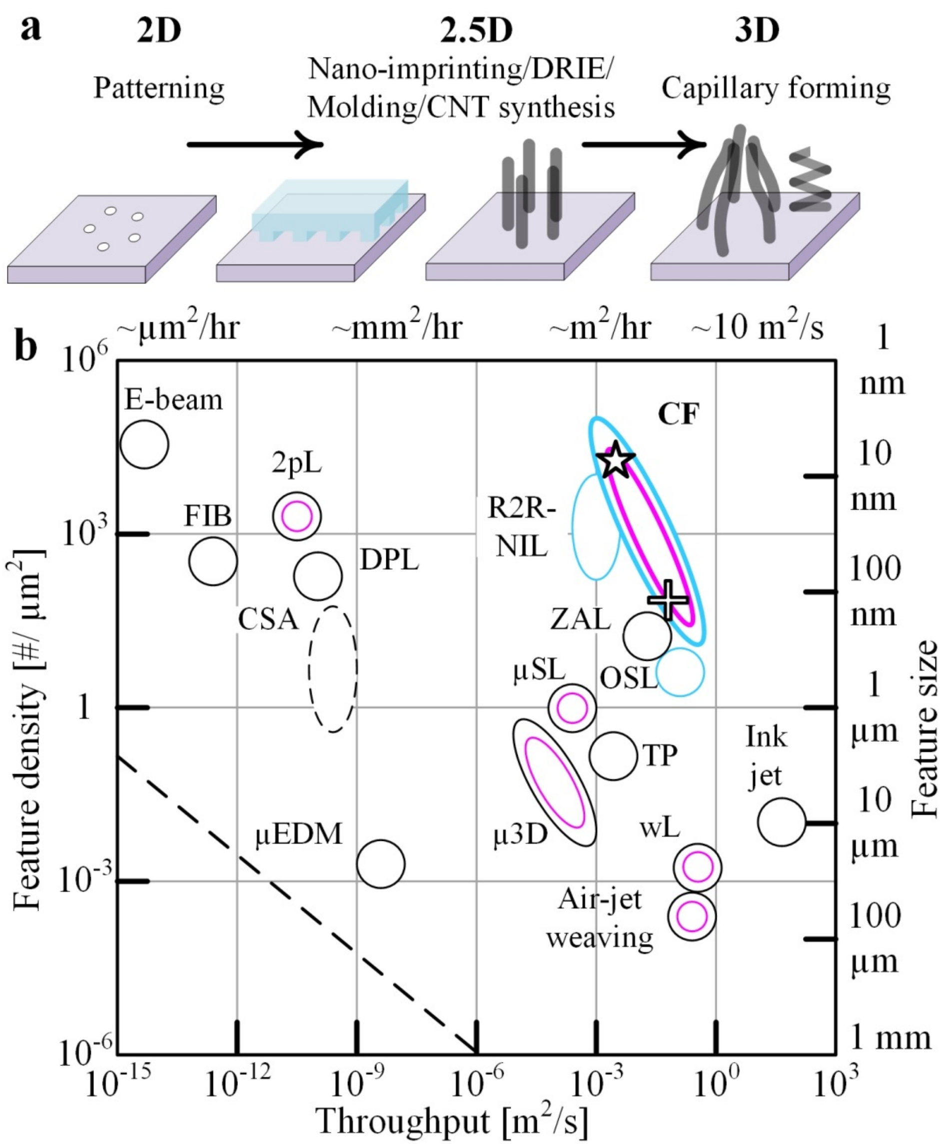


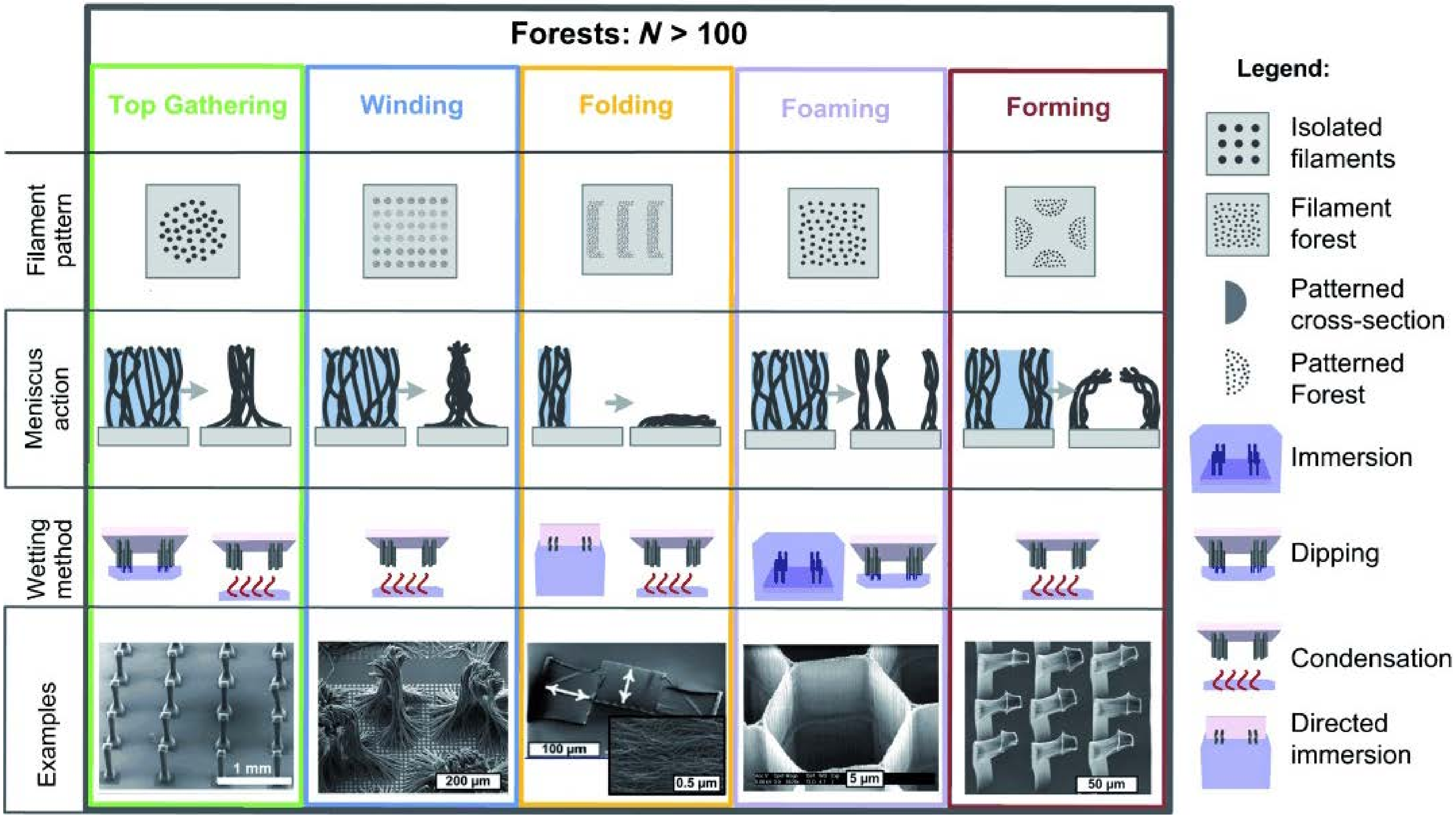
Large arrays of 3D hierarchical nanostructures





