SUPPLEMENTARY INFORMATION

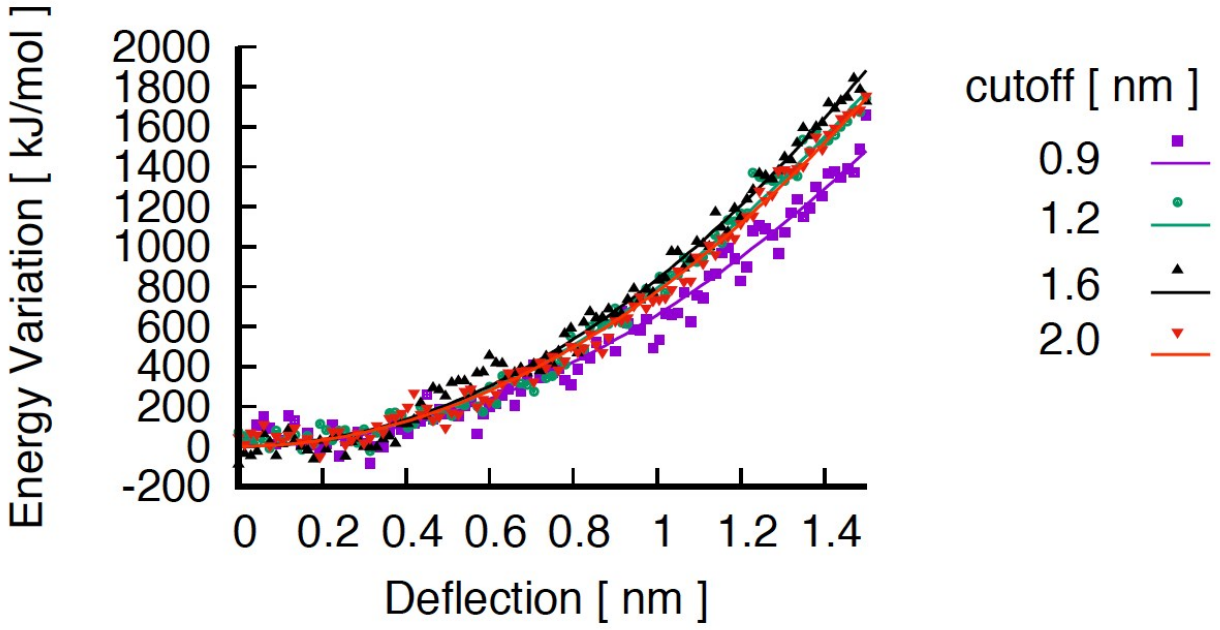


Figure S1. Energy deflection curves for bending by the 1-10 surface using Lennard-Jones and electrostatic cut-off values of 0.9 nm (violet), 1.2 nm, black 1.6 nm (black) and 2 nm (red).

Various cutoff values have been tested. The curve obtained by using 0.9 nm slightly underestimates the energy response of the cellulose crystal to bending whereas a similar resistance is obtained for cut-off values of 1.2 and 2 nm where the two curves superimpose. A cut-off of 1.2 nm is the most appropriate value.

