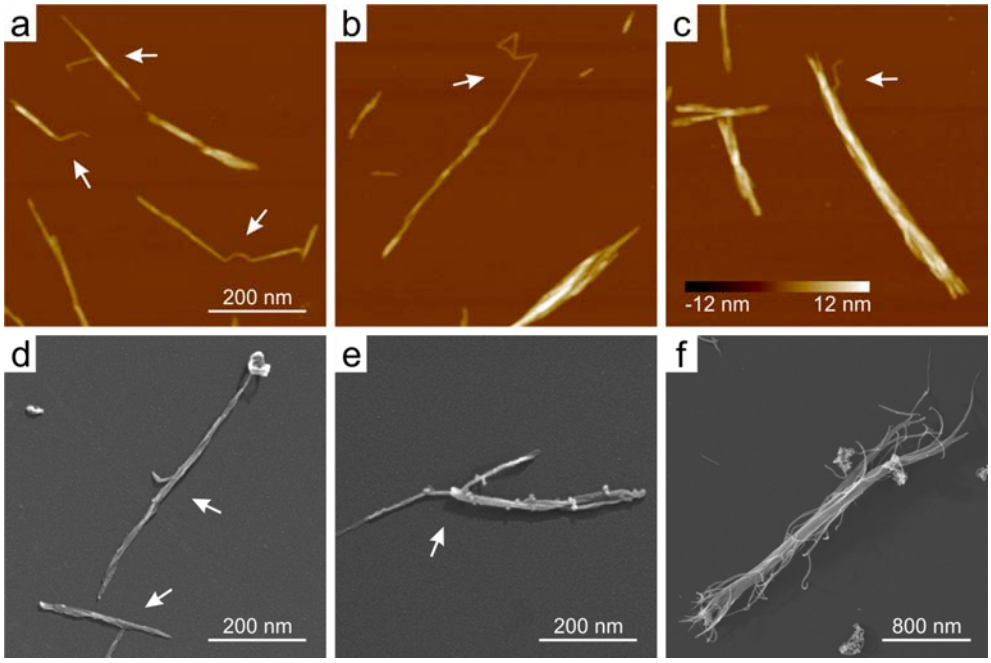
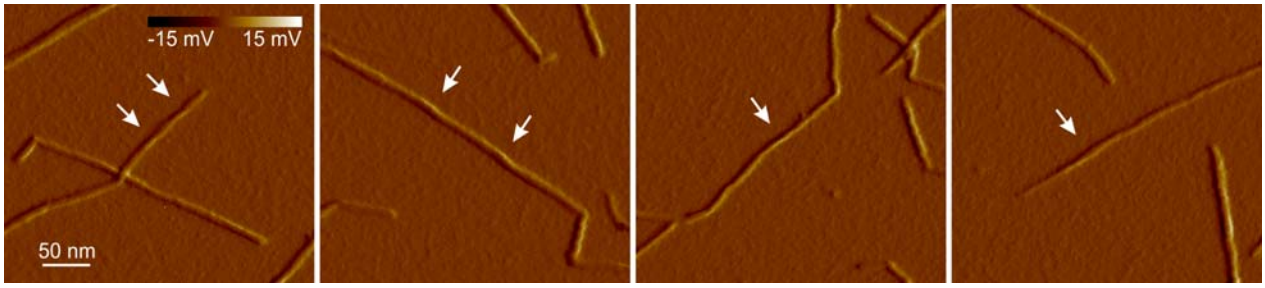
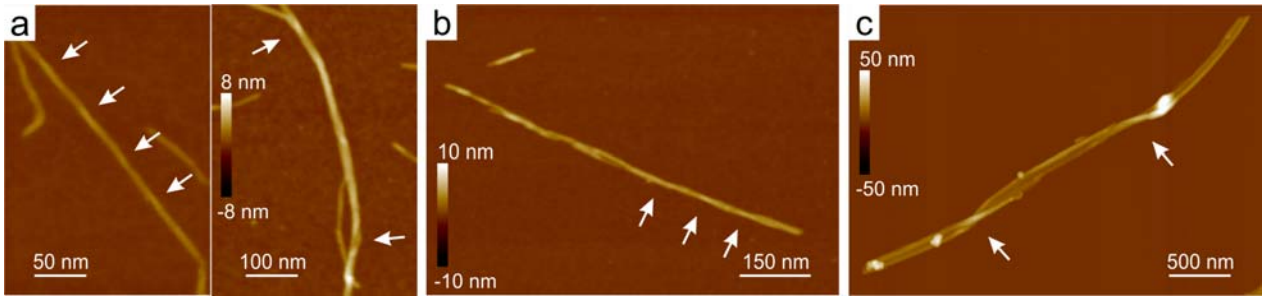


