

Supplementary Information

Tough hydrogel/hydroxyapatite bone-like composite fabricated *in situ* by electrophoresis approach

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Experimental

Swelling experiments

Swelling experiments were performed by immersing hydrogels in phosphate-buffered saline (PBS) at 37°C to reach swelling equilibrium. The swelling ratio (SR) was calculated by the following equation, $SR = (W_s - W_d) / W_d \times 100\%$, where W_s and W_d are the weights of the swollen hydrogel and the corresponding dried hydrogel, respectively. The average of three measurements was taken for each sample.

Results and discussion

Supporting Figures

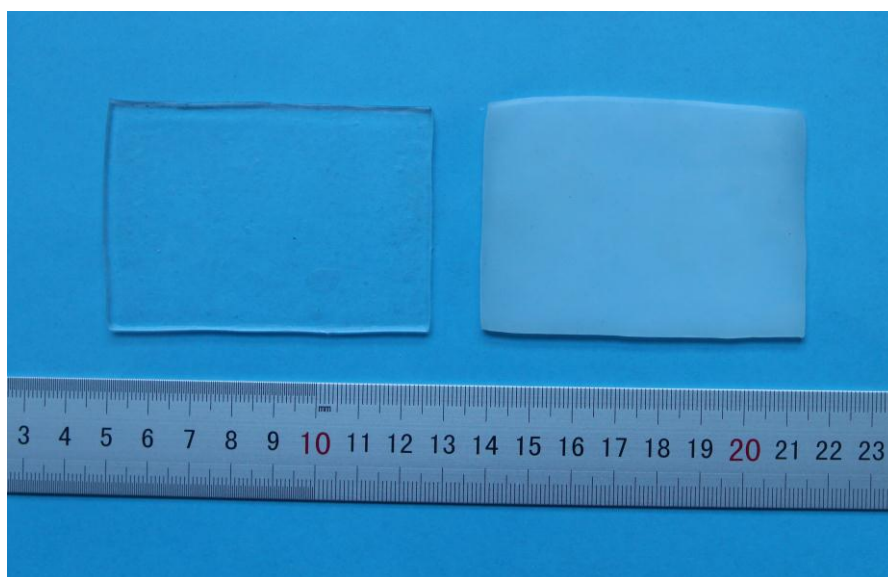


Figure S1. Photographs of the typical as-synthesized PAAm hydrogel (left) and hydrogel-HAp composite (right).