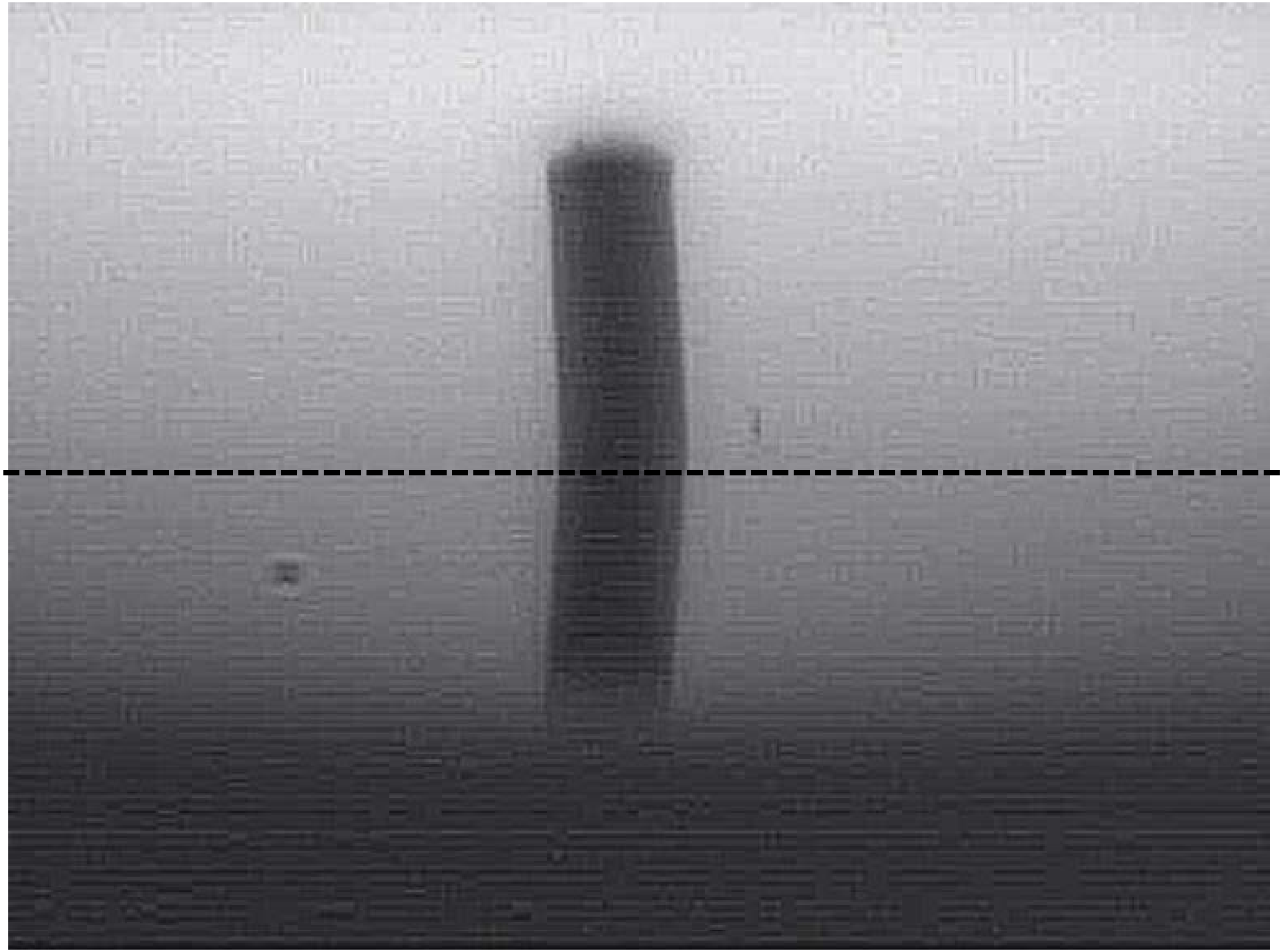
A manufacturing process




Forests: N > 100
Top Gathering
Winding
Folding
Foaming
Forming
Filament pattern
Meniscus action
Wetting method
Examples
1 mm
200 µm
100 µm
0.5 µm
5 µm
50 µm
Legend:
Isolated filaments
Filament forest
Patterned cross-section
Patterned Forest
Immersion
Dipping
Condensation
Directed immersion

Observing capillary forming

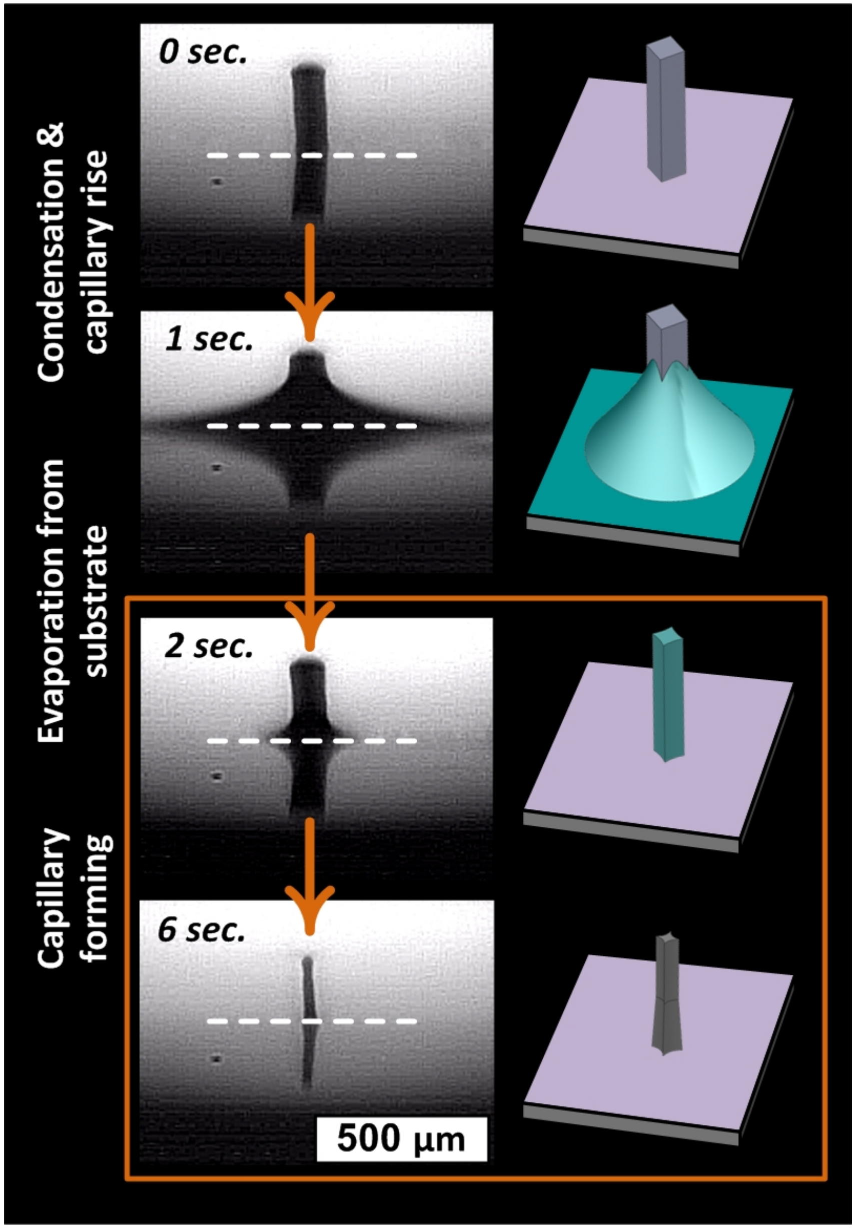
https://pubs.acs.org/doi/suppl/10.1021/la4002219