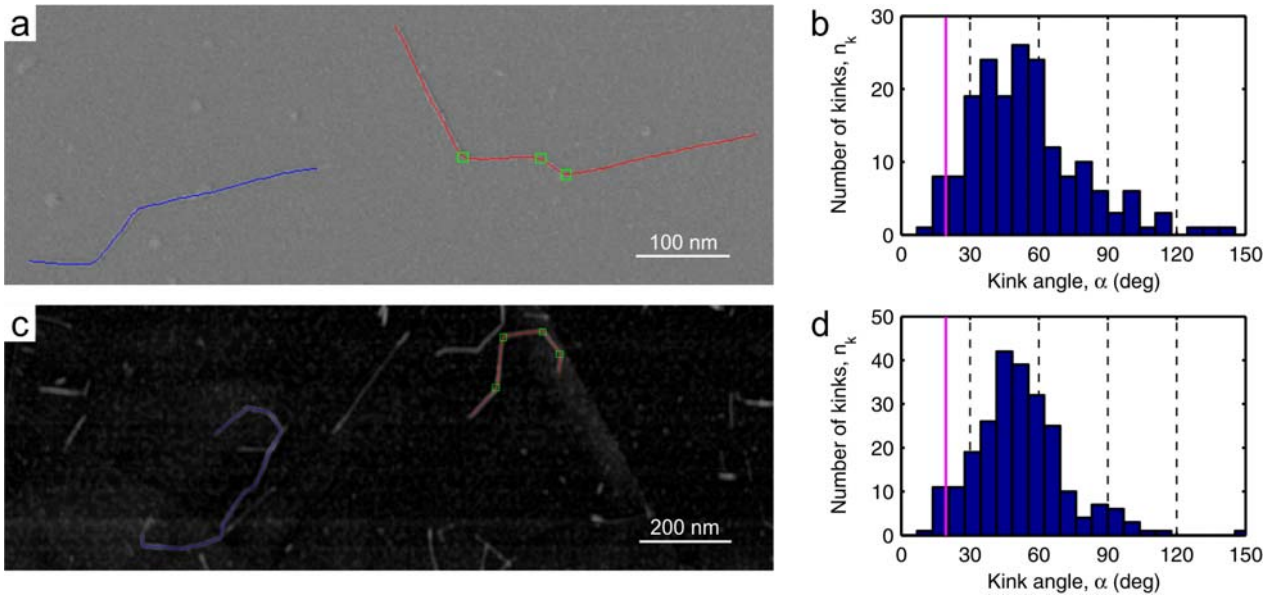

**Supplementary Figure 1.** Cellulose nanofibrils with kinks and/or splitting events that are pointed by white arrows in the images of the W-CNC, indicating that the hydrolysis was not fully completed. The images were acquired *via* (**a-c**) AFM and (**d-f**) Cryo-SEM. The scale bar in panel **a** and the color bar in panel **c** apply to all AFM images **a-c**.