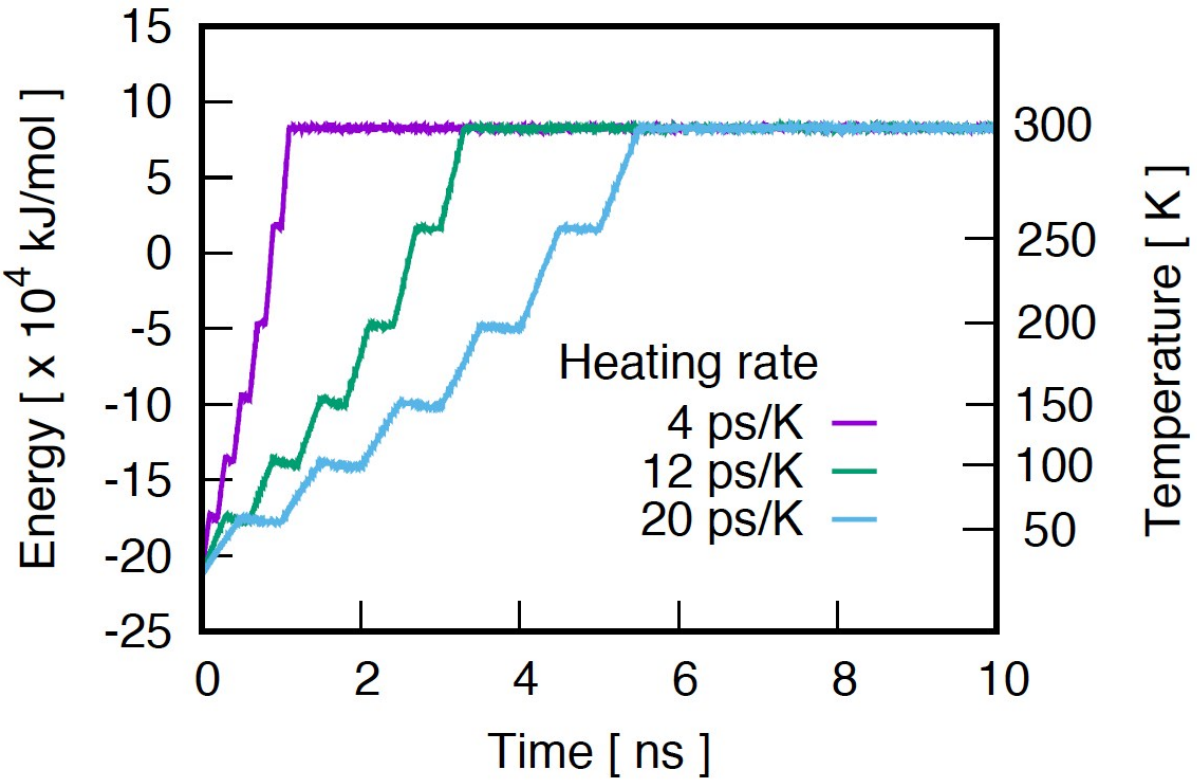


Figure S2. Variation of the energy of the finite crystal (the one exposing the (110) and (1-10) surfaces) as a function of time for various heating rates.

The duration of the heating up to 300 K may influence the equilibrated structure at room temperature. Various heating rates have been tested in order to identify the most appropriate value. All energies converge to the same value indicating that the final structure at 300 K is independent on the speed of heating. Consequently the value we use in our study is suitable.

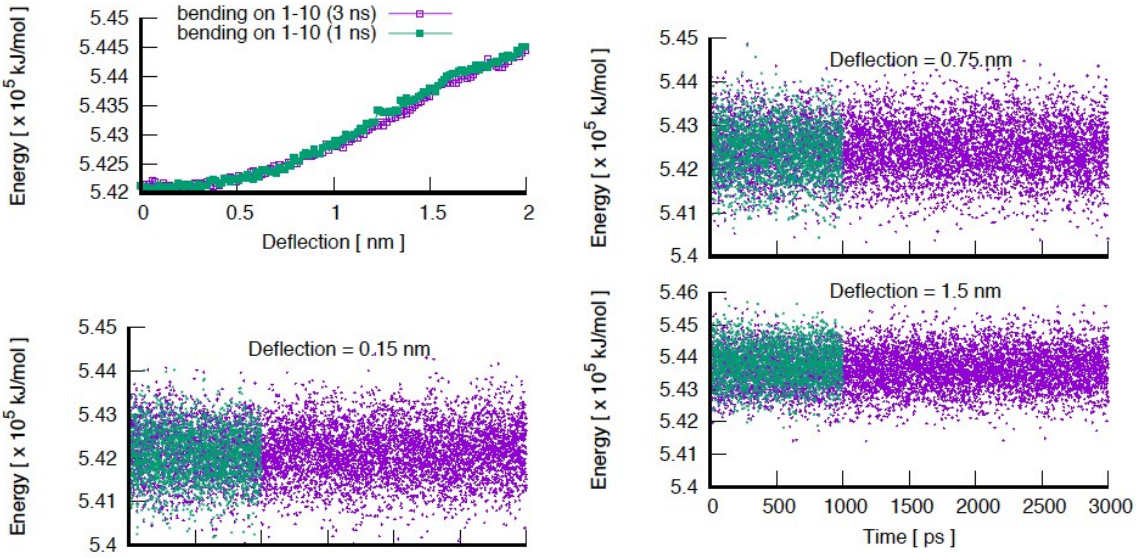


Figure S3. Effect of MD duration for equilibration at each bending step for a crystal bend by its 1-10 surface in the deflection range 0 – 2 nm. A: energy – deflection curves, energies as a function of time for deflections of 0.15 nm (B), 0.75 nm (C) and 1.5 nm (D).

The structures have to be equilibrated at each bending step; we used 1 or 3 ns for this purpose. Only minute differences can be seen in the energy-deflection curves (Figure S3A), similarly the time series at 3 deflection values are very similar, suggesting that 1 ns is enough.