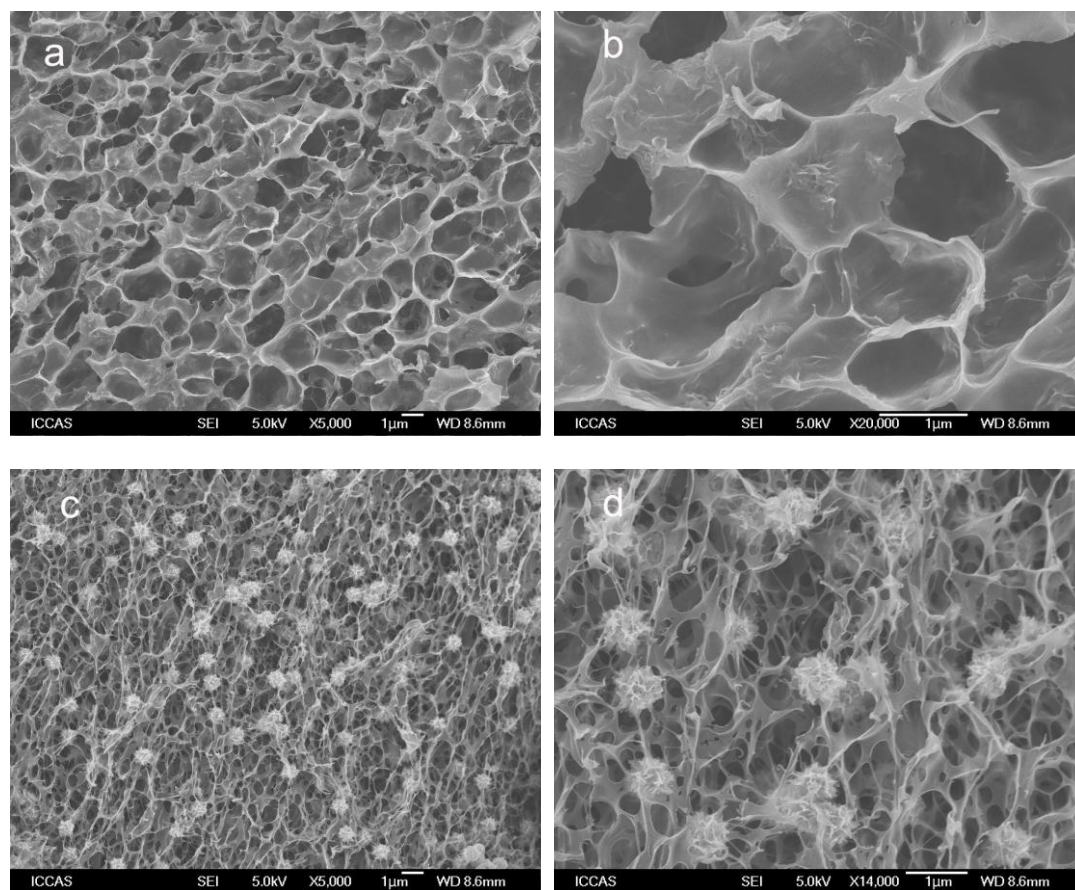


Figure S2. SEM images showing the morphologies of calcium phosphate in the hydrogel composites prepared from the as-synthesized hydrogels swollen in the CaCl_2 solutions with different pH values. (a, b) pH = 7.0, (c, d) pH = 8.0. The hydrogel composites were made from the as-synthesized hydrogels with 90 wt% water content.

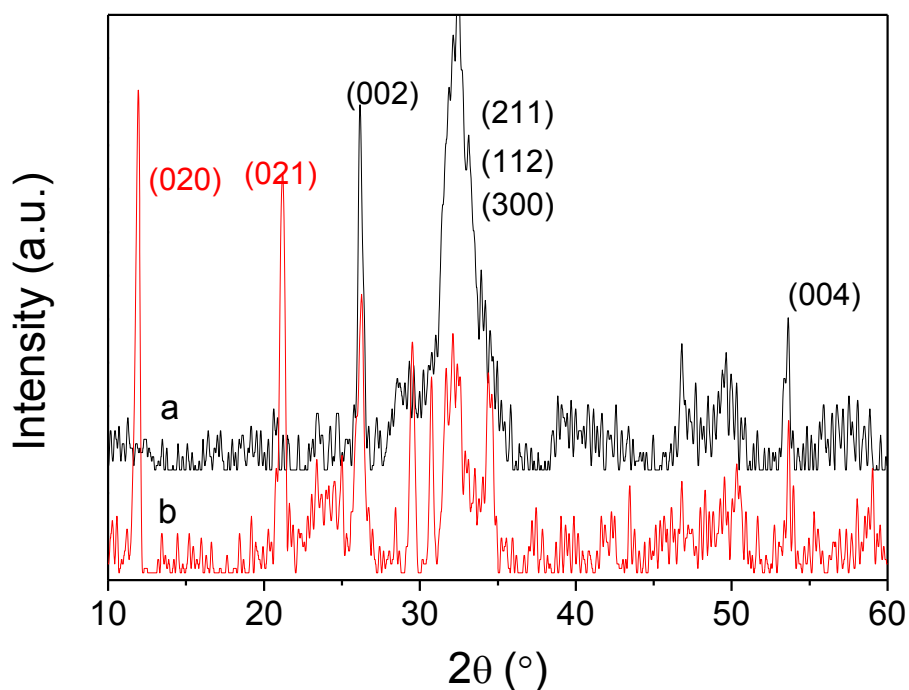


Figure S3. XRD patterns of the hydrogel composites prepared from the as-synthesized hydrogels swollen in the CaCl_2 solutions with different pH values. To distinguish the patterns easily, the amorphous regions have been eliminated. (a) pH = 8.0, the inorganic component in the composite is HAp, (b) pH = 7.0, the inorganic component in the composite is an admixture composed of HAp and DCPD. Peaks marked (002), (211), (112), (300), (004) are for HAp, and peaks marked (020), (021) are for DCPD.

