Supporting Information

Wet-expandable capsules made from partially modified cellulose

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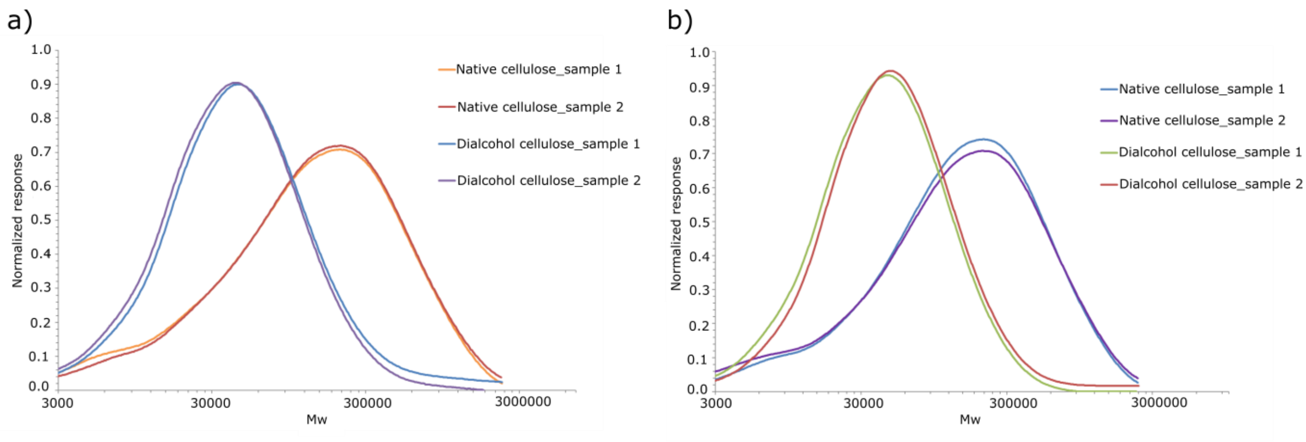

Fig. S1. Normalized SEC chromatograms for regenerated native cellulose and dialcohol cellulose solutions determined by using a) RI and b) UV detectors.

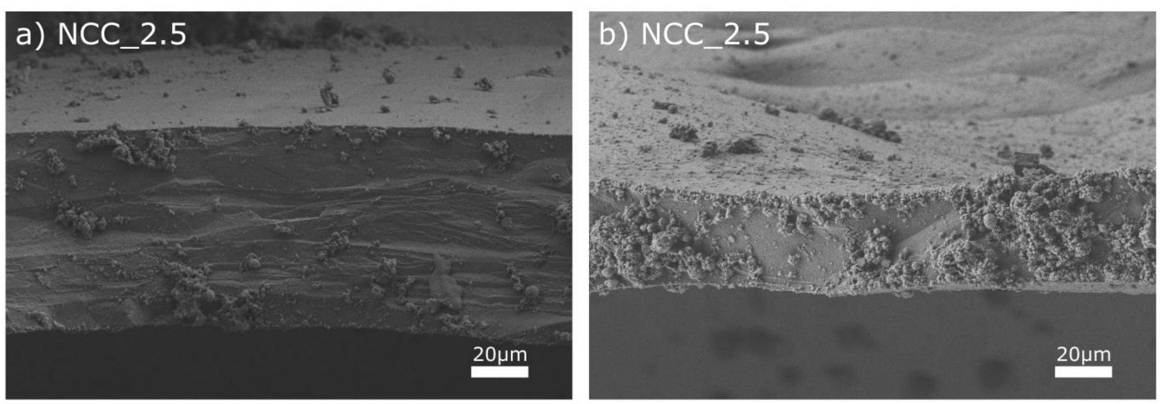

Fig. S2. Cryo-SEM images of the wet shell of native cellulose capsules in the a) thickest and b) thinnest region.

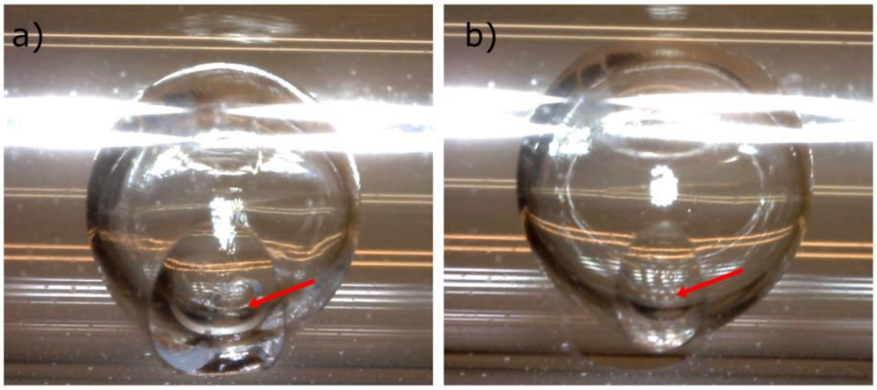

Fig. S3. Dialcohol-modified cellulose capsule tested using a "fast" rate of pressure change a) at the beginning of the expansion experiment when there is no decrease in external pressure, b) when a pressure difference of 100 mbar is applied. The water meniscus is denoted by a red arrow.