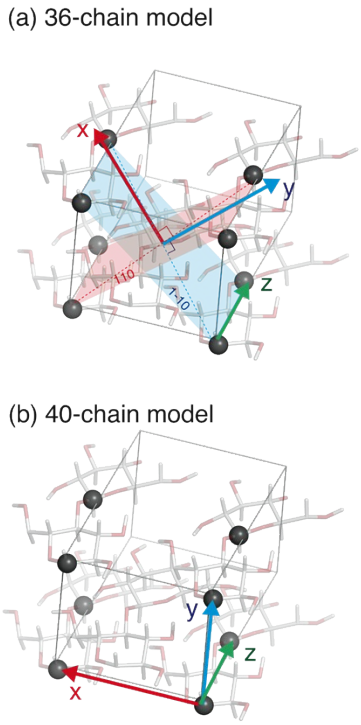


Figure S4. The definition of vectors for local strain tensor estimation. C1 atoms are shown in black spheres. The z-axis is along the fiber direction of cellulose crystal.

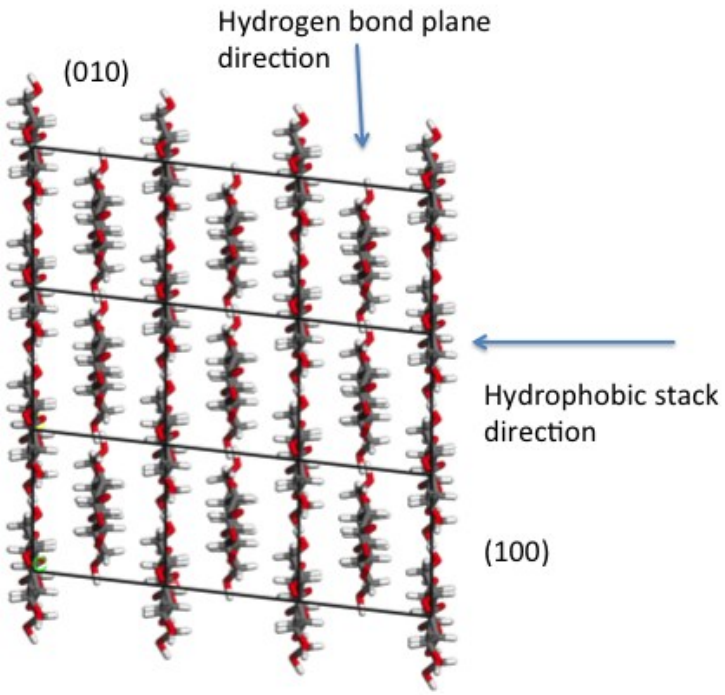
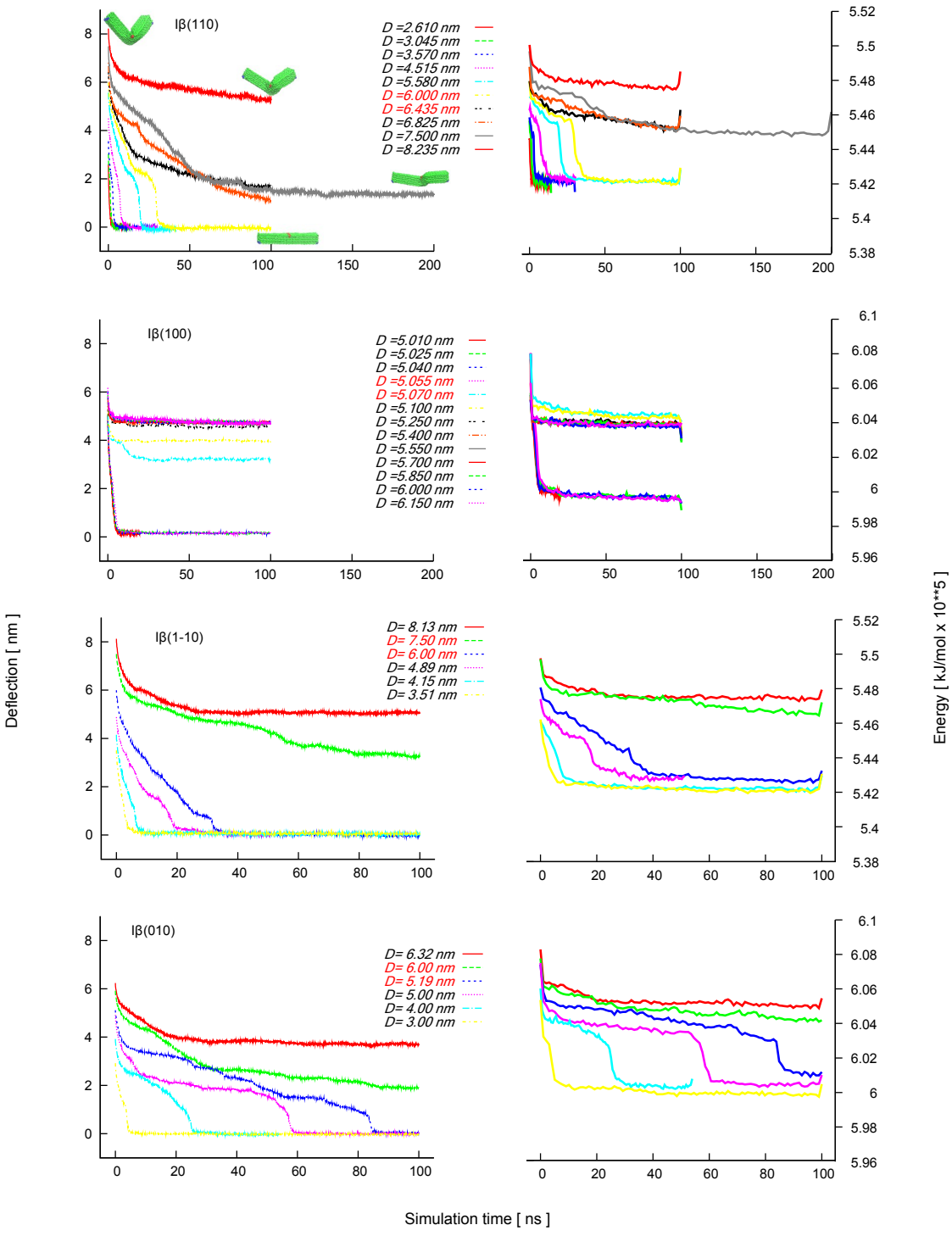


