Supplementary Information

Facile preparation of cellulose IV_{II} using ionic liquids

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Figure S1. Curve fitting analyses of WAXS patterns for (a) original cellulose II via mercerization (crystallinity: 54%), (b) cellulose II regenerated from 5 mol% cellulose II/[C_2 mim][OAc] at 298 K (crystallinity: 33%), (c) cellulose I (crystallinity: 61%), and (d) cellulose IV_{II} regenerated from 5 mol% cellulose I/[C_1 mim][DMP] at 373 K (crystallinity: 31%). Black and blue lines are experimental and fitted patterns, respectively. Crystalline components are represented by a red line and an amorphous component is in a gray line. Each component is in the Gaussian function.



Figure S2. Curve fitting analyses of WAXS patterns of cellulose regenerated at 373 K from cellulose

II/[C₂mim][OAc] with various cellulose concentrations. Black and blue lines are experimental and fitted patterns, respectively. Crystalline components are represented by a red line and an amorphous component is in a gray line. Each component is in the Gaussian function. (a) 1 mo%, (b) 3 mol%, (c) 5 mol%, (d) 7.5 mol%, (e) 10 mol%, (f) 20 mol%, (g) 25 mol%, (h) 30 mol%, (i) 40 mol%, and (j) 50 mol%.



Figure S3. WAXS patterns of cellulose regenerated from the 5 mol% cellulose $I/[C_1mim][DMP]$ mixtures at 373 K. Upper: using dried cellulose I and $[C_1mim][DMP]$ as in Figure 7 (a). Bottom: using undried cellulose I (6.5 wt% H₂O) and $[C_1mim][DMP]$ (1.6 wt% H₂O). Their water contents were determined with the thermogravimetric analysis and the Karl Fischer titration, respectively.



Figure S4. ¹³C MAS solid-state NMR spectrum of the 5 mol% cellulose $I/[C_1mim][DMP]$ mixture measured at room temperature (299 K). High power dipolar decoupling was applied without CP. The spectrum was obtained with 16384 accumulations and 10 s recycle delay.



Figure S5. ¹³C CP/MAS solid-state NMR spectra of the 40 mol% cellulose I/[C_1 mim][DMP] mixture measured at (a) room temperature (299 K) and (b) 343 K. Black and blue lines are experimental and fitted patterns, respectively. The peak position of the tg (65.6 ppm) and gt (63.1 ppm) was fixed, the values of which were determined from the NMR spectra of cellulose I and cellulose II, respectively. Each component is in the Gaussian function.