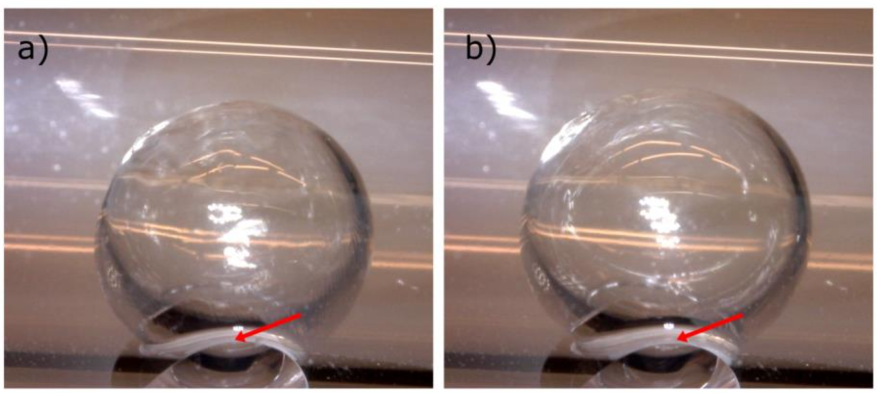

Fig. S4. Dialcohol-modified cellulose capsule tested using a "slow" rate of pressure change a) at the beginning of the expansion experiment when there is no decrease in external pressure, b) when a pressure difference of 350 mbar is applied. The water meniscus is denoted by a red arrow.

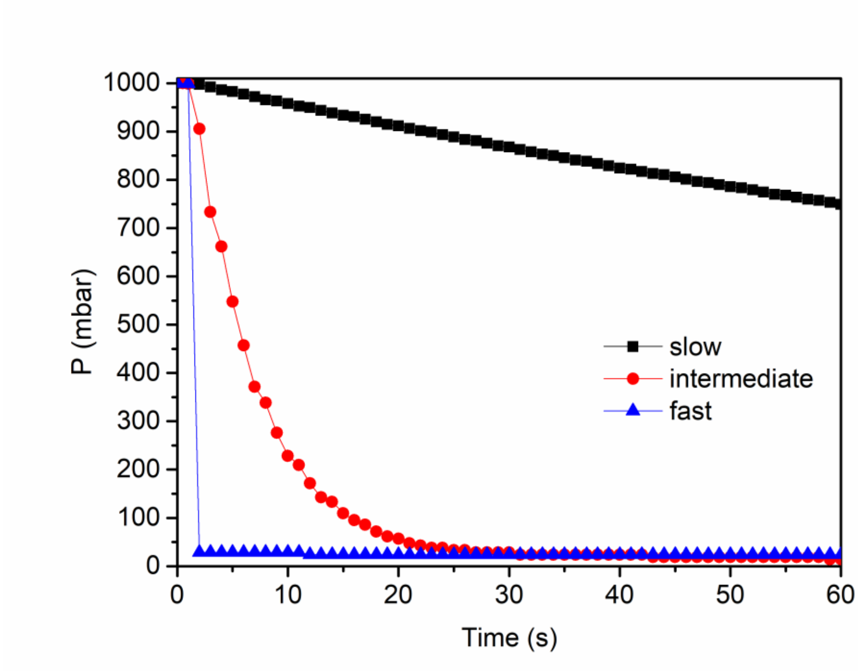

Fig. S5. Decrease in ambient pressure in the sample chamber over time at different positions of the leak valve. To perform the experiments, a pressure of 1 mbar was set in the pre-chamber, next the leak vale was opened and the time needed to reach the equilibration pressure was measured.

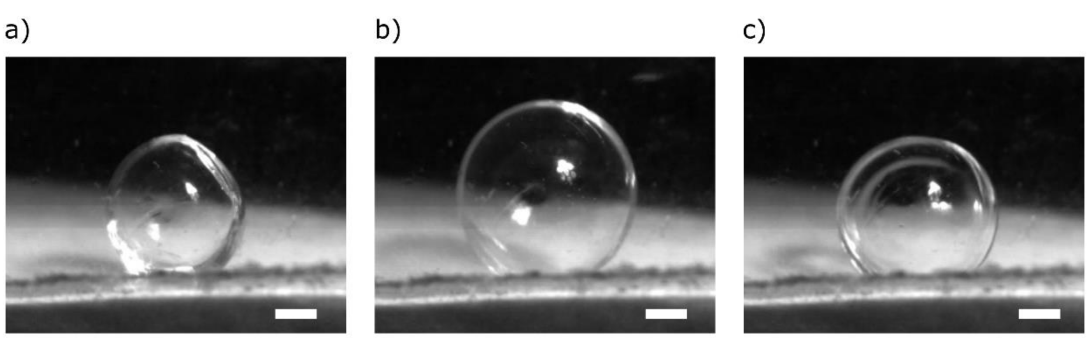

Fig. S6. Images showing the behavior of the modified capsule when the external pressure is cycled. a) Capsule in its initial (non-expanded) state, b) expansion of the capsule when the external pressure is decreased, c) shrinkage of the capsule when the external pressure is increased back to 1 atm. The scale bars indicate 10 mm.

