Supporting Information


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The CHC hybrid hydrogels were synthesized by combining Genipin-crosslinked CHC together with acid-hydrolyzed TEOS in different ratios of composition. The synthetic procedure was described as follows. In the first step, the synthesis of genipin-crosslinked CHC network is originated from nucleophilic attack by amino group of CHC toward the olefinic carbon atom at C-3 of deoxyloganin aglycone followed by the opening of the dihydropyran ring to form heterocyclic amine (Figure S1(a)). The conformation of the network segments of genipin-crosslinked CHC gels depends on the added amount of genipin. In the second step (Figure S1(b)), after genipin-crosslinked CHC network was formed, acid-hydrolyzed TEOS was added to complete sol-gel reaction.
Step 1: Genipin-crosslinked CHC network

Step 2: Acid-hydrolyzed TEOS

Figure S1 Structural illustration of (a) Genipin-crosslinked CHC network (b) acid-hydrolyzed tetraethoxysilane (TEOS) by mixing TEOS with ethanol and HCl acidified H₂O.
**Figure S2** Mechanical enhancement to the hybrid hydrogels can then be expected with higher TEOS content.
Figure S3 TG curves for pure CHC and pure TEOS.