Tawfick et al., Langmuir 2013

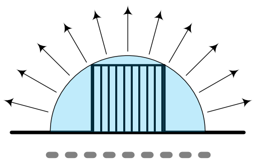
Observing capillary forming

■

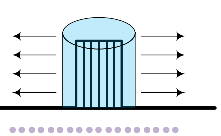


Tawfick et al., Langmuir 2013

Shrinkage during liquid evaporation

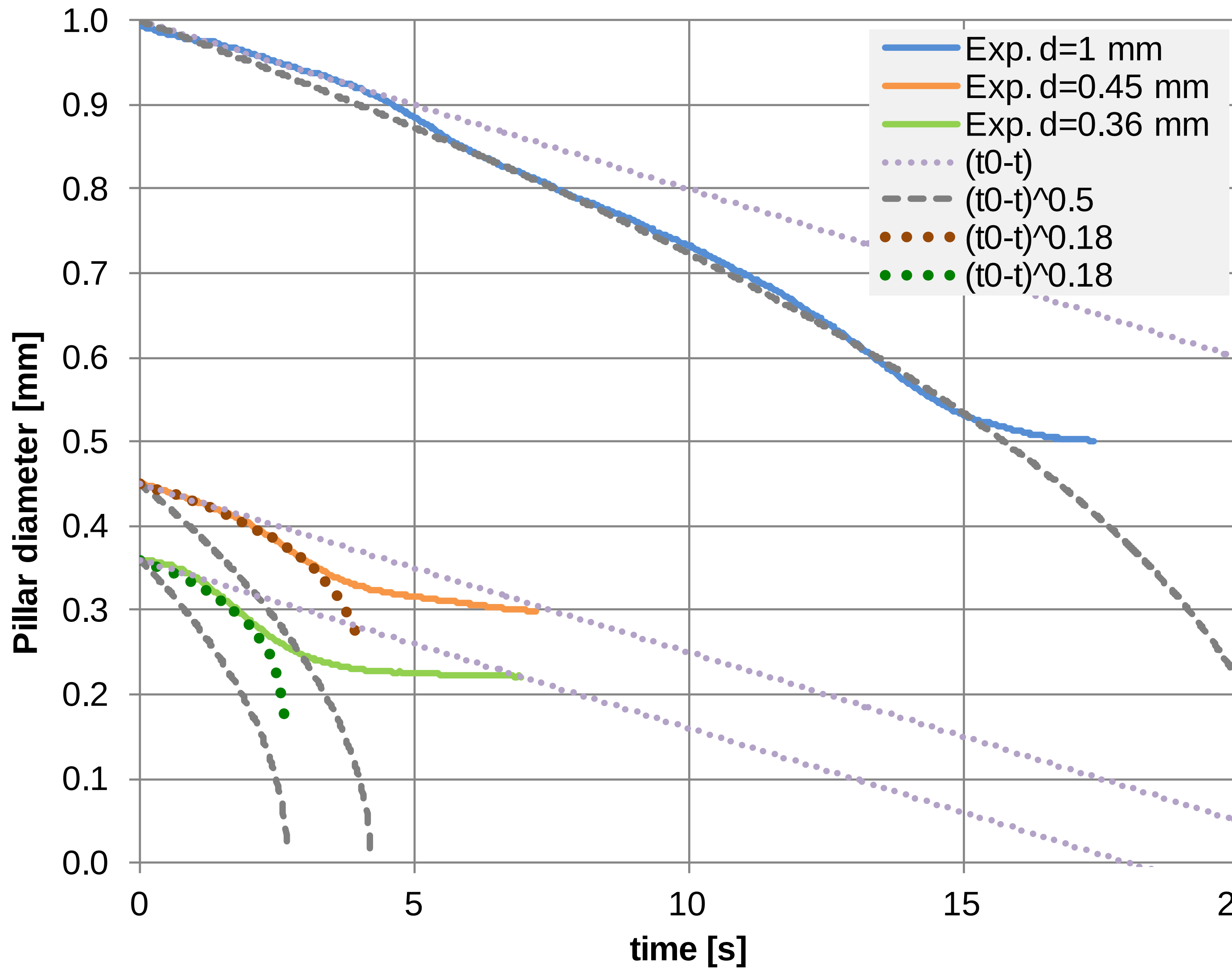


$$d(t) = \alpha\sqrt{t_0 - t}$$



$$d(t) = \alpha(t_0 - t)^{0.18}$$

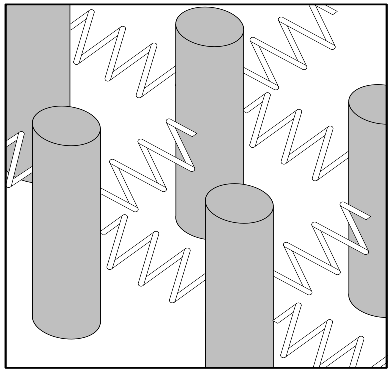
$$d(t) = \alpha(t_0 - t)$$

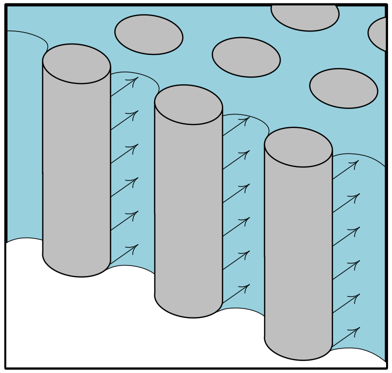


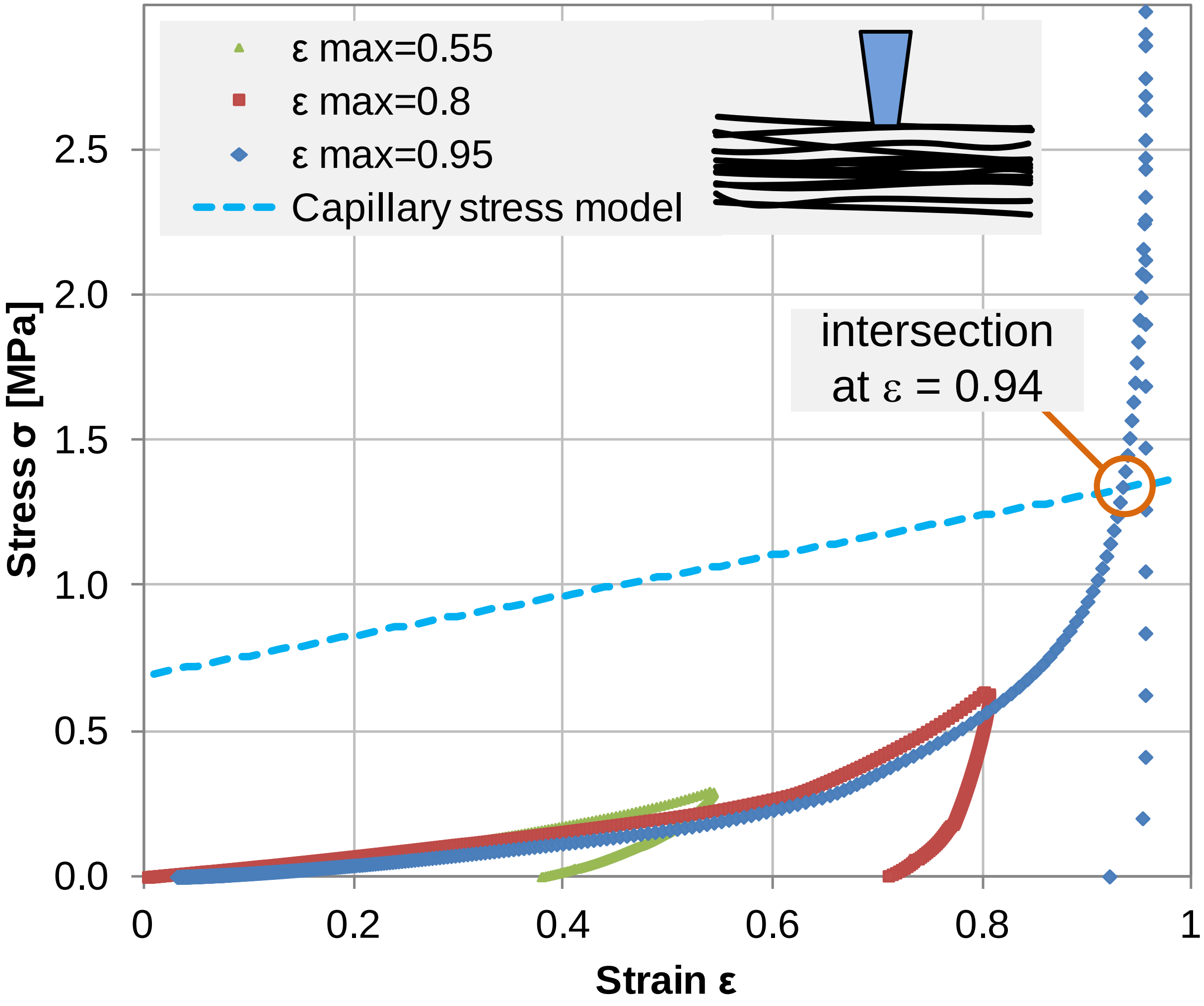
Tawfick et al., Langmuir 2013

When will densification stop?