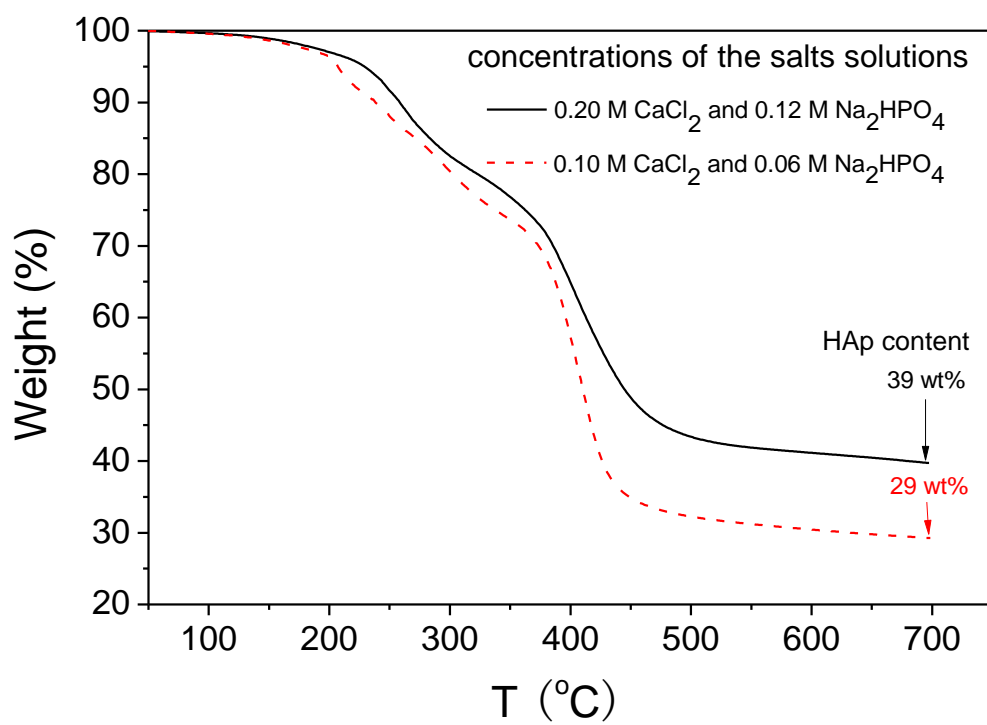


Figure S4. TGA curves of the hydrogel-HAp composites fabricated by using salts solutions with different concentrations.



Figure S5. Photographs of the mineralized products after the same mineralizing time based on the as-synthesized hydrogel with different water contents. The water contents of the as-synthesized hydrogels are 80 wt% (left), 85 wt% (middle) and 90 wt% (right), respectively.

