



Unless otherwise noted, the sequence linking the fluorescent protein to the CBM is composed of a glycine. **A**) The crystalline cellulose probe was formed by linking eGFP via a thrombin cleavage site to CBM3a (46.26 kDa). **B**) The amorphous cellulose probe is composed of mCherry linked to CBM17 (50.56 kDa). **C**) The xylan probe (44.68 kDa) was formed by mOrange2 and CBM15 while the mannan probe **D**) was constructed using eCFP and CBM27 (48.06 kDa). All fluorescent proteins C-terminal ends are linked to the N-terminal ends of the CBMs. The red portions in the ORFs represent N-terminal six histidines tags. The red spheres represent metal ions.





Binding curves of eGFP-CBM3a (A), mCherry-CBM17 (B), mOrange2-CBM15 (C) and eCFP-CBM27 (D) as calculated from solid state depletion assays. Cellulose substrates were: Whatman paper (*open circle*), kraft paper (*filled diamond*) and HYK (*open diamond*) unrefined papers. Binding was recorded after a 1h incubation at 23 °C of the various cellulose supports with the probes (100 μ g/well) in a 20 mM Tris-HCl pH 7.5 buffer containing 20 mM NaCl and 5 mM CaCl₂.



Fig. S3. XPS analysis of the HYK (A-B) and Kraft (C-D) pulps.

Low-resolution spectra of the surface of the pulps (**A-C**). Deconvolution of the C 1s band of the high-resolution spectra (**B-D**). The 300 W monochromatic Al K-a radiation source was used to study the surface composition of the pulps. The instrument resolution was 0.6 eV. The average of three different spots were recorded and analyzed using the CasaXPS software.

Table S1

Binding affinities (K_a) and capacities (N_o) of CBM probes for various carbohydrates polymers after 1h incubation at 23 °C in 20 mM Tris-HCl pH 7.5 containing 20 mM NaCl and 5 mM CaCl₂.

Probe	Binding support	<i>K</i> a (μM ⁻¹)	N₀ (μmol/g cellulose)
eGFP-CBM3a	Avicel	7.999	0.277
	Whatman	0.083	0.205
	КР	0.059	0.049
	НҮКР	0.101	0.019
mCherry-CBM17	Whatman	0.041	0,112
	КР	0.019	0,084
	НҮКР	0.008	0.089
mOrange2-CBM15	Xylohexaose	0.034 ^a	0.920 ª
	Whatman	0.007	0.335
	КР	0.026	0.049
	НҮКР	0.013	0.038
eCFP-CBM27	Mannohexaose	0.227 ^b	1.100 ^b
	Whatman	0.070	0.039
	КР	0.008	0.066
	НҮКР	0.009	0.317

These values (a-b) were determined by ITC.

^a Reference 21.

 Table S2

 Chemical composition of kraft pulps measured using NREL/TP-510-42618 methodologies.^{21,25}

Pulp polymer	Kraft (%)	НҮК (%)
Extractives	0.28 ± 0.01	0.43 ± 0.01
Lignin	4.8 ± 0.07	6.7 ± 0.15
Glucose	86.9 ± 1.22	81.93 ± 1.38
Xylose	7.1 ± 0.67	7.4 ± 0.87
Mannose	8.4 ± 0.58	8.7 ± 0.55

Table S3 XPS analysis of kraft pulps. Results include O/C ratios and contributions (%) from each carbon type (C1-C4) to curve fitting of the C 1s peak measured by low- and high-resolution XPS.^{11,21,29-30}

Functionality	Kraft (%)	НҮК (%)
O/C*	0.63 ± 0.02	0.56 ± 0.01
C1	16.06 ± 0.36	23.10 ±1.92
C2	67.63 ± 0.57	61.25 ±1.35
C3	15.33 ± 0.27	14.59 ± 0.68
C4	0.98 ± 0.07	1.07 ± 0.25

Spectra were taken from unextracted pulp samples.

*Low-resolution XPS spectra was used to obtain the oxygen and carbon percentage in order to ascertain that the O/C ratio does not vary as a function of chemical treatment.