

## **ADDITIONAL FILE 1**

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**Additional File : Figure S1. Impact of loosenin-like proteins on *NpXyn11A* hydrolysis of azo-xylan in solution.** Reactions were optimized to ensure absorbance values were within the linear range of the assay. The azo-xylan solution was treated with 0.003 mg/ mL PcaLOOL2, PcaLOOL12, BSA or buffer only at pH 5.0, 25°C for 24 h, followed by addition of 0.001 mg/ mL *NpXyn11A*, or buffer only, and incubation at 40°C, 300 rpm for 10 min. After precipitation of non-hydrolyzed substrate, absorbance of the reaction solution was recorded at 590 nm. n=3, errors correspond to standard deviation of mean.

**Additional File : Figure S2. Impact of loosenin-like proteins on cellulose/azo-xylan composites.** Composites were treated with 0.1 mg/ mL PcaLOOL2, PcaLOOL12, BSA or buffer only at pH 5.0 and room temperature for 24 h. This was followed by incubation of the resulting mandrels at pH 5.0, 40°C and 300 rpm for up to 6 h (A) Representative cellulose/azo-xylan composites dried after treatment. (B) Average intensity values of dried cellulose/azo-xylan composites per segment. (C) Absorbance of the reaction solution at 590 nm recovered after composite incubation. n≥4, errors correspond to standard deviation of mean.

**Additional File : Figure S3. In-solution hydrolysis of azo-xylan by *Pm25* and its mutants.** The azo-xylan solution was treated with 0.03 mg/ mL wild-type *Pm25*, 0.03 mg/ mL M5 (*Pm25* Y213A + Y378A), 0.017 mg/ mL M6 (*Pm25*ΔCBMs), 0.03 mg/ mL M1 (*Pm25* E546A), or buffer only, at pH 5.0 and 40°C, 300 rpm for 3-20 h. After precipitation of non-hydrolysed substrate, absorbance of the reaction solution was recorded at 590 nm. n≥4, errors correspond to standard deviation of mean.

**Additional File : Figure S4.** SDS-PAGE analysis of *CmXyn10B* (lane 2), *NpXyn11A* (lane 3) and *TfXyn11A* (lane 4). Lane 1 shows the molecular weight standard.

**Additional File : Figure S5.** Replicate cellulose/azo-xylan composites after treatment for 1-h with (A) *NpXyn11A* or (B) *TfXyn11A*

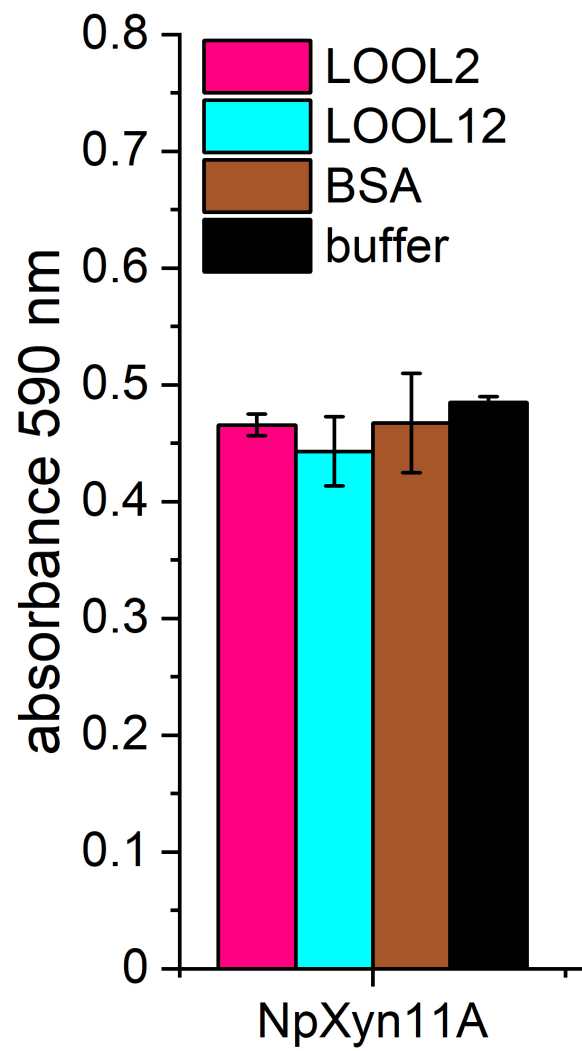
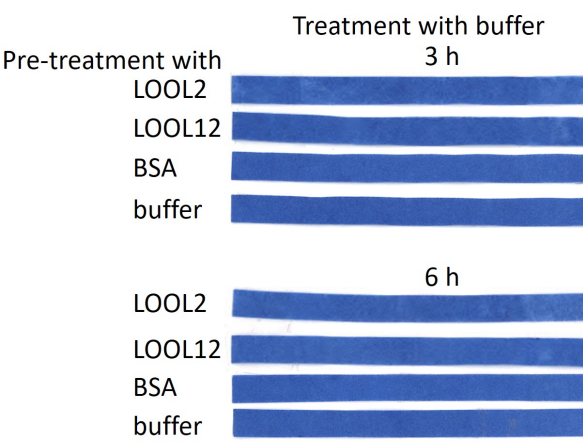
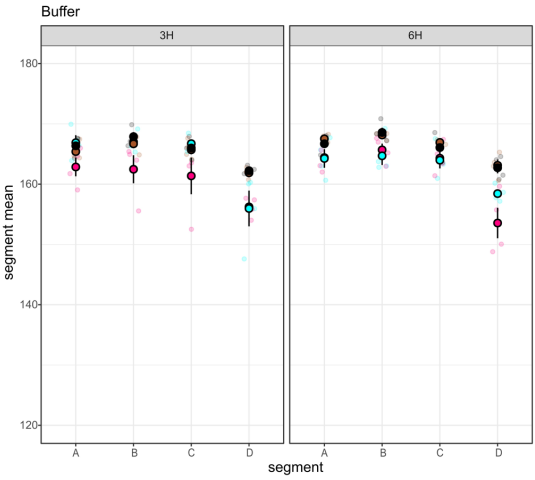


