Title:

Construction of Versatile Multilayered Composite Nanoparticles from a Customized Nanogel Template

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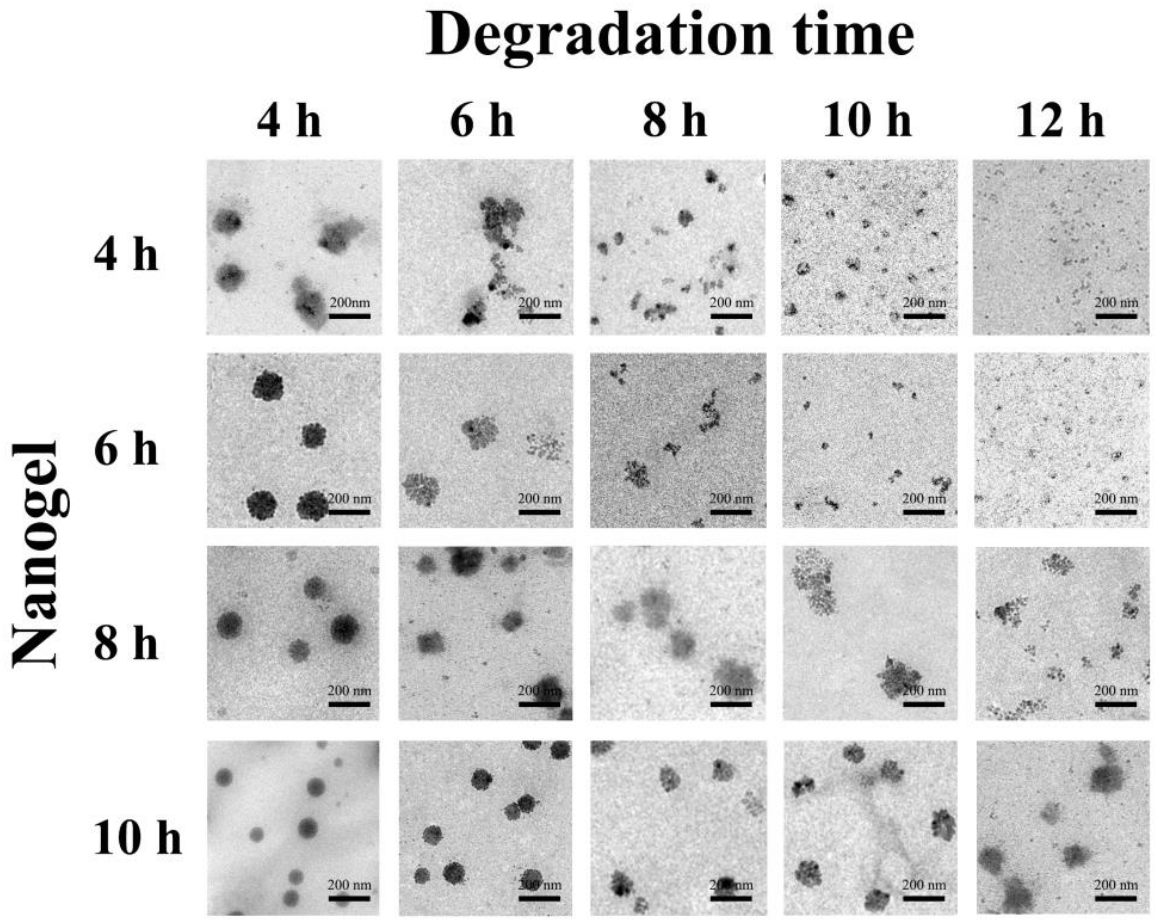

Fig. S1. The degradation of nanogels for 4, 6, 8, 10 and 12 h degradation times (x-axis), with gelation times of the nanogels labeled on the y-axis. Because nanogels are swollen nanosized networks that can absorb a lot of liquid in aqueous solutions, the sizes obtained from TEM and DLS results had a reasonable discrepancy.

