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Supporting Information

Crystallinity Improvements in Cellulose II after Multiple Posttreatment Cycles with a Dilute NaOH Solution

Yuki Kugo[1], Satoshi Nomura[1], Takuya Isono[2], Masashi Fujiwara[2],

Toshifumi Satoh[2,3], Hirofumi Tani[2], Tomoki Erata[2], and Kenji Tajima [2]*

[1]Graduate School of Chemical Sciences and Engineering, Hokkaido University, N13W8, Kita-ku, Sapporo 060-8628, Japan

[2]Faculty of Engineering, Hokkaido University, N13W8, Kita-ku, Sapporo 060-8628, Japan

[3]ICReDD List-PF, Hokkaido University, N21W10, Kita-ku, Sapporo 001-0021, Japan

*Corresponding Author: Email: ktajima@eng.hokudai.ac.jp

Results

Multicycle posttreatments

Fig. S1 shows the XRD profiles of mercerized initial cellulose II and the samples obtained by one to three cycles of posttreatment. The widths of peaks at 12°, 19°, and 21° were wider as the number of post-treatment cycles increased, indicating that the crystal sizes in the ($1\bar{1}0$), (110), and (020) planes increased as the number of posttreatment cycles increased.

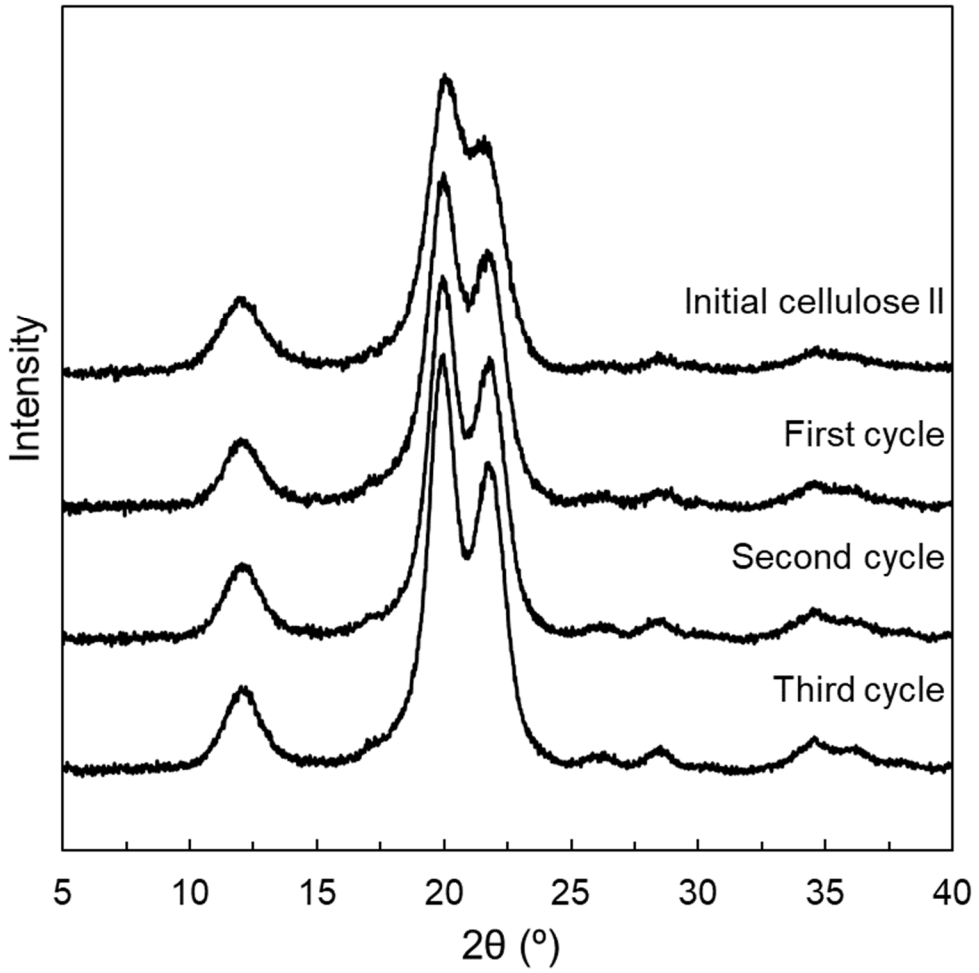

Fig. S1. XRD profiles of the mercerized initial cellulose II and the samples obtained by one to three cycles of posttreatment.

