Supporting information

Moldable crystalline α-chitin hydrogel with toughness and transparency toward ocular applications

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Figure S1. Typical collapsed N-acetylated chitosan hydrogel during preparation according to the original protocol. The chitosan solution gelled during mixing with acetylating agent, acetic acid anhydride (Ac₂O), before pouring into molds.
Figure S2. Preparation protocol of N-acetylated chitosan hydrogel developed in this study.
Figure S3. Preparation protocol of N-acetylated chitosan hydrogel beads developed in this study.
**Figure S4.** Representative stress-strain (S-S) curves of N-acetylated chitosan hydrogels prepared by using methanol (same as "2.3 wt%" in Fig. 3a) and ethanol.

**Figure S5.** Unmodified electron micrograph of Fig. 5. Cross-sectional cryo-SEM image of N-acetylated chitosan gel based on high DP chitosan (DP: 7600).
Figure S6. Cross-sectional cryo-SEM image of N-acetylated chitosan gel based on low DP chitosan (a) just after gelation and (b) after subsequent washing with water.

Figure S7. Wide angle X-ray diffraction pattern of the N-acetylated chitosan hydrogel based on high DP chitosan (DP: 7600) (the same specimen as in Fig. 2f).
**Figure S8.** Unmodified optical micrograph of Fig. 8a. Optical micrograph of micro-beads based on N-acetylated chitosan hydrogel (DP: 7600). Scale bar represents 1 mm.