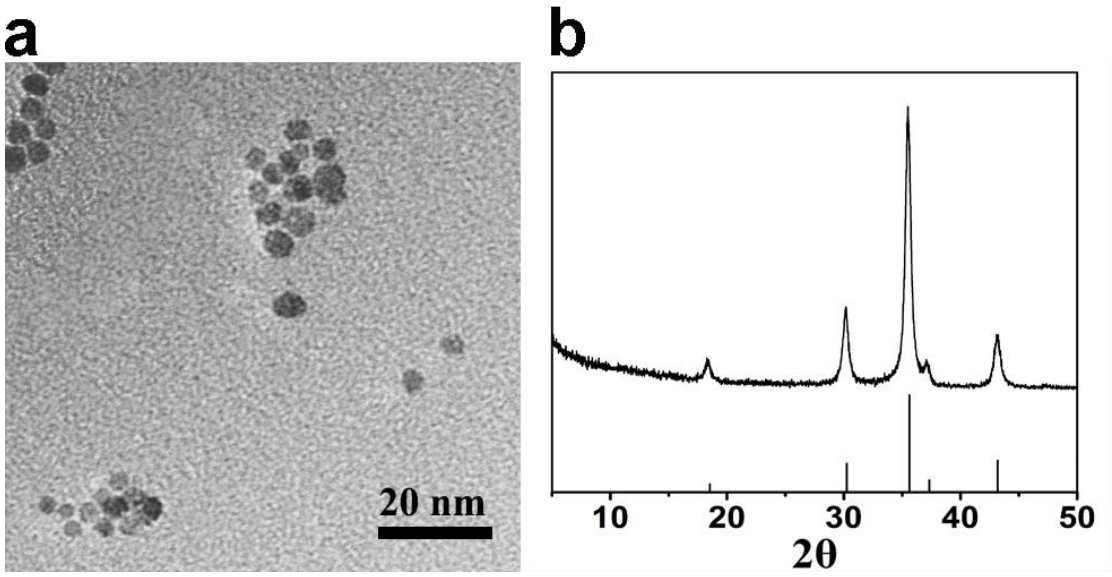

Fig. S2. (a) TEM image of Fe_3O_4 nanoparticles and (b) XRD pattern of Fe_3O_4 nanoparticles (curve) and the reference PDF for Fe_3O_4 (column).

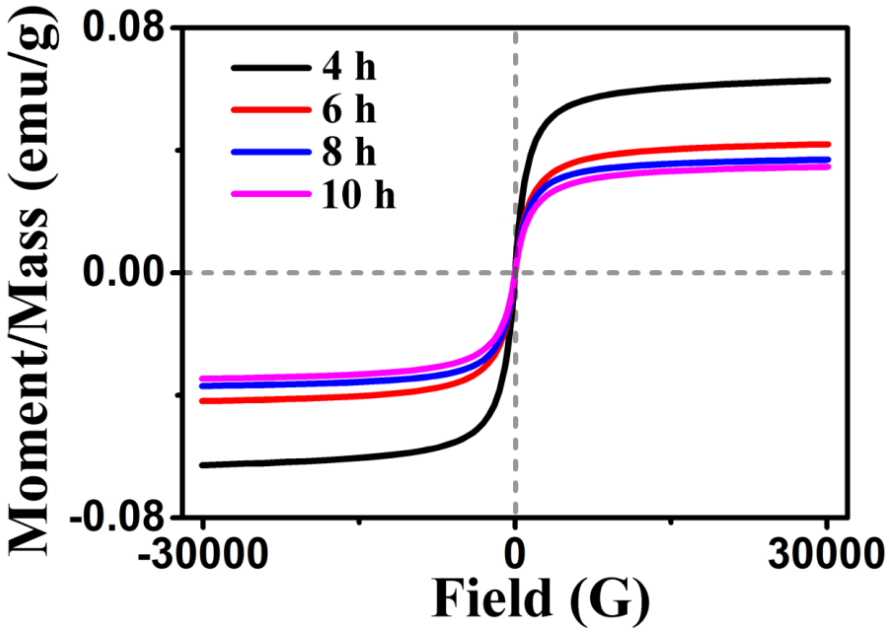

Fig. S3. Magnetic field curves for magnetic nanogel/SiO_2 NPs. Legend: 4h = magnetic nanogel/SiO_2 NPs obtained from a nanogel template with 4 h gelation time.

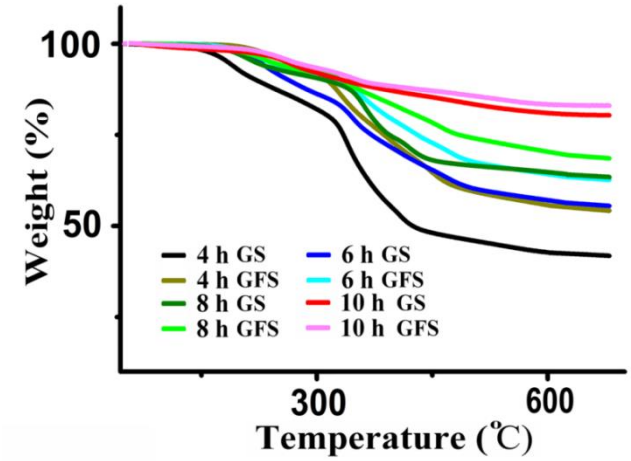

Fig. S4. The TGA results of thermal weight loss of nanogels in nanogel/SiO_2 NPs (labeled GS) and magnetic nanogel/SiO_2 NPs (labeled GFS). Legend: 4h = magnetic nanogel/SiO_2 NPs obtained from a nanogel template with 4 h gelation time.

