**Cationic surfactants as a non-covalent linker for oxidised cellulose nanofibrils and starch-based hydrogels**

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**Electronic supplementary information (ESI)**

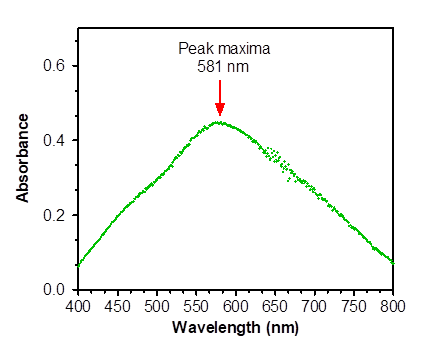
**Amylose content determination:**

The amylose content in the starch used was determined by using the method of (Sadasivam & Manickam, 1996; Yuliana, Huynh, Ho, Truong, & Ju, 2012). Briefly, 100 mg of starch was taken in a 100 mL volumetric flask, then first 1 mL of absolute ethanol followed by 10 mL 1N NaOH was added and allowed to store overnight at room temperature. The suspension was made up to 100 mL and placed on a boiling water bath for 10 min. After cooling down to room temperature, 2.5 ml extract was taken into another 50 mL volumetric flask. Then 20 mL of DI water was added followed by the addition of 2 drops of phenolphthalein (0.1%) indicator. The resultant mixture was titrated against HCl (0.1N) until the pink colour disappeared. 1 mL of Iodine reagent (0.25 g of I2 and 2 g of KI in 100 mL of DI water) was added and the volume was made up to 50 mL (thus, the concentration of starch in the test solution used for the UV measurement was 0.05mg/ml).

The absorbance of the suspension was taken using a UV/visible spectrometer (Varian Cary 50 Probe) by scanning over the wavelength range of 300 to 800 nm. The peak maximum was observed at 581 nm. Absorbance at 581 nm (0.45) corresponds to the concentration of the sample in the final test solution.

Synthetic amylose (MW 100 KDa, purchased from Shoko Ltd, Japan) was used as a standard to draw the calibration curve. The absorbance at 581 nm of various concentration of the standard amylose suspension (0.008, 0.012, 0.016, 0.020 and 0.050 wt%) was taken and plotted against the concentration.

From the standard calibration curve, absorbance at 0.45 was obtained for 0.017 wt% of amylose. Thus, the amylose content in the starch sample was calculated to be 0.017/0.05 = 34%.



**ESI 1: a)** UV-Visible spectrophotometry curve of the starch and **b)** calibration curve obtained from the absorbance vs concentration of standard synthetic starch suspension.

**References**

Sadasivam, S., & Manickam, A. (1996). *Biochemical methods*. India: New Age International Publishers.

Yuliana, M., Huynh, L.-H., Ho, Q.-P., Truong, C.-T., & Ju, Y.-H. (2012). Defatted cashew nut shell starch as renewable polymeric material: Isolation and characterization. *Carbohydrate Polymers,* 87(4), 2576-2581.



**ESI 2:** Tan delta values show the effect of surfactant concentration on the gelation of OCNF/surfactant systems.



**ESI 3:** Frequency sweeps of OCNF:Starch gels. Closed and open symbols represent G’ (storage modulus) and G’’ (loss modulus), respectively.



**ESI 4:** Frequency sweep curves of the starch (2 wt%), OCNF (2wt%) and OCNF:starch (1:1 wt%) hydrogels. In the blend hydrogels, the addition of starch to the OCNF resulted in stronger rheological properties compared to the starch gels, where starch polymers were suggested to form an entangled network with cellulose fibrils.



**a**

**b**

**c**

**d**

**ESI 5:** Photo images of OCNF:Starch/surfactant systems: **a)** OCNF:Starch (1:0.5 wt%)/DTAB-5 mM, **b)** OCNF:Starch (1:1 wt%)/DTAB-5 mM, **c)** OCNF:Starch (1:0.5 wt%)/CTAB-5 mM, **d)** OCNF:Starch (1:1 wt%)/CTAB-5 mM (photos taken 24 h after gel formation).



**a**

**b**

**c**

**d**

**e**

**f**

**ESI 6:** Photo images of Starch (1wt%) solution at various concentrations of surfactants: **a)** DTAB-1 mM, **b)** DTAB-5 mM, **c)** DTAB-10 mM, **d)** CTAB-1 mM, **e)** CTAB-5 mM, and **f)** CTAB-10 mM.



**ESI 6:** ζ-potential values of OCNF:Starch blend (0.1 wt%) systems.



**ESI 7:** ζ-potential values of diluted starch (0.1 wt%)/surfactant systems.



**a)**

**b)**

**ESI 8:** **a)** Amplitude sweeps of OCNF:Starch/DTAB and **b)** OCNF: Starch/CTAB gel systems. Closed and open symbols represent G’ and G’’, respectively.



**a)**

**b)**

**ESI 9:** Photographs show the **a)** iodine stock solution (50% of Lugol’s solution), and **b)** dilute iodine solution (25x dilution with DI water).