Calculations of the moduli of the capsules shell

Provided that the diameter of the capsules is much larger than the shell thickness, the relationship between the linear circumferential expansion of the capsule and the pressure difference across the capsule shell can be described by Equation 1.[1,2]

$$\frac{r-r_0}{r_0} = \frac{1-\nu}{E}\frac{\Delta P r_0 \lambda^2}{2t_0} \qquad \text{valid if } {t_0}^2 << 2r_0 t_0 \qquad \text{(Equation 1)}$$

Where r is the capsule's radius at a pressure difference ΔP, r_0 is the initial radius of the capsule, ν is the Poisson's ratio of the shell, ΔP is the pressure difference between the inside and outside of the capsule, E is the linear deformability/modulus of the shell, t_0 is the initial shell thickness, and $\lambda = \frac{d}{d_0}$ is a factor that compensates the difference between engineering stress and true membrane stress i.e. to normalize with the non-deformed area.[2]

By fitting the experimental data, i.e. the results from the expansion measurements at different pressures, to Equation 1, it is possible to calculate the modulus (elastic or strain hardening) of the shell assuming that Poisson's ratio is known. In a previous study, the Poisson's ratio of films made of cellulose fibers partially modified to dialcohol cellulose was found to lie between 0.2 and 0.33 (depending on the relative humidity).[3] Another study reports a value of 0.3 for films made from the regeneration of native cellulose.[4] Consequently, the moduli of DCC and NCC were calculated assuming a Poisson ratio of 0.3. Since the expansion of a capsule is given by $\frac{r-r_0}{r_0}$ and the engineering stress by $\frac{\Delta P r_0 \lambda^2}{2t_0}$, a stress–strain curve can be plotted (where the slope is $\frac{E}{1-\nu}$) as shown in Fig. S7.

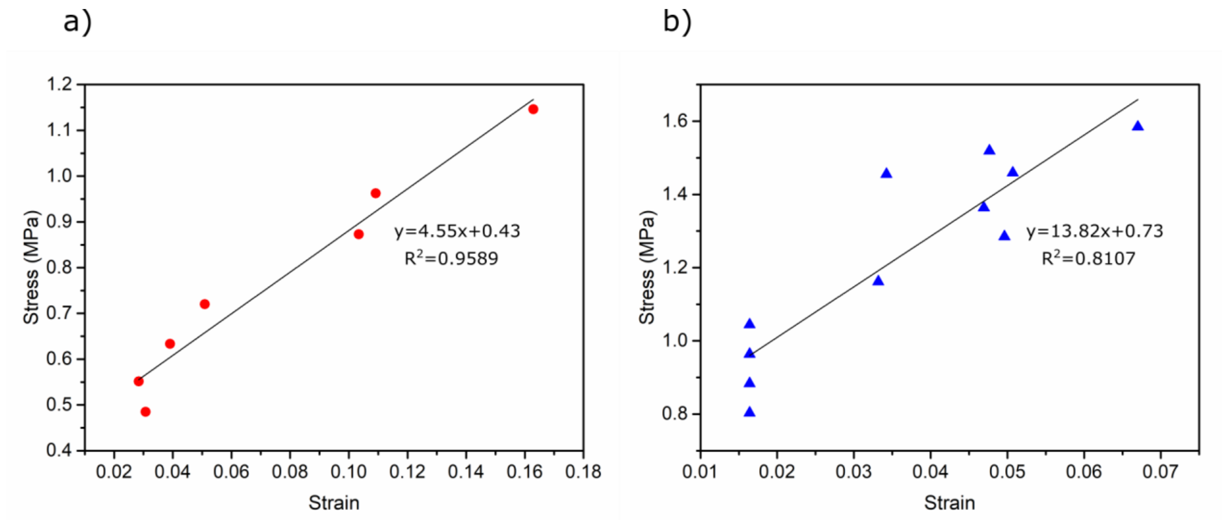

Fig. S7. Stress–strain curves determined for a) DCC_2.5 and b) NCC_2.5.

References:

1 P. Kelly, *Solid mechanics part I. I: An Introduction to Solid Mechanics*, 2014.

2 M. A. Johnson and M. F. Beatty, *Int. J. Eng. Sci.*, 1995, **33**, 223–245.

3 E. Linvill, P. A. Larsson and S. Östlund, *Mater. Des.*, 2017, **128**, 231–240.

4 W. Gindl, M. Reifferscheid, R. B. Adusumalli, H. Weber, T. Röder, H. Sixta and T. Schöberl, *Polymer (Guildf).*, 2008, **49**, 792–799.