$$\sigma_c = \frac{2\gamma N}{1-\varepsilon}$$

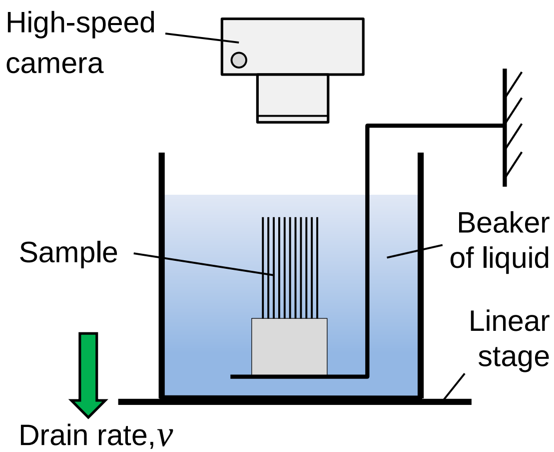


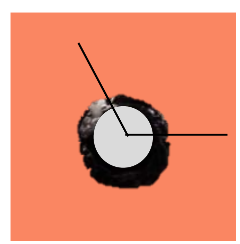




Tawfick et al., Langmuir 2013

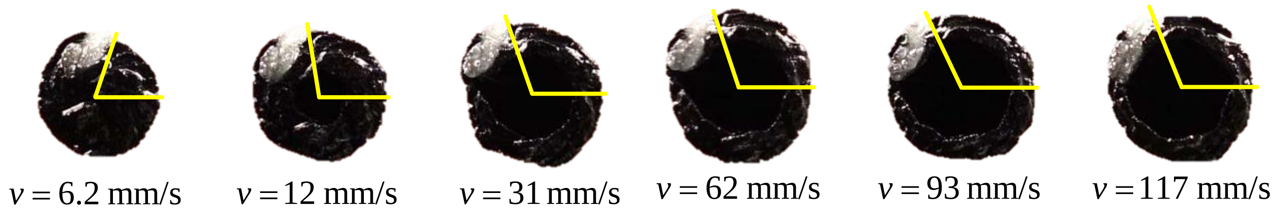
Watching the Kinetics of Capillary Twisting

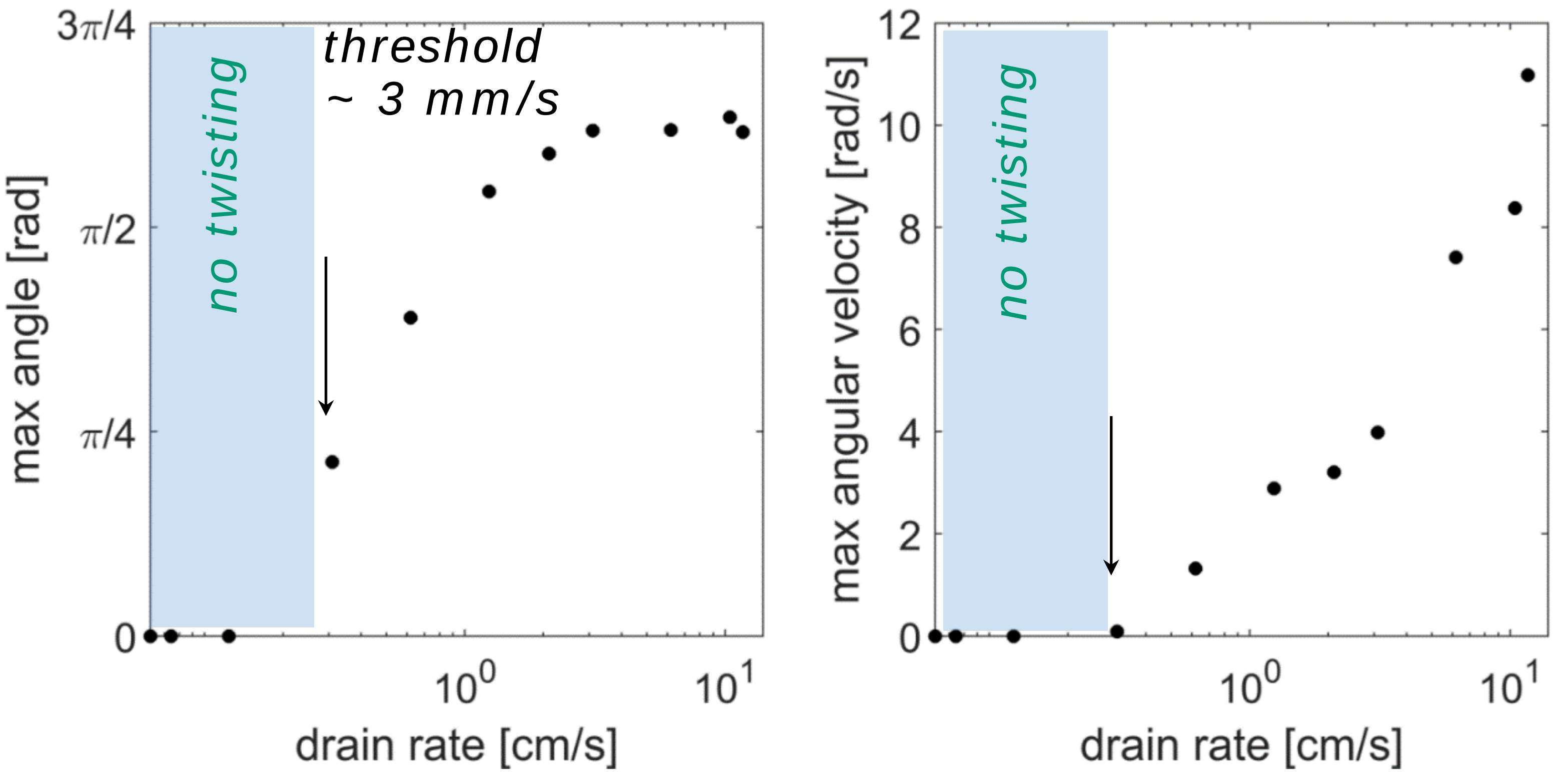




Shin and Tawfick Langmuir (2018)
Kovanko and Tawfick, in preparation

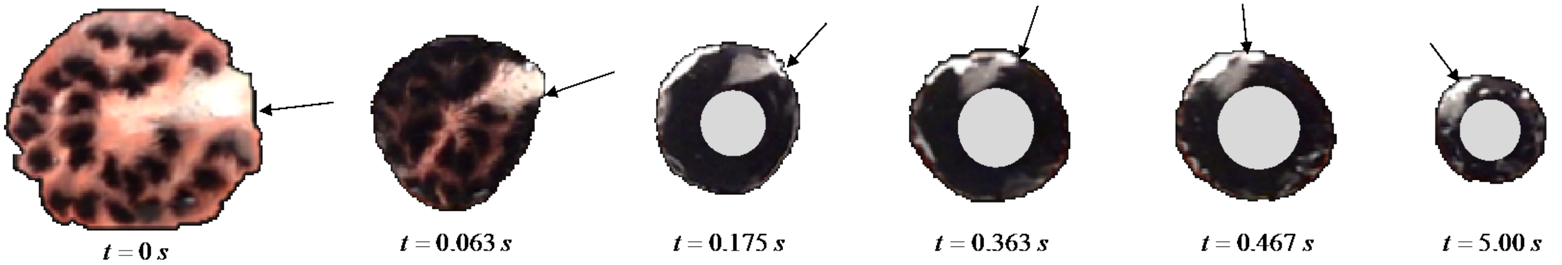
Threshold and Scaling of Capillary Twists