Figure S1

A



B



C

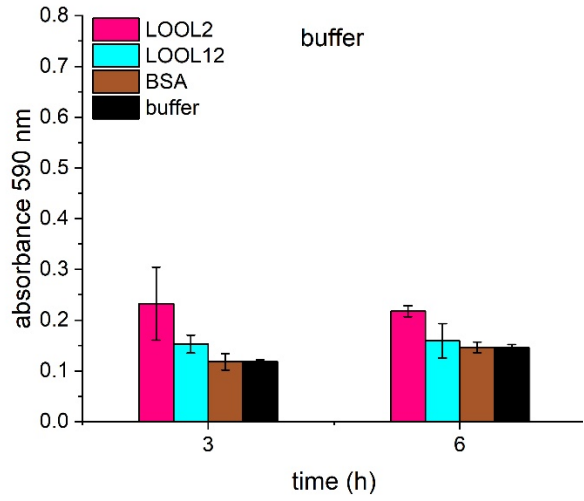


Figure S2

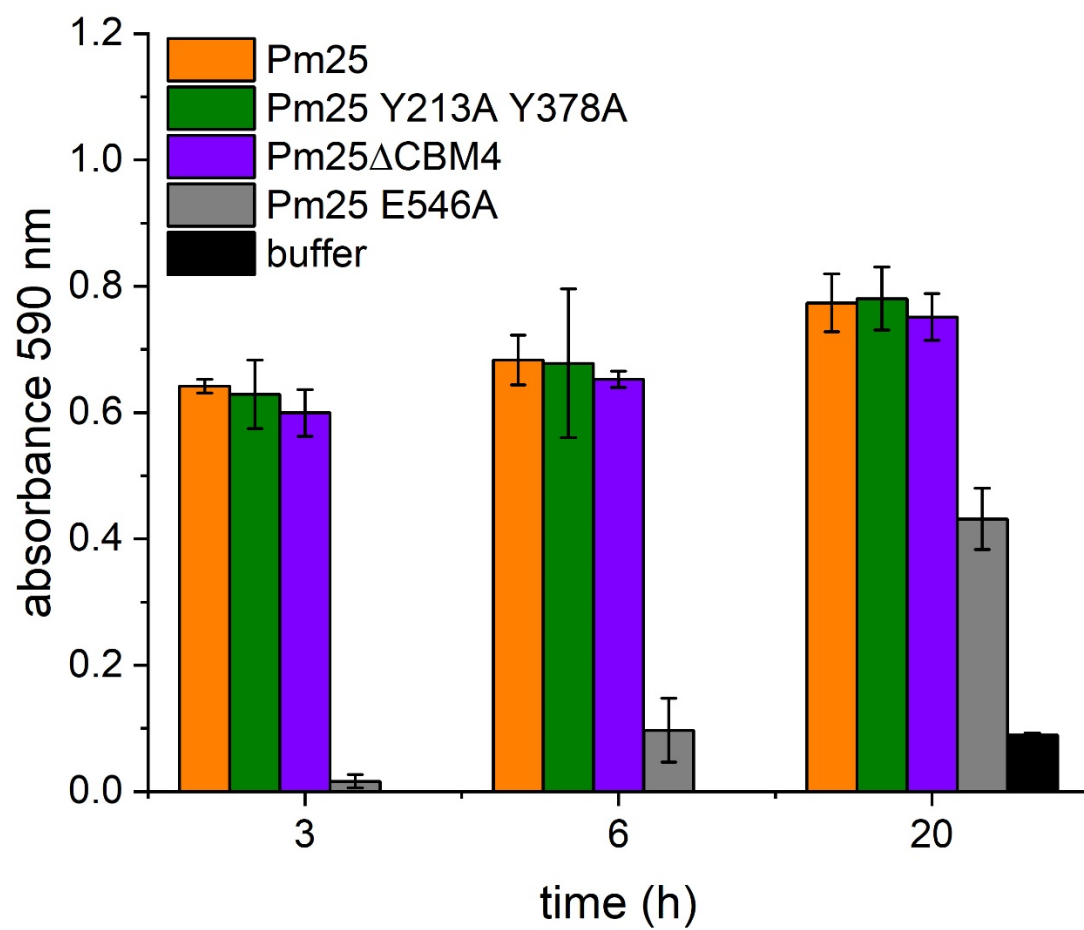