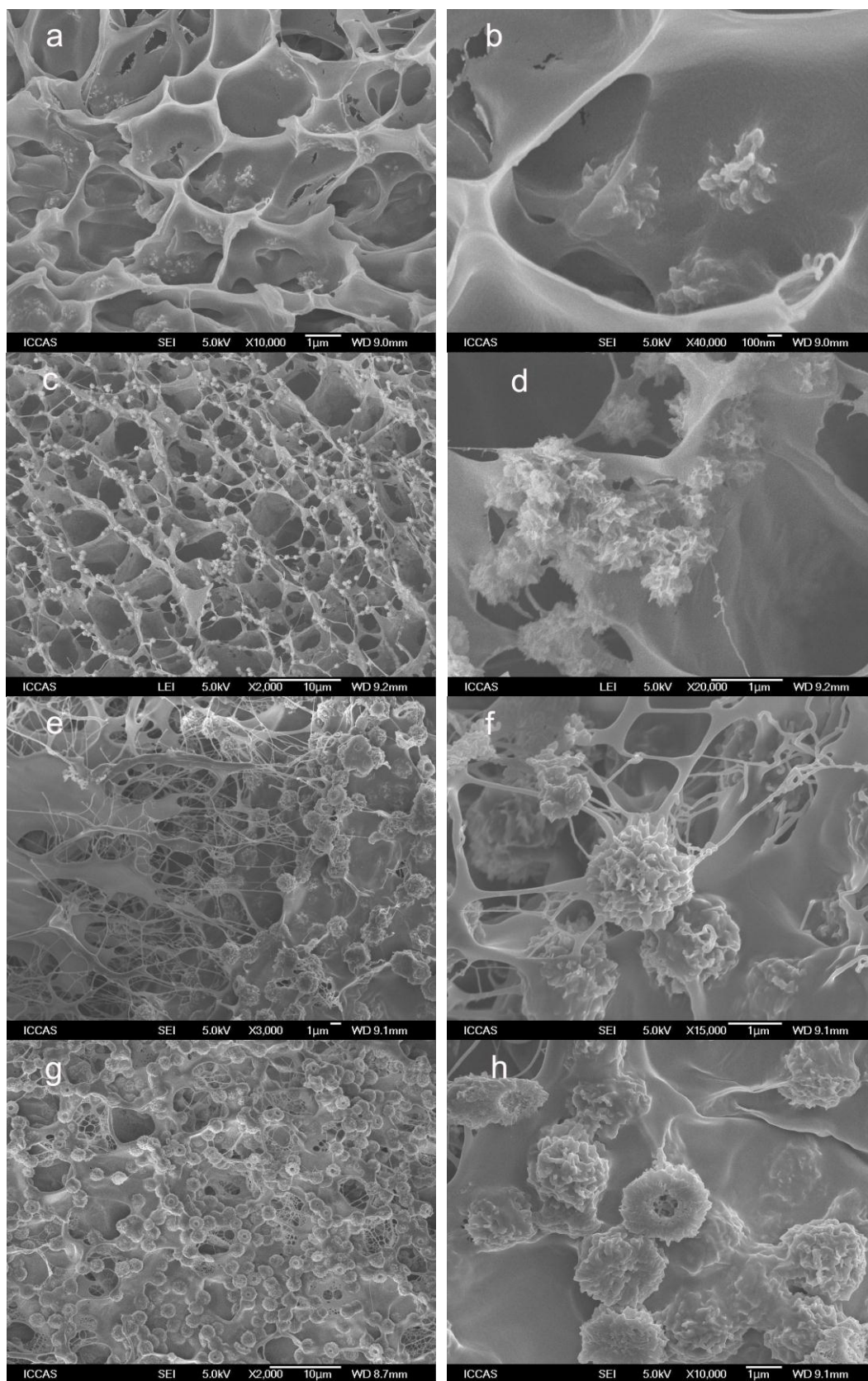


Figure S6. SEM images of the hydrogel-HAP composites swollen to different water contents. (a, b) 80 wt%; (c, d) 85 wt%; (e-h) 95 wt%.