Figure S5. Anisotropy in the crystal structure of cellulose Iβ.

Along the *b* axis (in the vertical direction), cellulose forms sheets stabilized by hydrogen bonds, corresponding to the vertical piles of molecules in the drawing. Along the *a* axis (in the horizontal direction), the pile of sheets are stabilized by hydrophobic forces.



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43 **Figure S6.** The evolution of total energies and bending displacements during force releasing

44 processes.

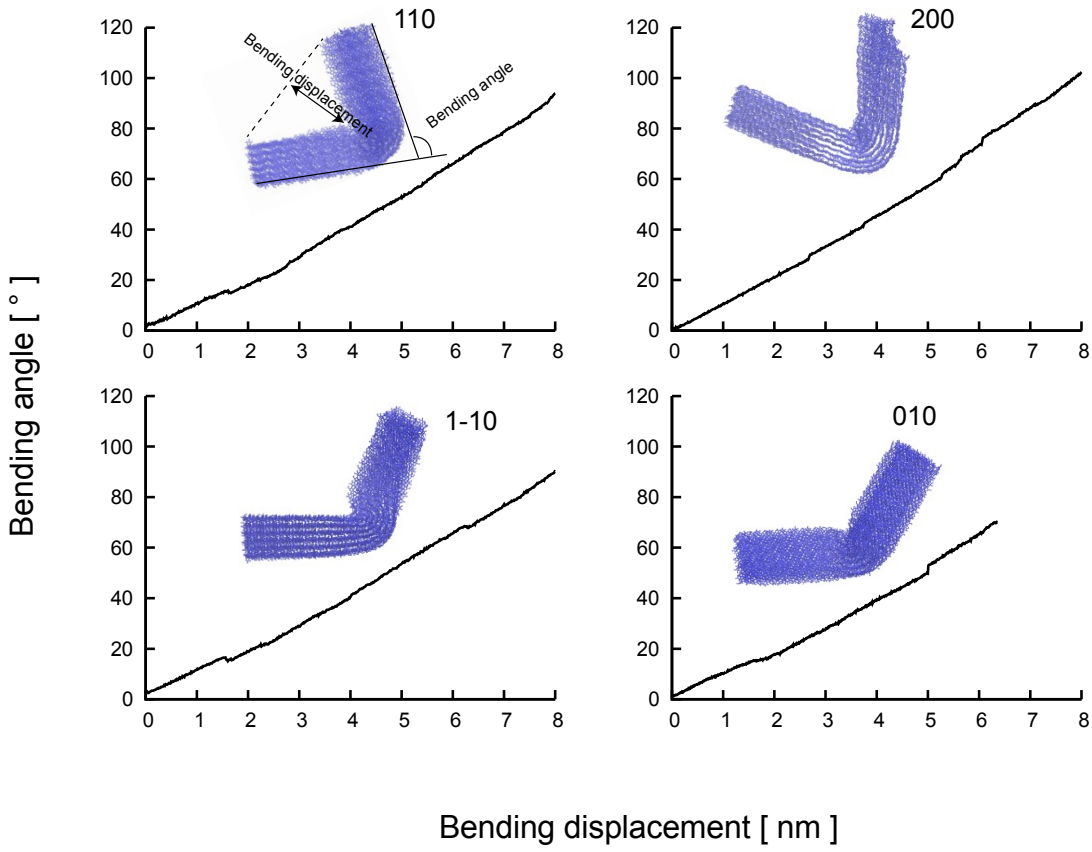

Figure S7. Relationship between bending deflections and the corresponding bending angles.