

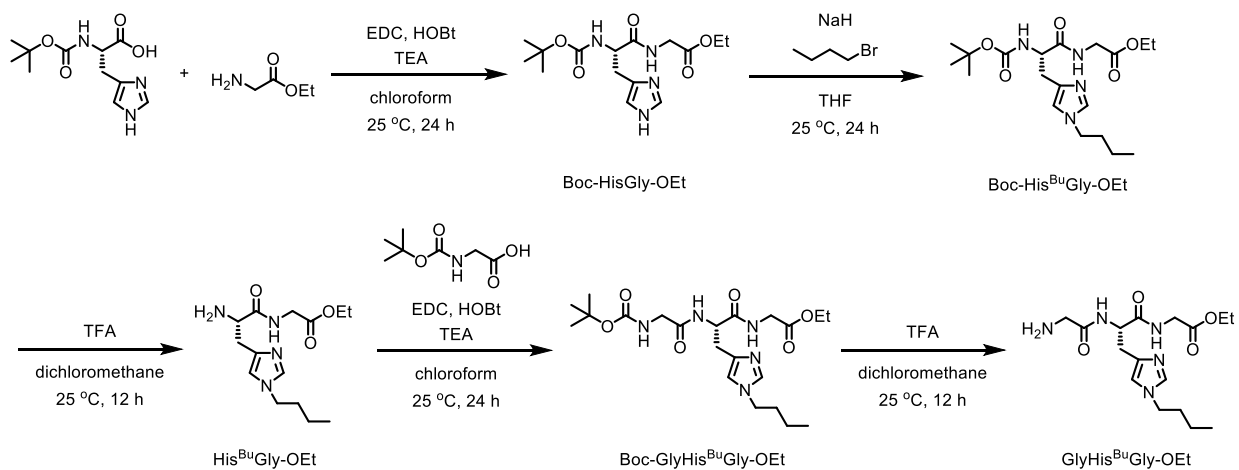
## Supporting Information

### Zwitterionic polypeptides: Chemoenzymatic synthesis and loosening function for cellulose crystals

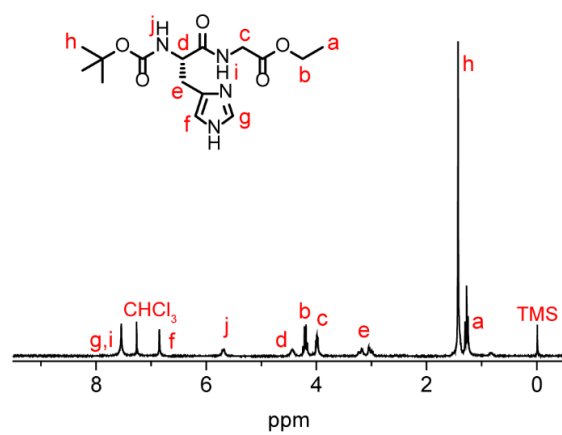
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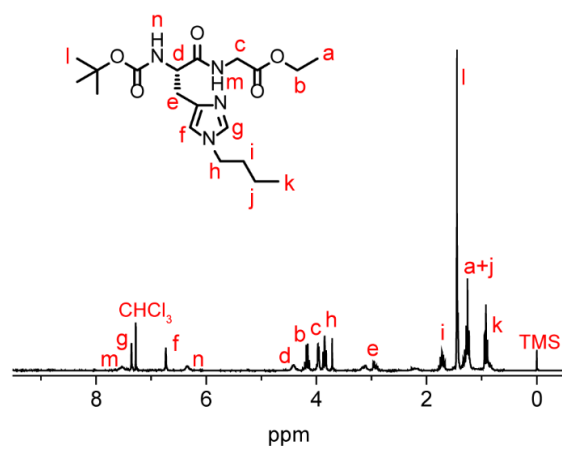
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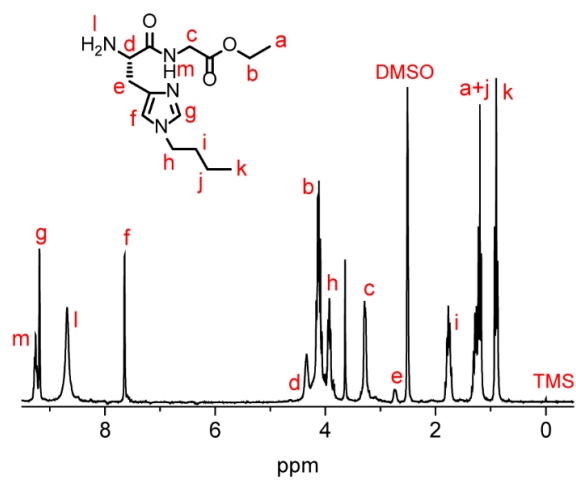
**Scheme S1.** Synthesis of GlyHis<sup>Bu</sup>Gly-OEt monomer.



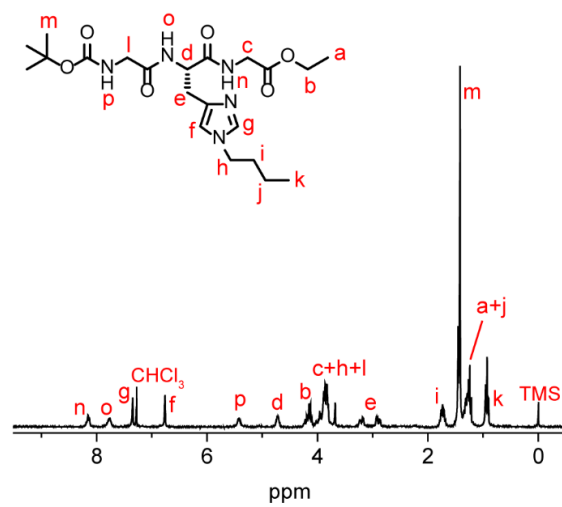
**Figure S1.** <sup>1</sup>H NMR spectrum of Boc-HisGly-OEt in CDCl<sub>3</sub>.