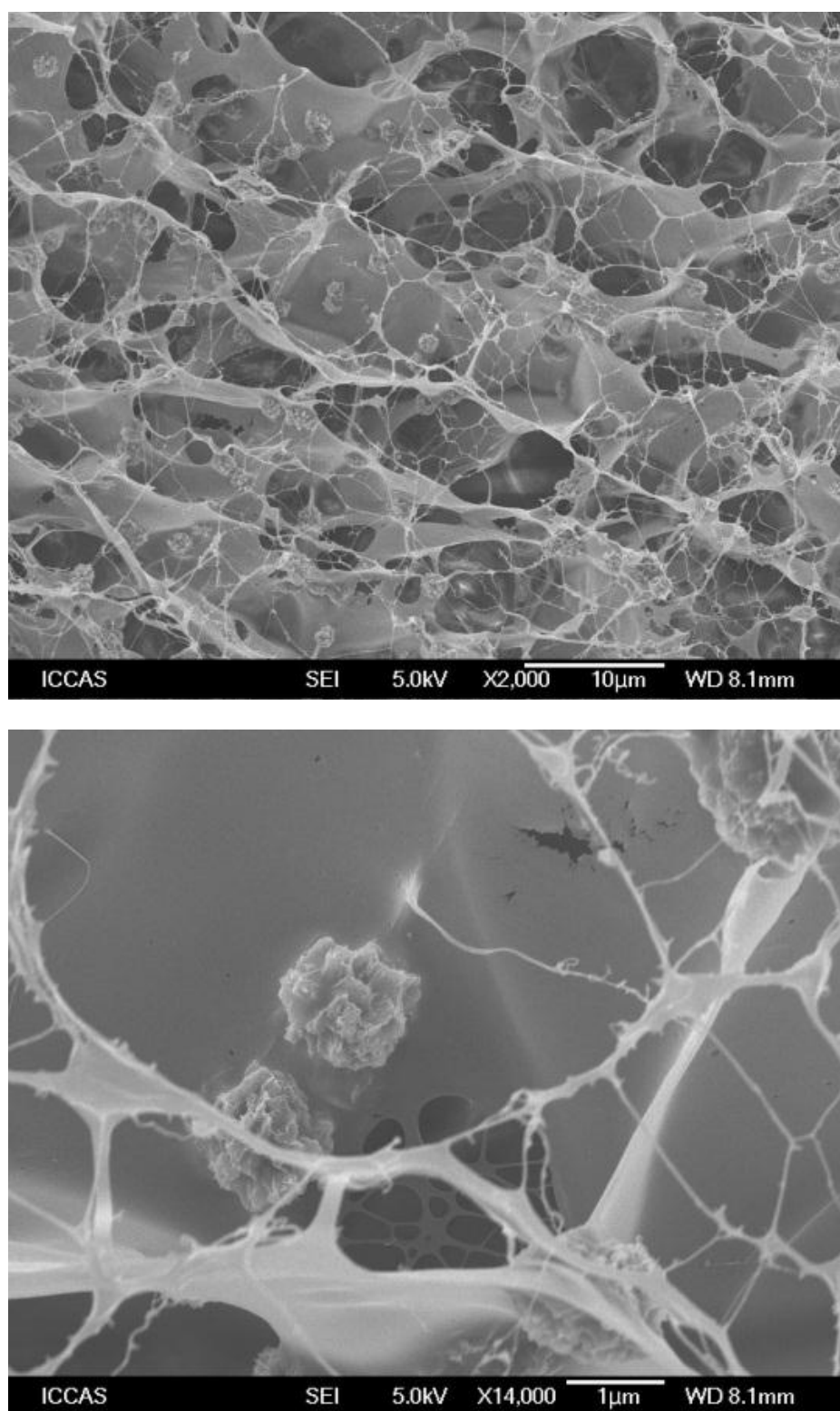


Figure S7. SEM images of the hydrogel-HAp composite before the compression test. (For the convenience of observation, the gel sample was equilibrium swollen). The hydrogel-HAp composite was made from the as-synthesized hydrogel ($C_M = 7$ M) with 90 wt% water content and 0.1 M CaCl_2 solution.

