Electronic Supplementary Information



Figure S1. Illustrative flowchart of hydrothermal synthesis procedure



Figure S2. XRD pattern (A) and FTIR spectrum (B) of the product synthesized at pH=5.4. The asterisks in (A) mark the diffraction peaks from CaHPO₄. The vibrational bands at 866 cm⁻¹ and 1635 cm⁻¹ in (B) are assigned to HPO₄ group and bending mode of water respectively.



Figure S3. Bright-field TEM and HRTEM images of the nanoparticles prepared at varying pH. (A1) pH = 6.6; (B1) pH = 8; (C1) pH = 10; and (D1) pH = 12. Insets in the HRTEM images are the corresponding fast Fourier transform (FFT) patterns. (A2) shows the typical HA nanorod. Three typical facets (001), $(1\overline{10})$ and (010) of CAp nanocrystals are shown in (B2), (C2) and (D2) respectively.



Figure S4. HRTEM image of the CAp nanocrystal with elongated hexagonal shape and corresponding Fast Fourier Transform (FFT). The beam direction is along [001].



Figure S5. TGA curves of CAp with different C/P atomic ratios in starting solution. (A) CAp with C/P = 1/3; (B) CAp with C/P = 2/3; and (C) CAp with C/P = 1.



Figure S6. XRD patterns of the samples with varying carbonate content. The black solid balls mark the CaHPO₄ impurity phase present in the sample free of carbonate.



Figure S7. FTIR spectra of CAp prepared at different temperatures for 12 h (C/P =2/3,

pH = 10).