Kovanko and Tawfick, in preparation

Kinematics of Capillary Twists



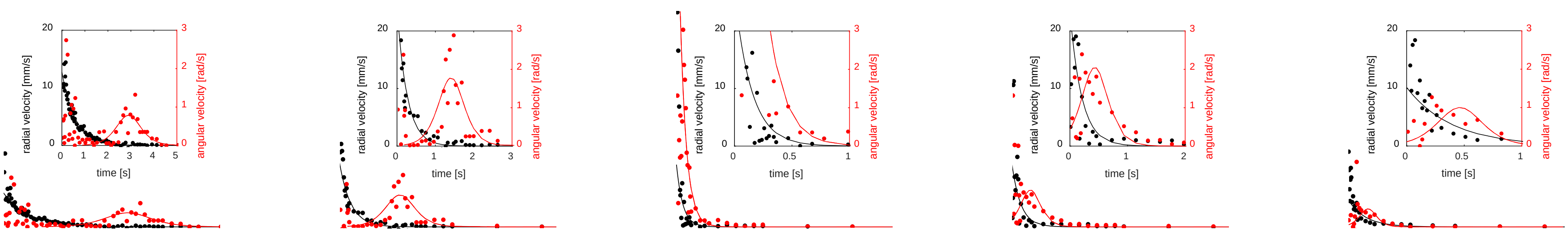
a l = 21 mm
v = 6.2 mm/s

b l = 21 mm
v = 12 mm/s

c l = 21 mm
v = 117 mm/s

d l = 19 mm
v = 117 mm/s

e l = 17 mm
v = 117 mm/s



Kovanko and Tawfick, in preparation

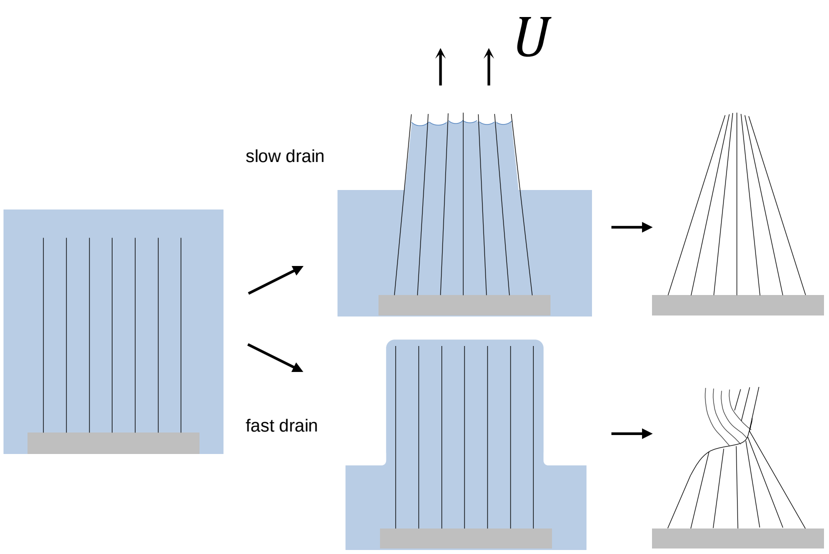
Threshold of Capillary Twists

Capillary work rate ~ $\frac{\gamma}{S} A v_{lat}$

Viscous work rate ~ $\mu \frac{U^2}{S} A$

Volume conservation $\frac{v_{lat}}{S} \sim \frac{U}{l}$

$$v_{lat} \sim \frac{\gamma}{\mu} \left(\frac{S}{l}\right)^2 \sim 2\ mm/s$$

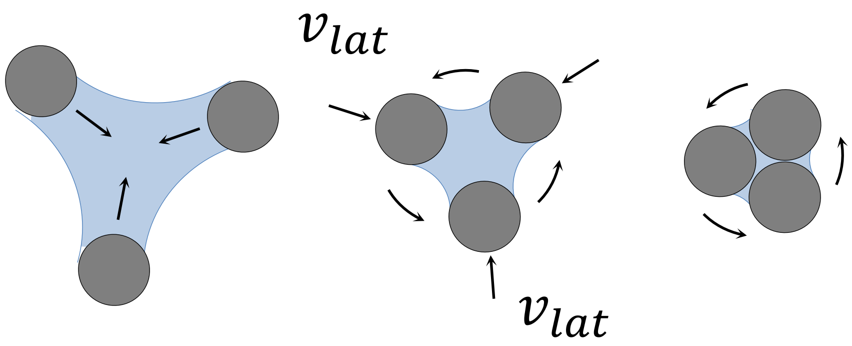


l fiber length
S fiber spacing
μ viscosity
γ *surface energy*

Kovanko and Tawfick, in preparation

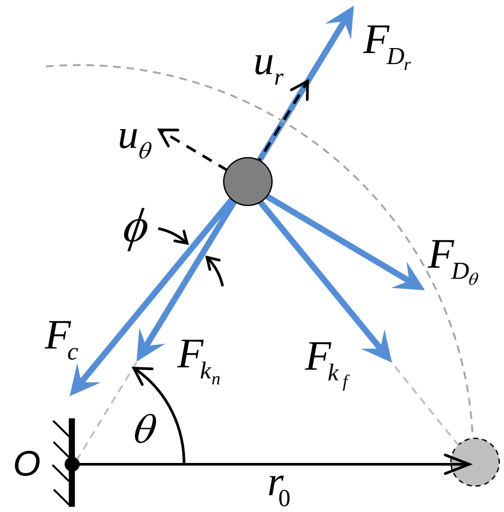
Forces Inducing Twists

Anisotropic forces



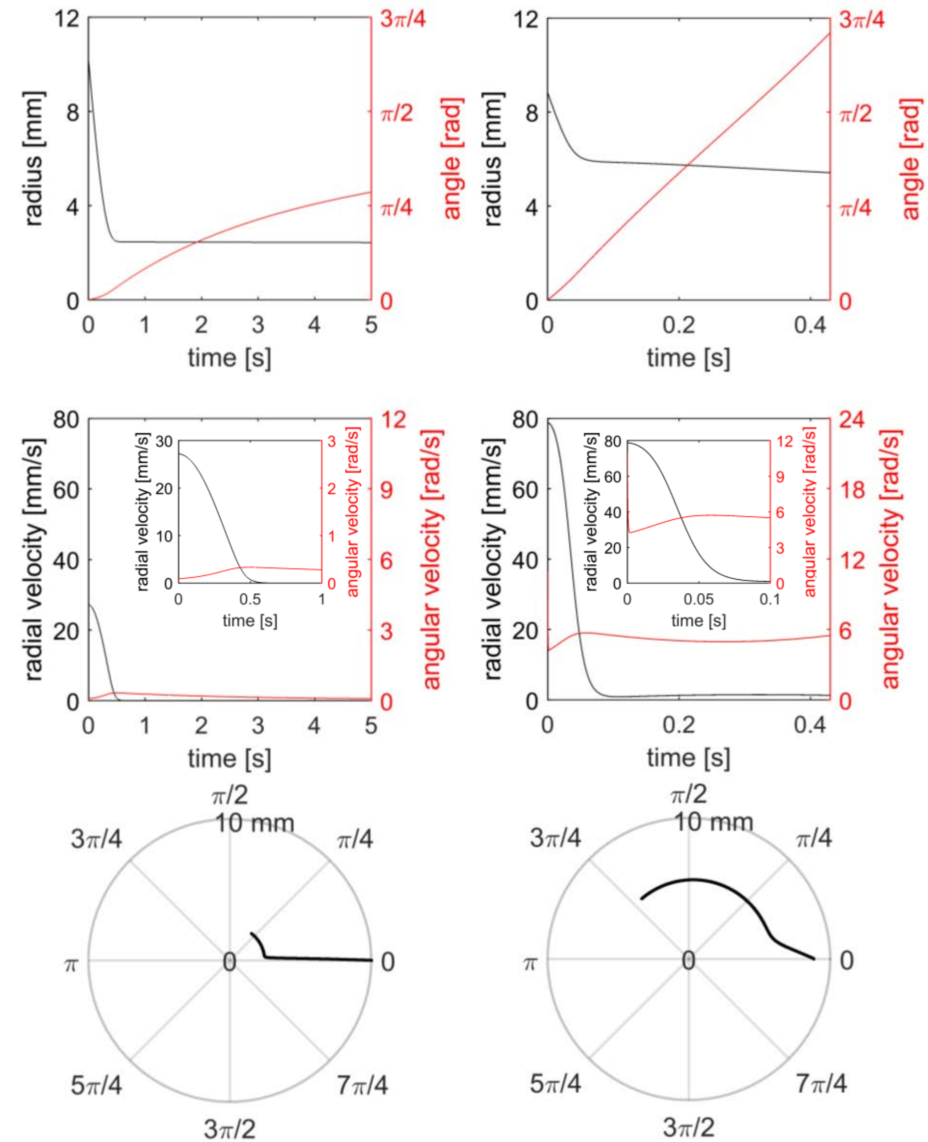
→ *Stiff in the radial direction, soft in the tangential direction*