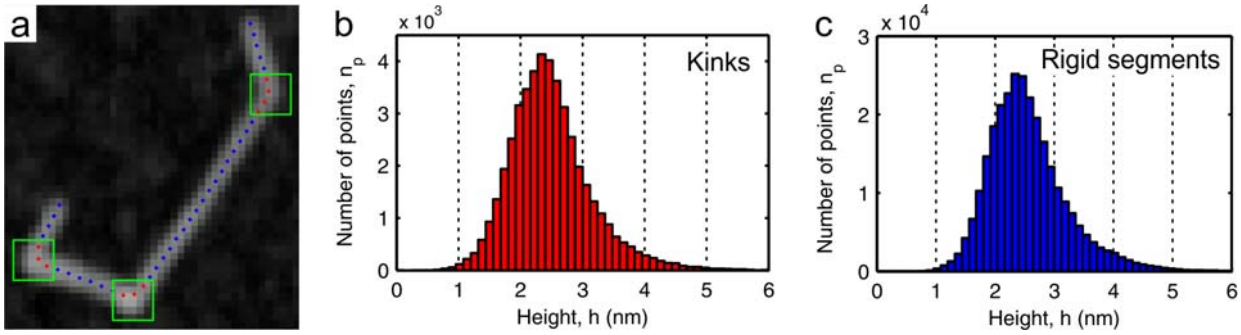

**Supplementary Figure 2.** Image data from the amplitude channel of AFM scanning showing W-CNF with observable right-handed twisting. The scale and color bars in the left panel apply to all AFM amplitude images.