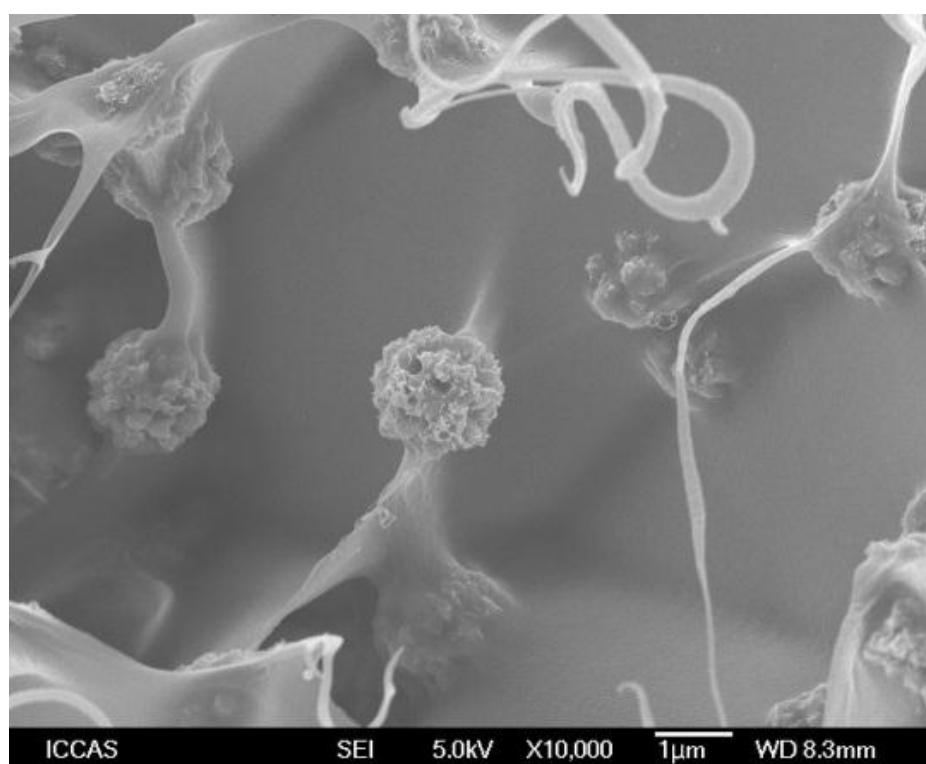
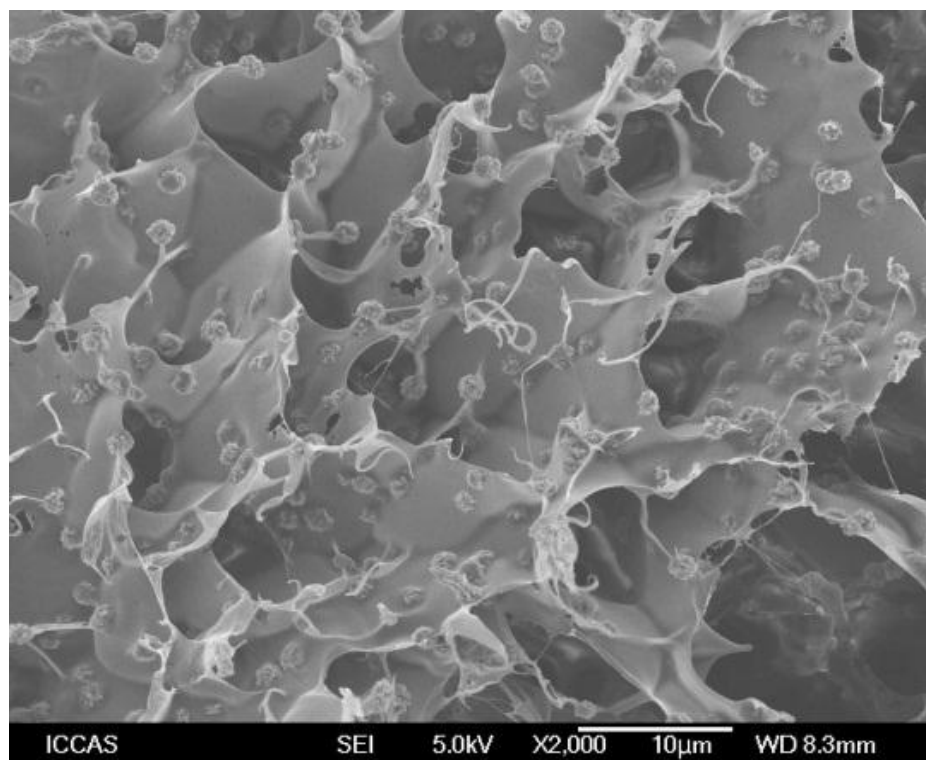


Figure S8. SEM images of hydrogel-HAp composite after compression test. (The specimen used for the compression test and the specimen in Figure S7 were cut out from the same big gel piece ($C_M = 7 M$)).