Modeling Capillary Twists



$$m\ddot{r} = \overbrace{\left(\boldsymbol{F}_{kn} + \boldsymbol{F}_{kf,r}\right)}^{\text{Spring forces}} \overbrace{\left[\frac{r_0 - r_D}{\left(r_0 - r_D - r_{el} + r(t)\right)}\right]^n}^{\text{Fiber densification}} + \boldsymbol{F}_{centripetal} + \overbrace{\boldsymbol{F}_{D,r}}^{\text{Radial drag}} + \boldsymbol{F}_{capillary,r}$$

$$mr\ddot{\theta} = \underbrace{\boldsymbol{F}_{kf,\theta}}_{\text{Spring force}} + \boldsymbol{F}_{coriolis} + \underbrace{\boldsymbol{F}_{D,\theta}}_{\text{Tangential drag}} + \boldsymbol{F}_{capillary,\theta}$$



Kovanko and Tawfick, in preparation

Polymorphic elastocapillary self-assembly

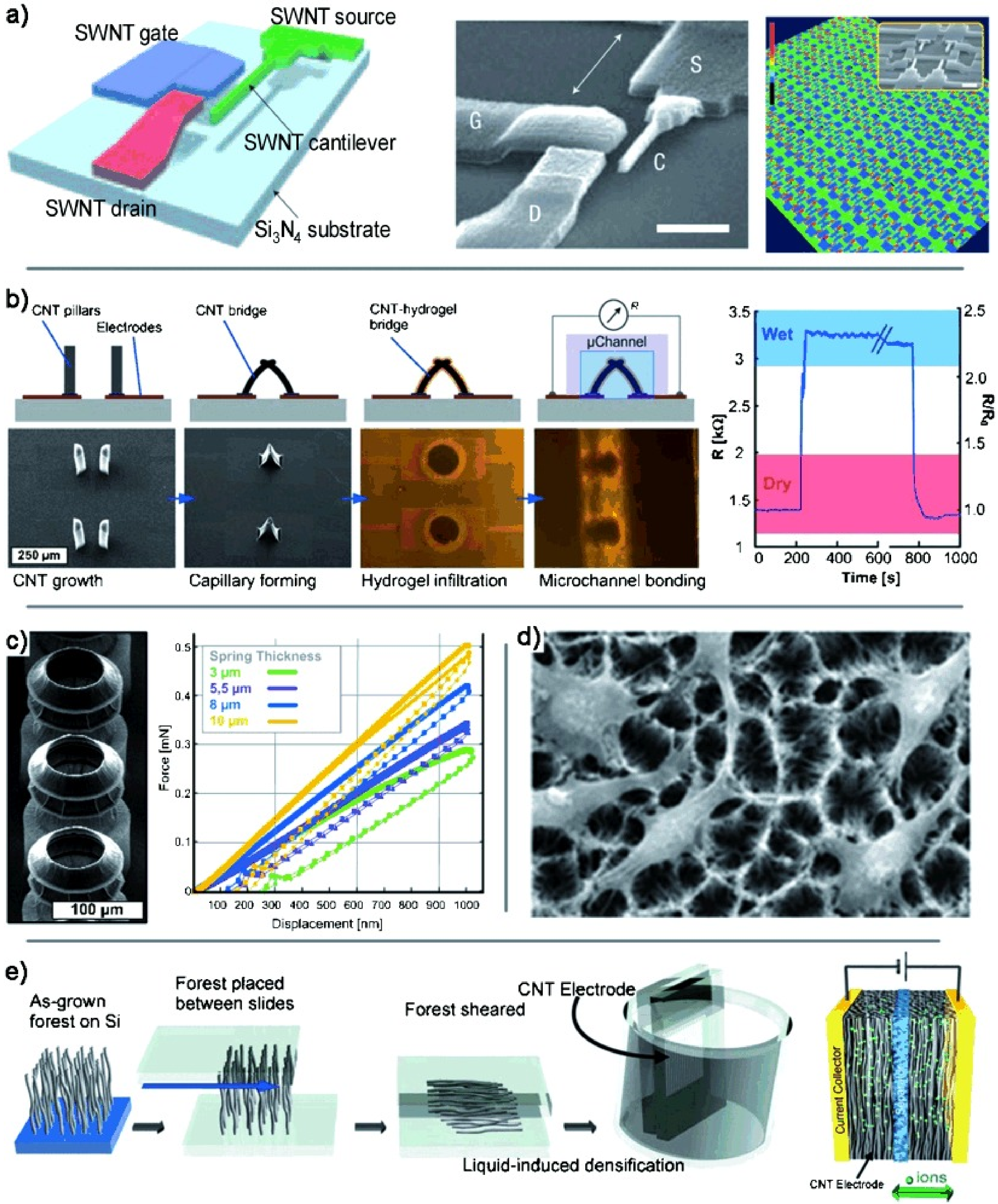
https://www.dropbox.com/s/x58pc3kp020uctn/S2-2018.avi?dl=0

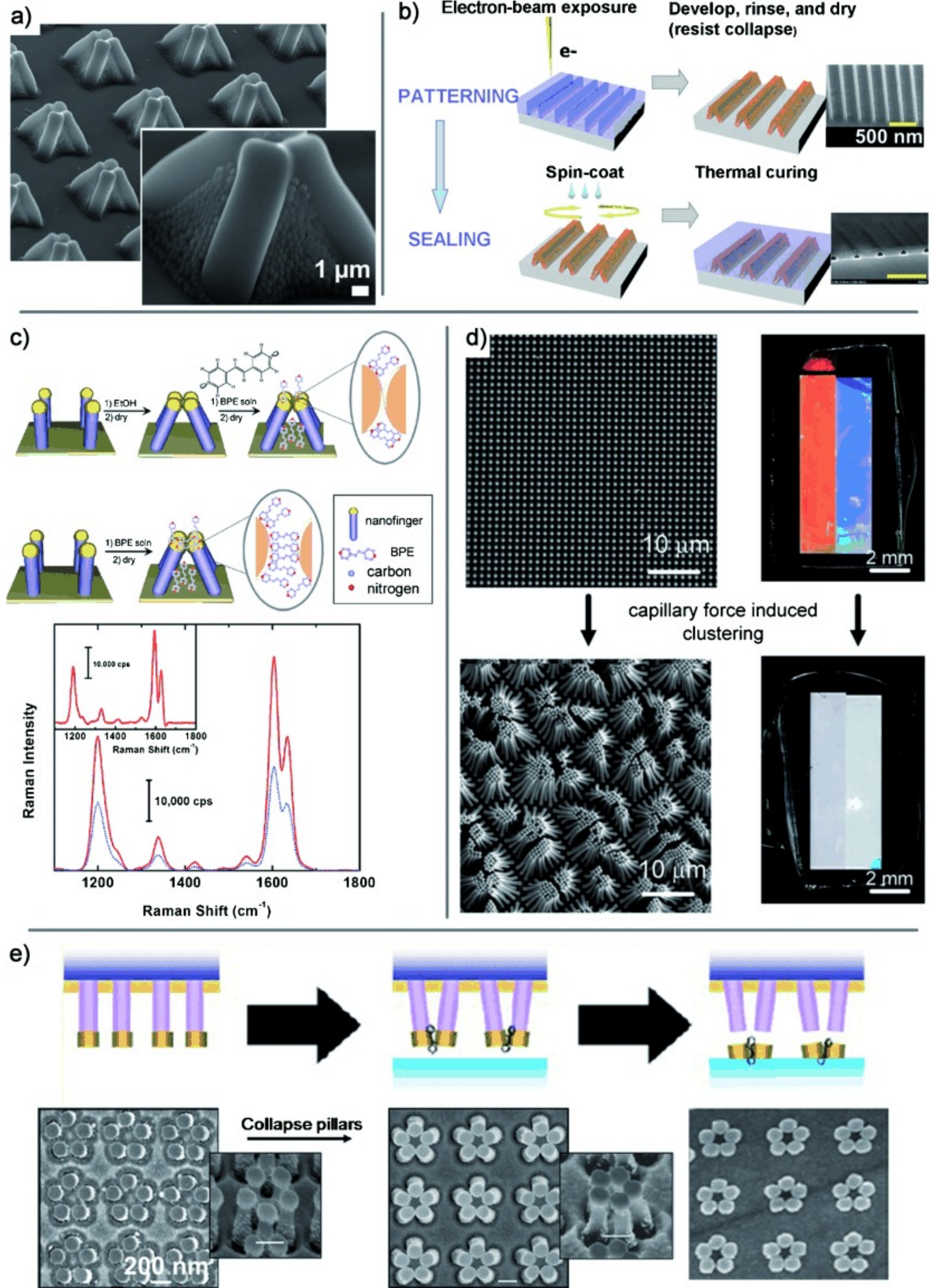
Shin and Tawfick, Langmuir (2018)