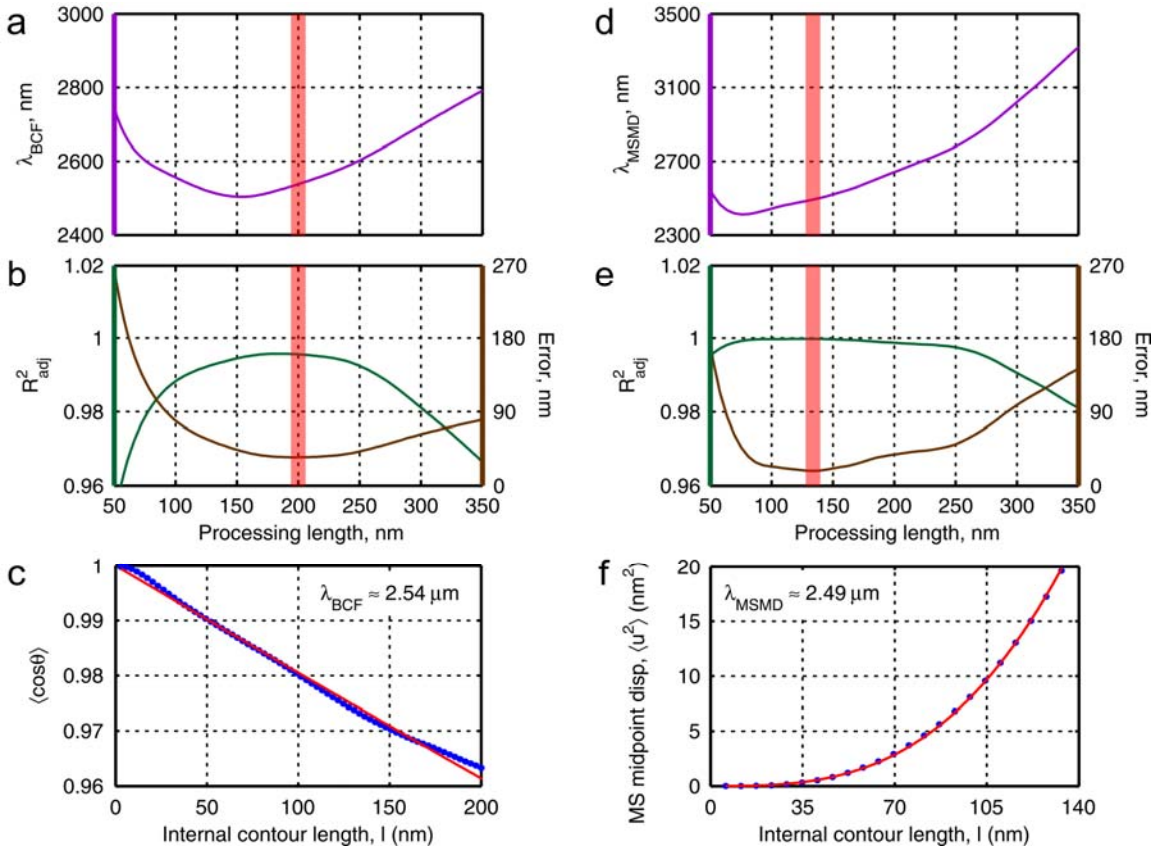

**Supplementary Figure 3.** Corresponding AFM height maps for the panels **a**, **d**, **g** of the Figure 2 in the main text, with the white arrows reproduced in the same positions.