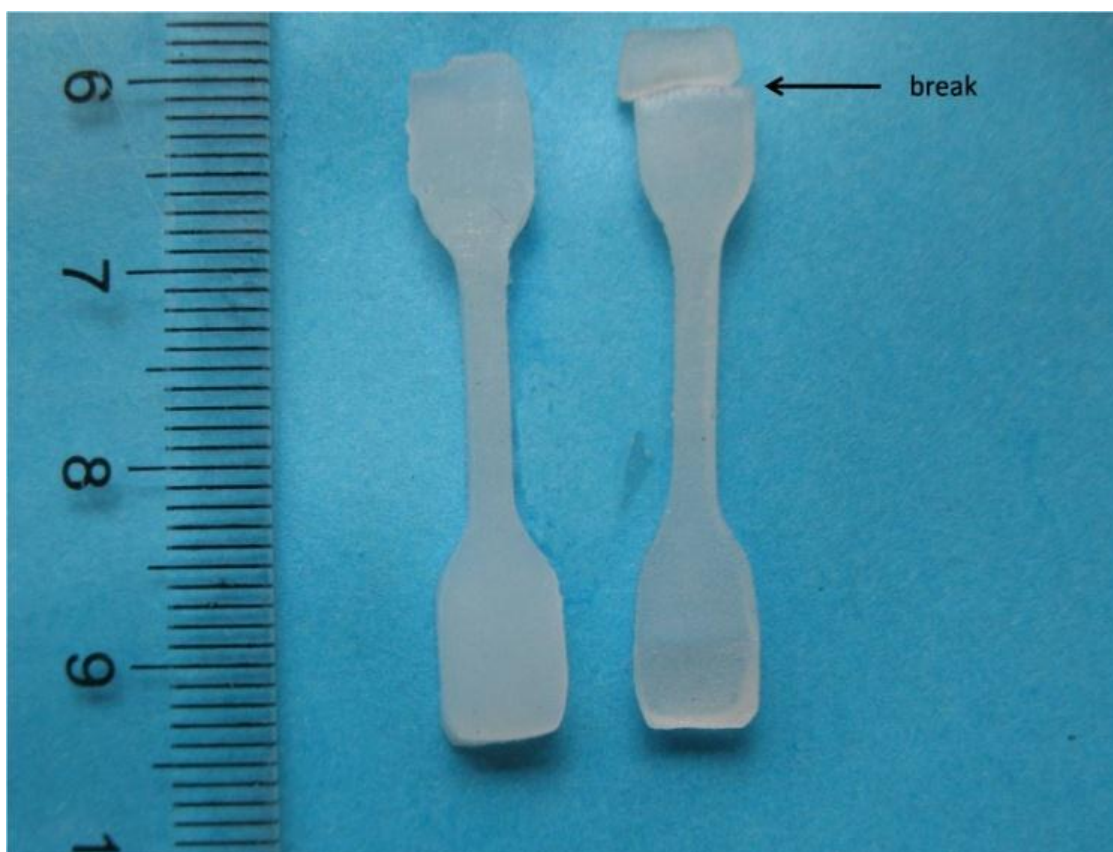


Figure S9. Photographs of the hydrogel-HAp composite specimens with 25 wt% polymer content. The as-prepared specimen (left) and the specimen after the tensile test (right) were cut out from the same big hydrogel piece ($C_M = 7$ M). The specimen on the right side has been elongated to a strain of 2200 %, it did not break in the gauge center part, whereas broke at the gripped end part. It can be seen clearly that the tested specimen has almost recovered back to its original length in comparison with the original specimen (left).