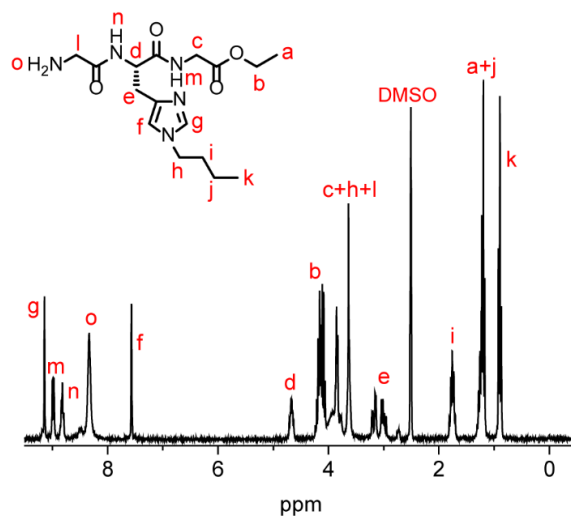
**Figure S2.**  $^1\text{H}$  NMR spectrum of Boc-His<sup>Bu</sup>Gly-OEt in  $\text{CDCl}_3$ .



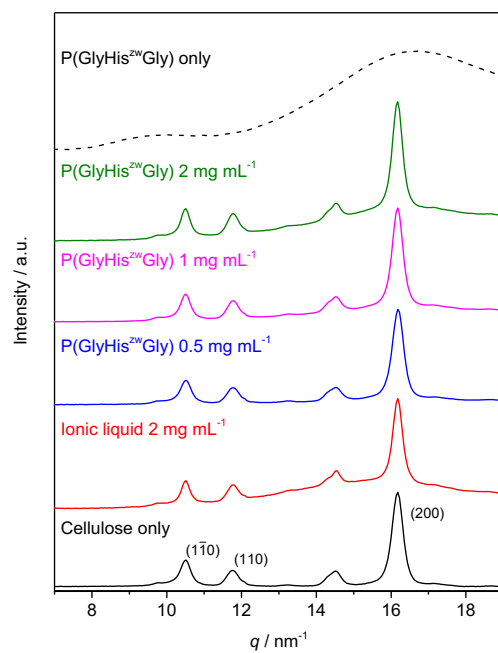
**Figure S3.** <sup>1</sup>H NMR spectrum of His<sup>Bu</sup>Gly-OEt in DMSO-*d*<sub>6</sub>.



**Figure S4.** <sup>1</sup>H NMR spectrum of Boc-GlyHis<sup>Bu</sup>Gly-OEt in CDCl<sub>3</sub>.

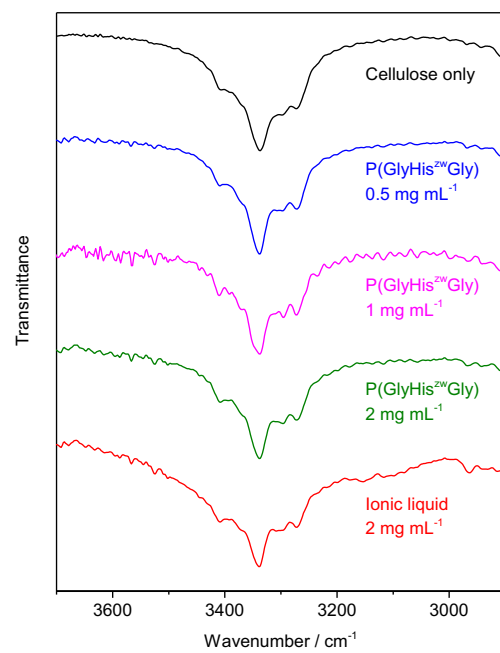


**Figure S5.** <sup>1</sup>H NMR spectrum of GlyHis<sup>Bu</sup>Gly-OEt in DMSO-*d*<sub>6</sub>.



**Figure S6.** WAXD 1D profiles of cellulose nanocrystals after treatment with P(GlyHis<sup>zw</sup>Gly) and ionic liquid BMImAc for 1 h (solid lines) and P(GlyHis<sup>zw</sup>Gly) powder sample (broken line).





**Figure S7.** IR spectra of cellulose nanocrystals after treatment with P(GlyHis<sup>zw</sup>Gly) and ionic liquid BMIImAc for 1 h.

## **Captions of Movies**

**Movie 1.** Morphological changes in the cell wall structures in BY-2 cells observed by HS-AFM after treatment with P(GlyHiszwGly) ( $0.4 \text{ mg mL}^{-1}$ ). Movie version of Figure 7a.

**Movie 2.** Morphological changes in the cell wall structures in BY-2 cells observed by HS-AFM after treatment with P(GlyHiszwGly) ( $0.4 \text{ mg mL}^{-1}$ ). Another example showing the dissociation of cellulose microfibrils.

**Movie 3.** Morphological changes in the cell wall structures in BY-2 cells observed by HS-AFM after treatment with P(GlyHiszwGly) ( $0.8 \text{ mg mL}^{-1}$ ). Movie version of Figure 7b.