Highly efficient posttreatment method for improving crystallinity

Figs. S2(a) and S2(b) present the proportions of crystalline regions, accessible surfaces, and inaccessible surfaces/amorphous regions of samples prepared with total posttreatment times of 8 and 10 h, respectively. These results confirmed that when the posttreatment time for the first cycle was sufficiently long and the posttreatment time for the second cycle was relatively short, the crystallinity of the sample tended to increase with fewer posttreatment cycles for total treatment times of 8 and 10 h, similar to the multicycle posttreatment for a total treatment time for 6 h, as shown in Fig. 12.

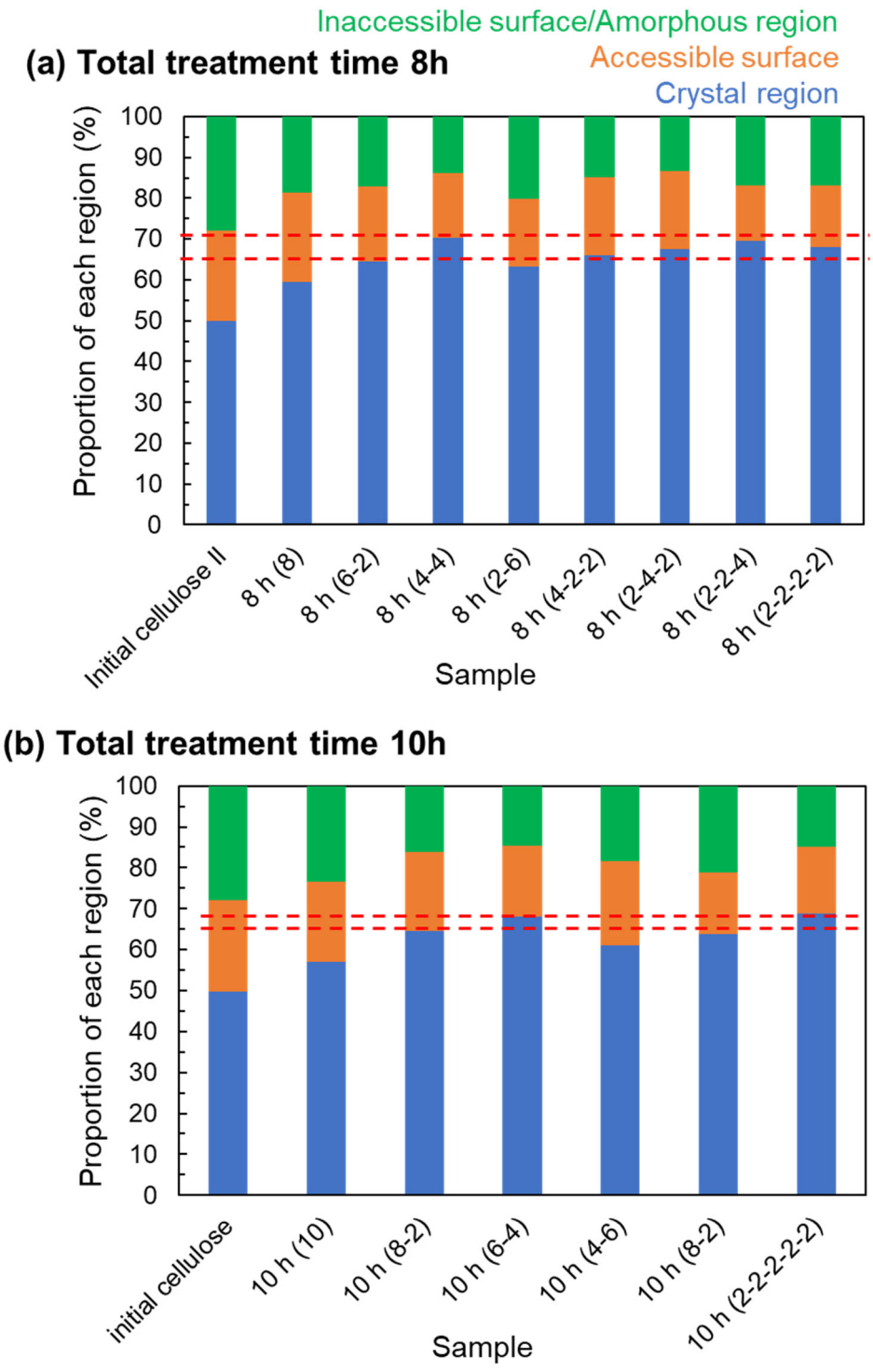

Fig. S2. Proportions of crystalline regions (blue bar), accessible surfaces (orange bar), and inaccessible surfaces/amorphous regions (green bar) evaluated from the solid-state ^{13}C CP/MAS NMR spectra of samples subjected to multicycle post-treatment with total posttreatment times of (a) 8 and (b) 10 h.