Supplementary Materials for

## Biomineralization-inspired Anti-caries Strategy Based on Multifunctional Nanogels as Mineral Feedstock Carriers



Figure S1 TEM observation of the degraded Cap-loaded PNPDC/PGA nanogels at 25°C

(A-C) and  $37^{\circ}$ C (D-F) at different time. At the time of 8 h, the nanogels have partially degraded, and the shape of nanoparticles is still clear.



**Figure S2** SEM observe of *S. mutans* UA159 biofilm under the action of Cap-loaded PNPDC/PGA nanogels (50mg/ml). The bacteria are in contact with the surrounding gel-like materials, and swelled, deformed (a-b), and lysed into fragments (c-d).



**Figure S3** SEM micrographs of dentin surface remineralization with Cap-loaded PNPDC/PGA nanogels for 7 d. The peony-patterned material was connected into networks and sheets, and completely blocked the dentin tubules.



**Figure S4** Unstained TEM images of recombinant type I collagen. The collagen fibrils show relatively clear light and dark striped stripes.