Polymorphic elastocapillary self-assembly

Shin and Tawfick, unpublished

De Volder and Hart, Angewandte Chemie, 2013

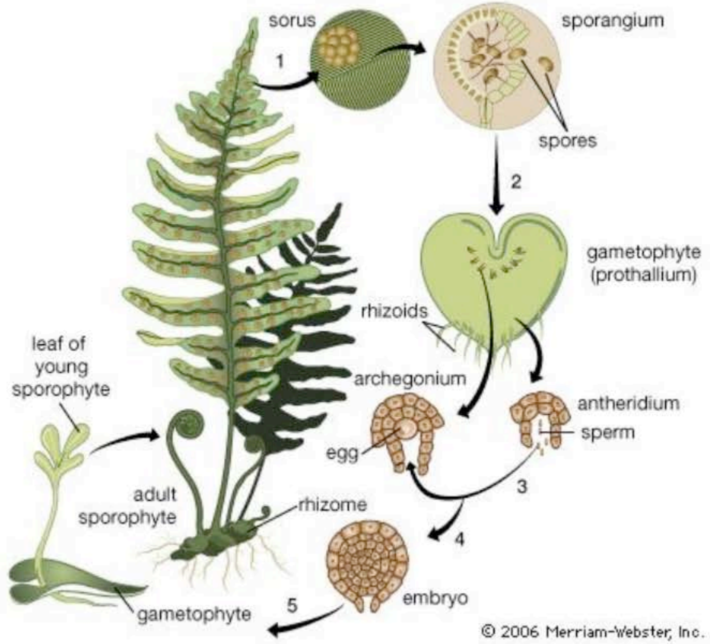




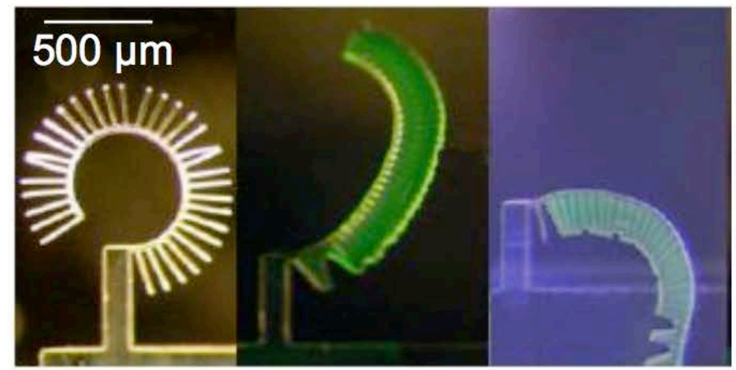
De Volder and Hart, Angewandte Chemie, 2013

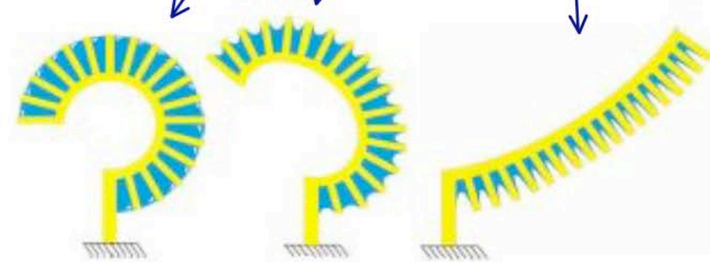
Capillary actuators

Slide by J. Bico, ESPCI France

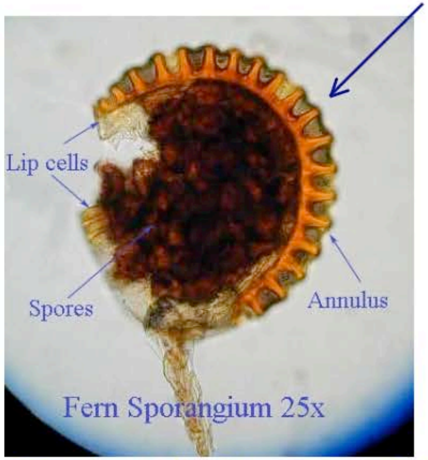


Wet hair actuator





rib like structure



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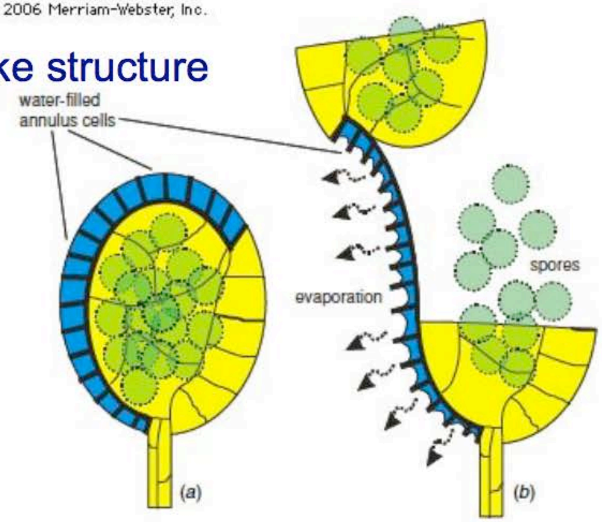
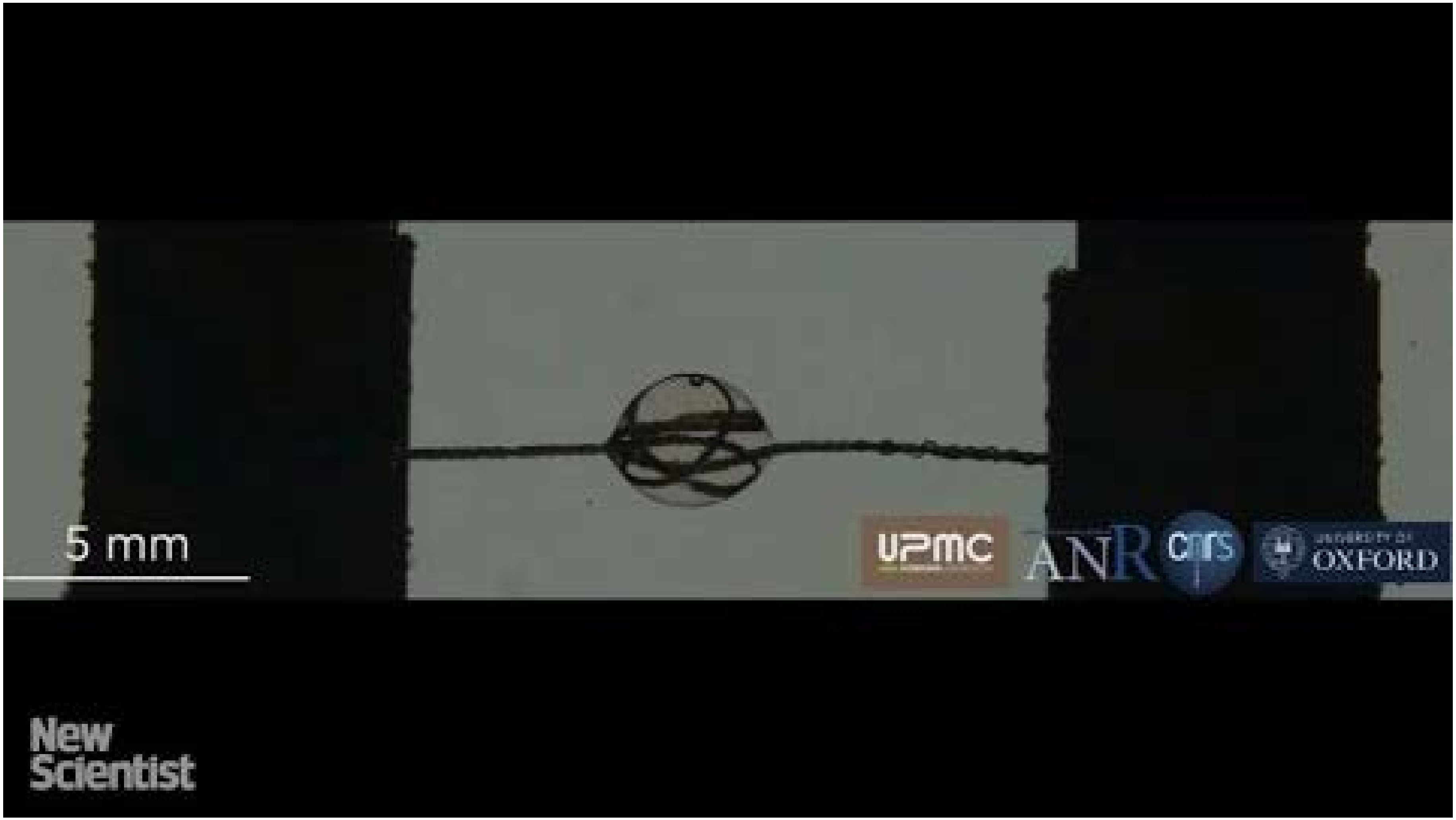