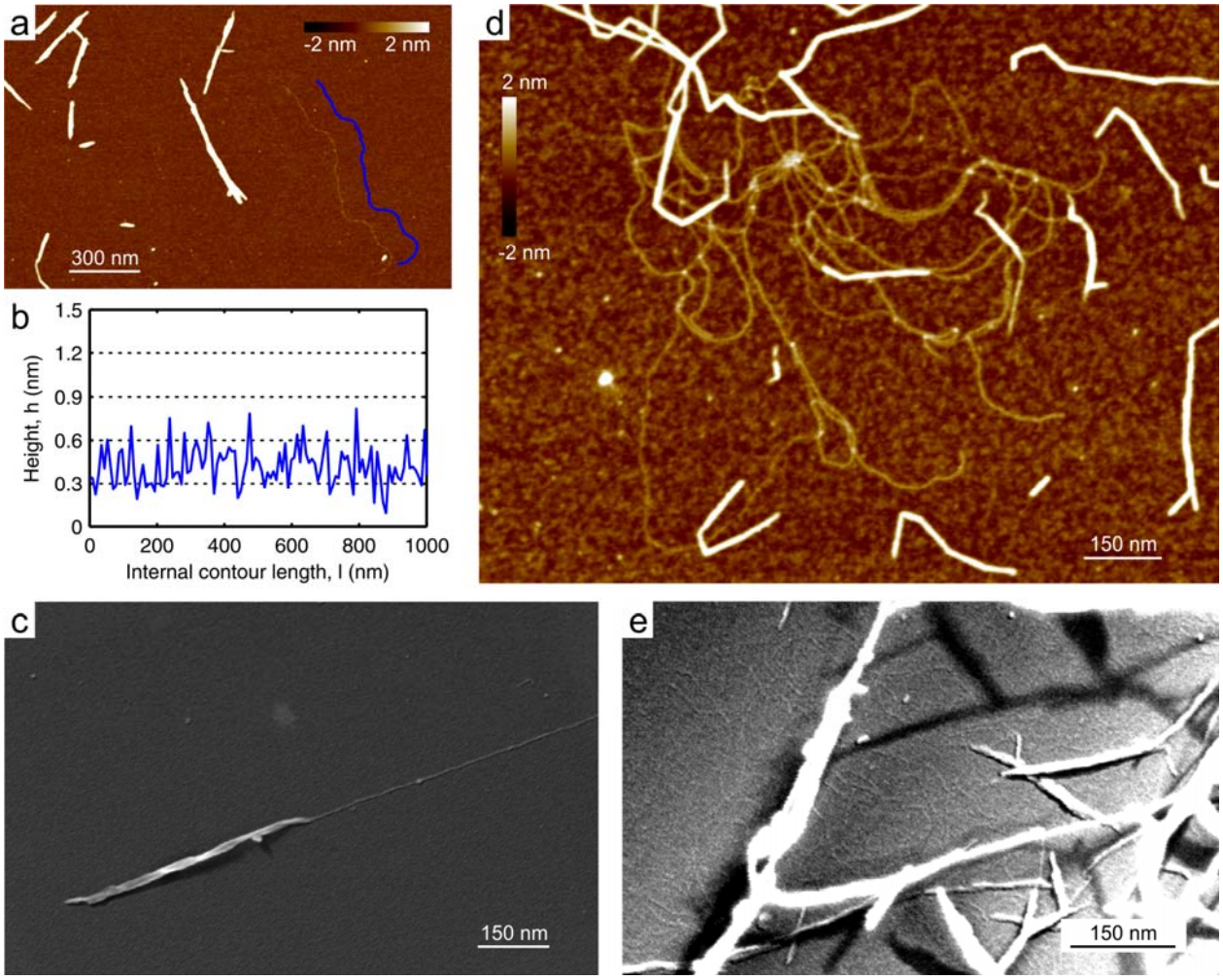

**Supplementary Figure 4.** An example of (**a**) Cryo-SEM image and (**c**) AFM on graphite with traced W-CNF and manually detected kinks. The corresponding kink angle distributions (**b** and **d**) for the corresponding samples (**a** and **c**).