Figure S3

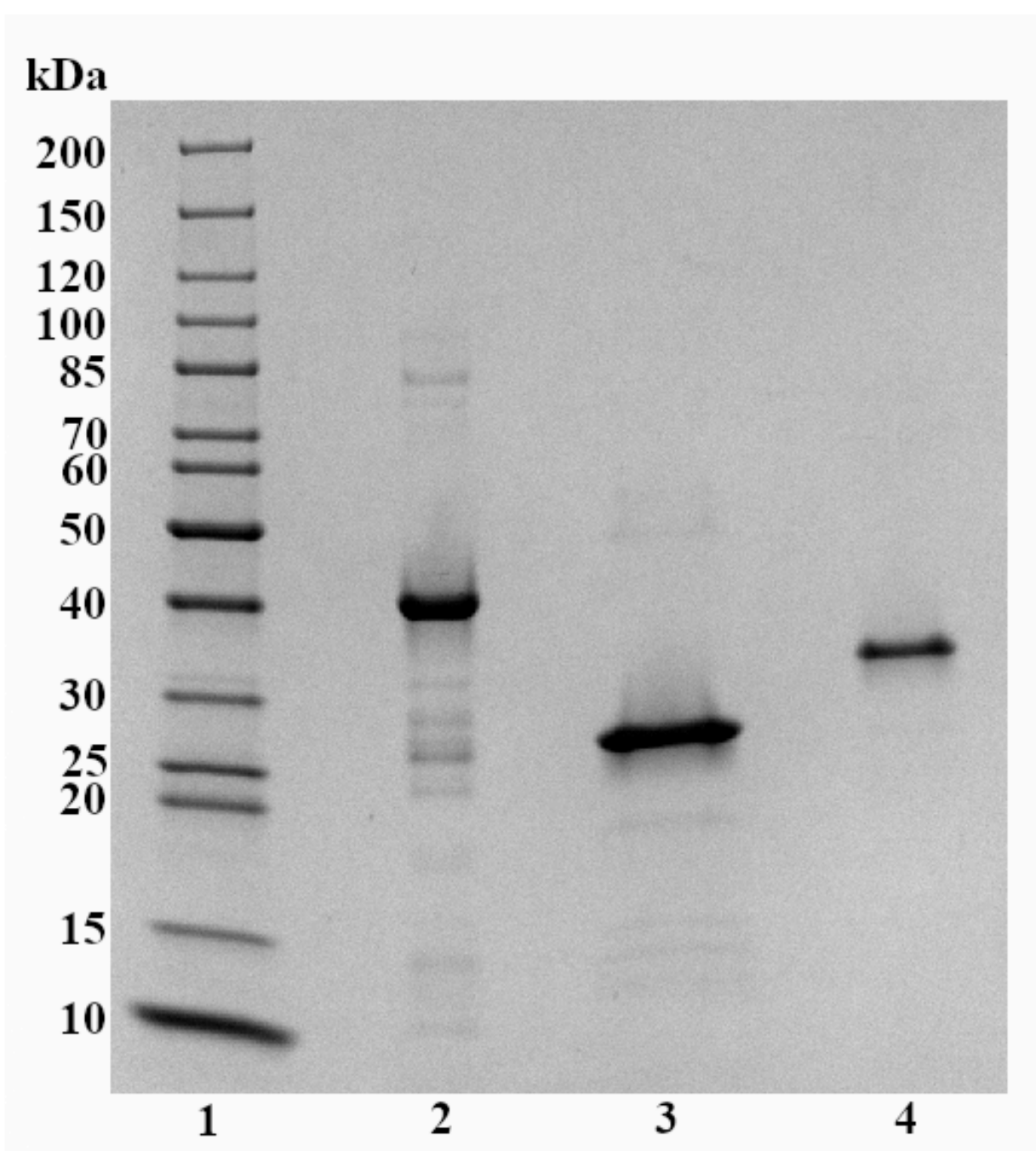


Figure S4

A. *NpXyn11A* treatments



B. *TfXyn11A* treatments



Figure S5