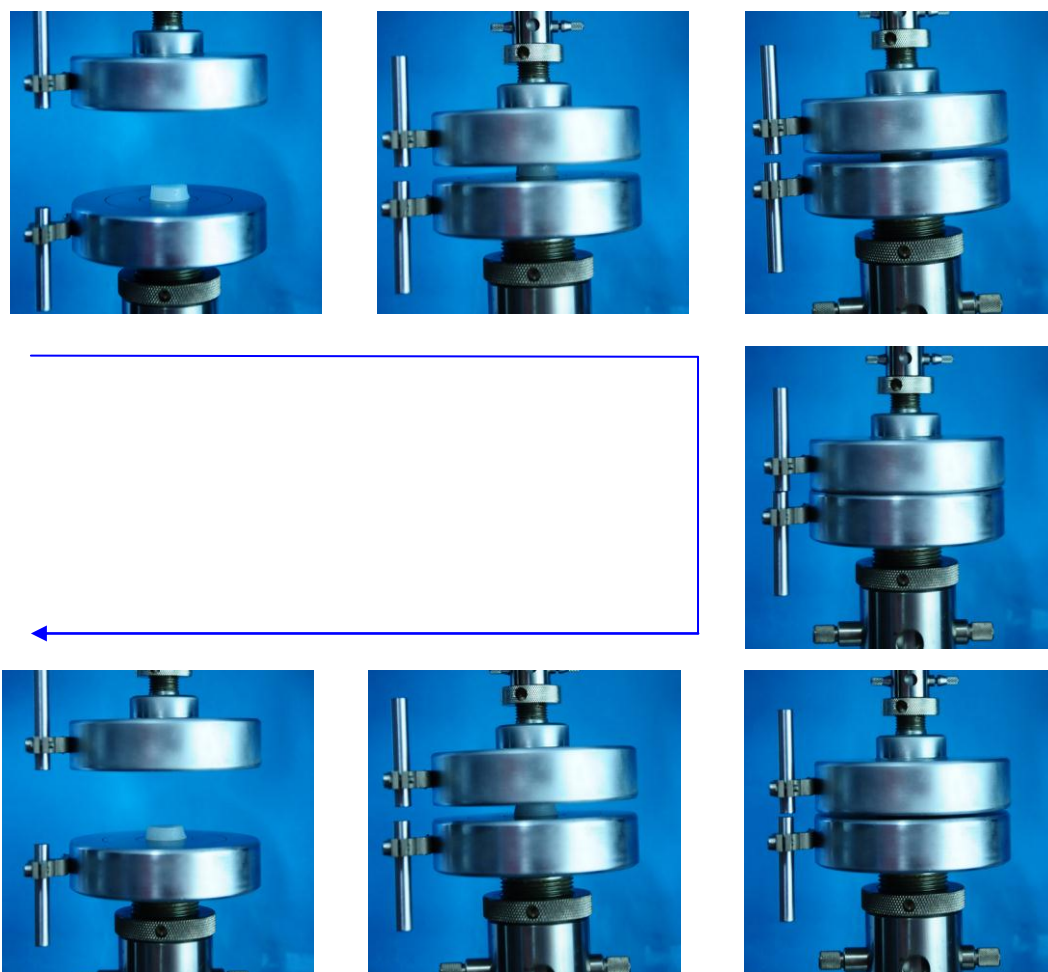


Figure S10. Photographs of a typical hydrogel-HAp composite during the compression test. The hydrogel-HAp composite could recover back to its original shape even after being compressed to a strain of 0.98.

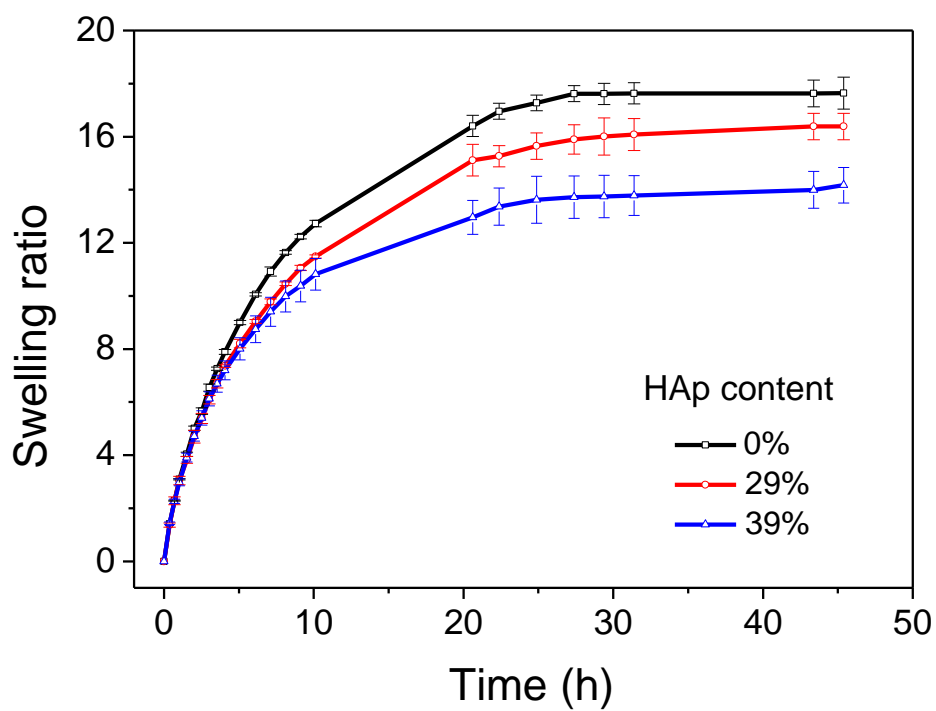


Figure S11. Swelling curves of the as-synthesized hydrogel and the hydrogel-HAp composites with different contents of HAp. The hydrogel-HAp composites were made from the as-synthesized hydrogels ($C_M = 7M$) with 80 wt% water content.