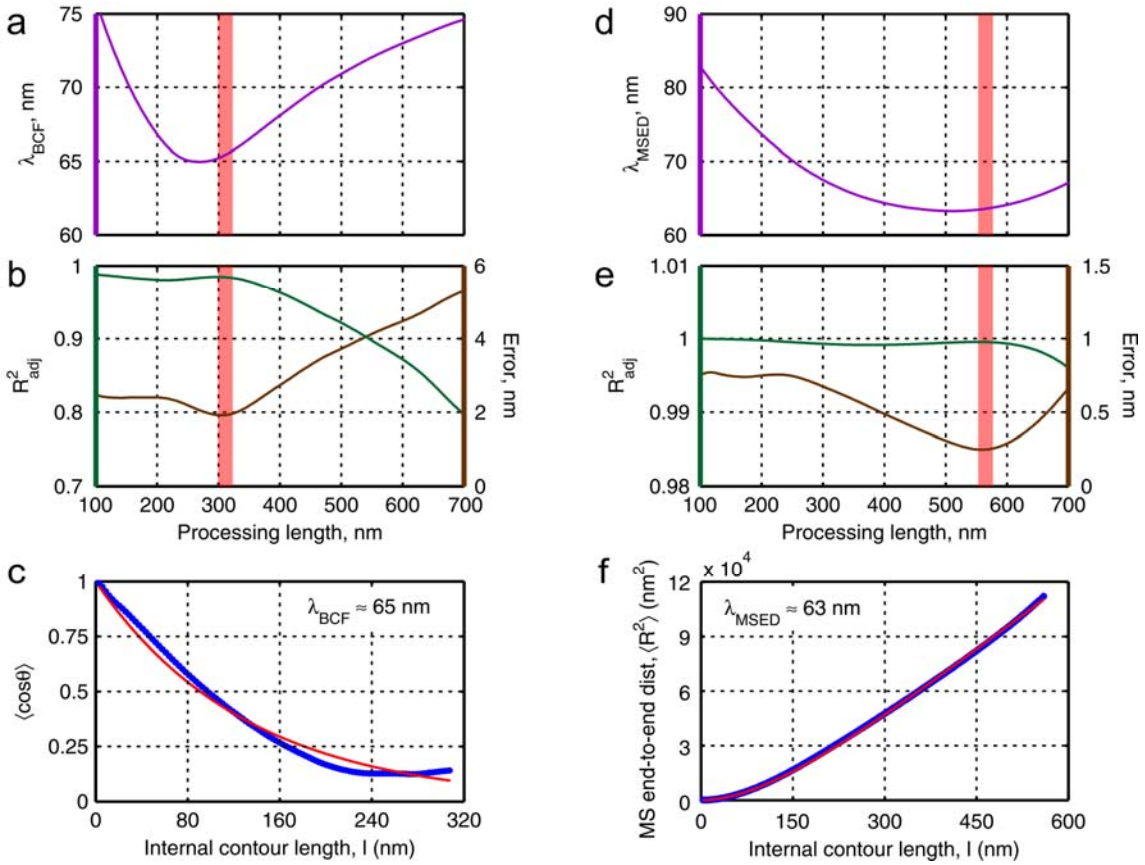

**Supplementary Figure 5.** A comparison between height distributions of contour points which are located in the vicinities of kinks and along stiff segments in W-CNF. (**a**) Example of the fibril from W-CNF and a corresponding contour with mask elements (green boxes), defining the kink areas. (**b**) The height distribution of all points inside regions of mask elements (red points). (**c**) The height distribution of all points outside regions of mask elements (blue points). Overall, these distributions do not differ from each other.




**Supplementary Figure 6.** The persistence length estimation of the rigid segments derived from W-CNF tracked contours *via* (**a-c**) the bond correlation function method, and (**d-f**) the mean-squared midpoint displacement method. (**a, d**) The persistence length values ($\lambda_{\mathrm{BCF}}$ and $\lambda_{\mathrm{MSMD}}$ respectively) plotted *versus* the processing length. (**b, e**) Adjusted coefficient of determination (goodness of fit) $R^2_{\mathrm{adj}}$ and fitting error *versus* the processing length. Horizontal axes from the panels **b** and **e** correspond also to the panels **a** and **d** respectively. The red vertical lines correspond to the processing length 200 nm for the bond correlation function and 135 nm for the mean-squared midpoint displacement methods, at which the $R^2_{\mathrm{adj}}$ is maximal and the fitting error is minimal. (**c, f**) The best fits for the bond correlation function and the mean-squared midpoint displacement respectively *versus* the internal contour length, truncated at the optimal processing length. The resulting persistence lengths are $\lambda_{\mathrm{BCF}} = 2.54$ µm and $\lambda_{\mathrm{MSMD}} = 2.49$ µm.




**Supplementary Figure 7.** (**a**) Observation of a single cellulose polymer chain in AFM image of the W-CNC. The blue line is a representation of the cellulose chain tracked contour, which is shifted for better visualization. (**b**) Height profile along the contour from the panel **a** with the average height value $\langle h \rangle \cong 0.45$ nm. (**c**) Observation of, possibly, a single cellulose polymer chain in Cryo-SEM image of the W-CNC. (**d**) AFM image of W-CNF, depicting single and 2×2 cellulose polymer chains forming a remarkable network with a well-defined entanglement center. (**e**) Cryo-SEM image with a high contrast of W-CNC, showing similar network in the background.




**Supplementary Figure 8.** The persistence length estimation of single cellulose polymer chain contours *via* (**a-c**) the bond correlation function method and (**d-f**) the mean-squared end-to-end distance method. (**a, d**) The persistence length values ($\lambda_{\mathrm{BCF}}$ and $\lambda_{\mathrm{MSED}}$ respectively) plotted *versus* the processing length. (**b, e**) Adjusted coefficient of determination (goodness of fit) $R^2_{\mathrm{adj}}$ and fitting error *versus* the processing length. Horizontal axes from the panels **b** and **e** correspond also to the panels **a** and **d** respectively. The red vertical lines correspond to the processing length 307 nm for the bond correlation function and 560 nm for the mean-squared end-to-end distance methods, at which the $R^2_{\mathrm{adj}}$ is maximal and the fitting error is minimal. (**c, f**) The best fits for the bond correlation function and the mean-squared end-to-end distance respectively *versus* the internal contour length, truncated at the optimal processing length. The resulting persistence lengths are $\lambda_{\mathrm{BCF}} = 65$ nm and $\lambda_{\mathrm{MSED}} = 63$ nm.