Borno et al. J. Micromech. Microeng. 16 (2006)

Capillary origami: wrapping droplet → Drug delivery!

Paulsen et al. Nature Materials (2015) https://www.youtube.com/watch?v=nfu9AjacvtY

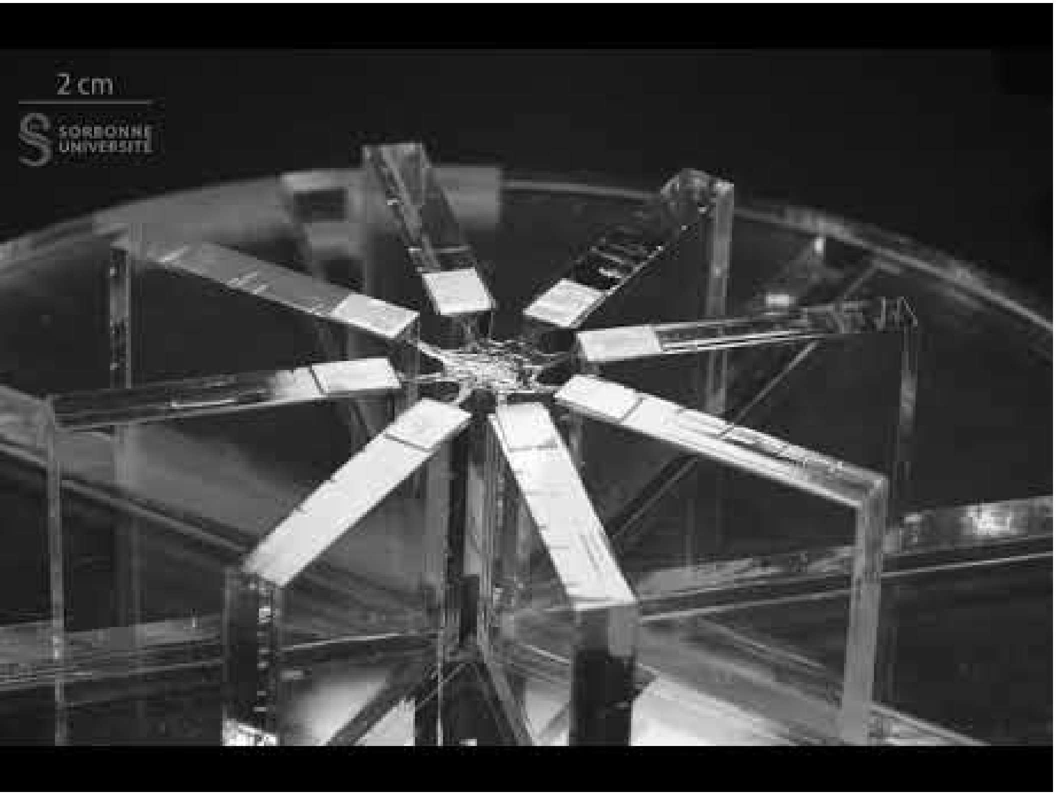
Elastocapillary spooling

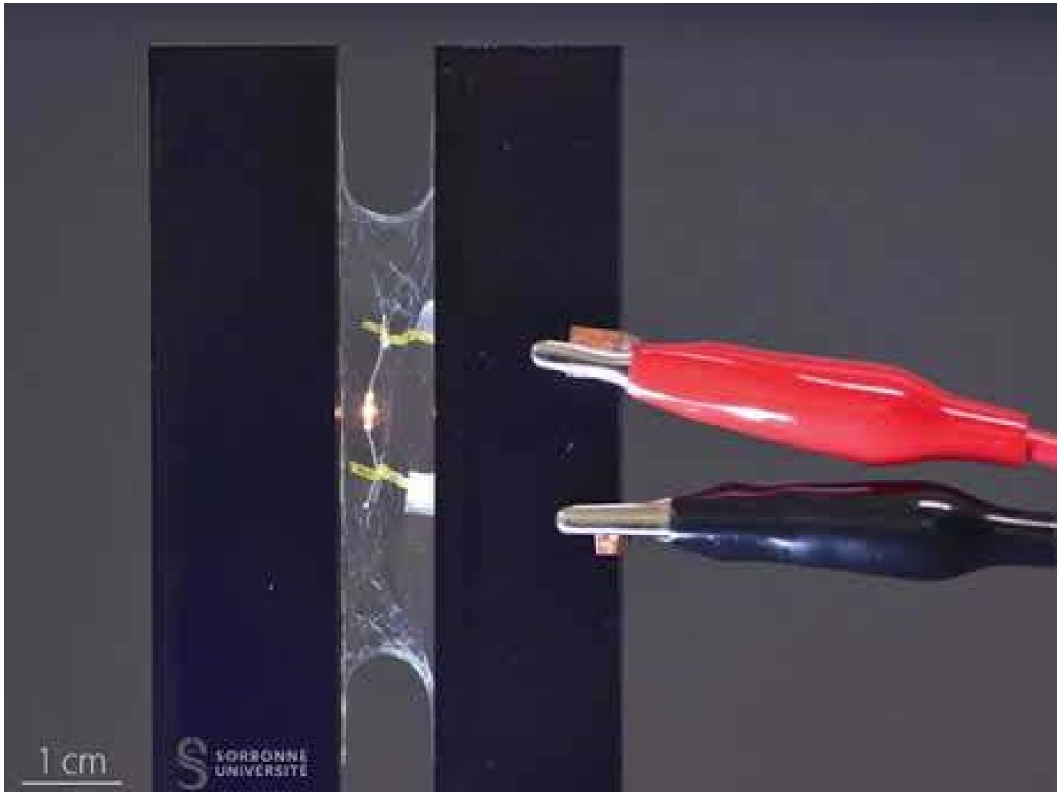


Elettro et al. PNAS (2016)

https://www.youtube.com/watch?v=NUGI5VW4frI

Capillary elasticity: wicked membranes





Grandgeorge et al. Science (2018)

https://youtu.be/CXJletDh4N8

Summary

Elastocapillarity: elastic energy ~ surface free energy

→ self-assembly from the nano- to the millimeter scales
→ Complex programmable geometries
→ Unusual behavior such as spontaneous twisting or spoolin
→ Shape morphing