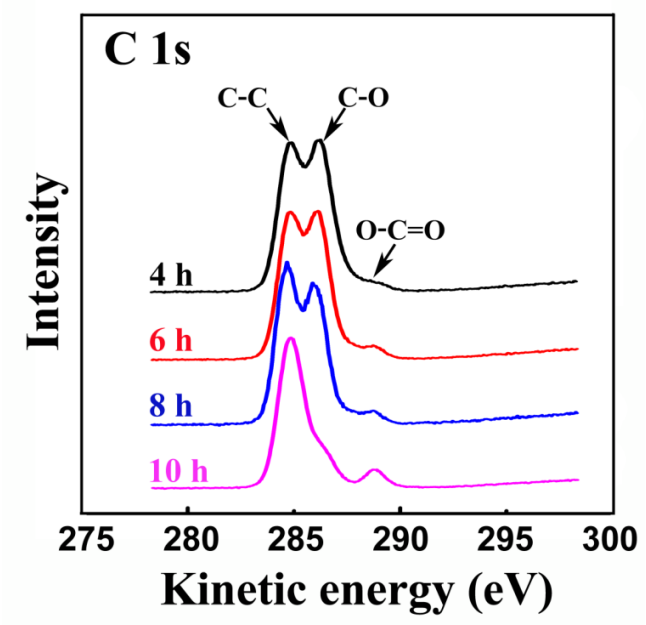

Fig. S5. XPS results of nanogel/SiO_2/PAA NPs. Legend: 4h = nanogel/SiO_2/PAA NPs obtained from a nanogel template with 4 h gelation time.

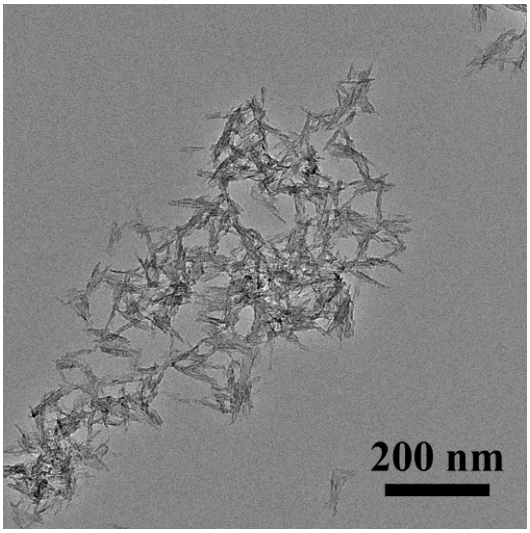

Fig. S6. TEM image of hydroxyapatite NPs.

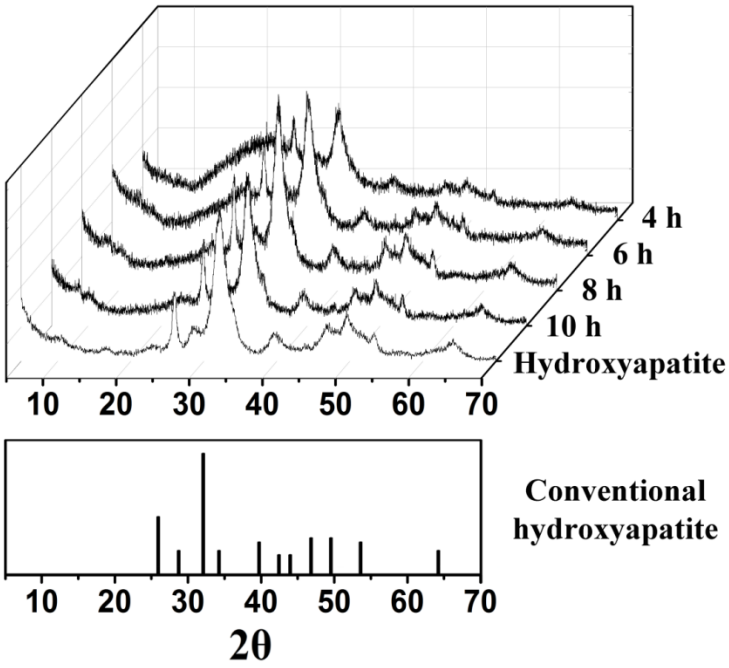

Fig. S7. XRD patterns of the nanogel/SiO_2/PAA/HA NPs (curve) and the reference PDF for hydroxyapatite (column). Y-axis: 4h = nanogel/SiO_2/PAA/HA NPs obtained from a nanogel template with 4 h gelation time.

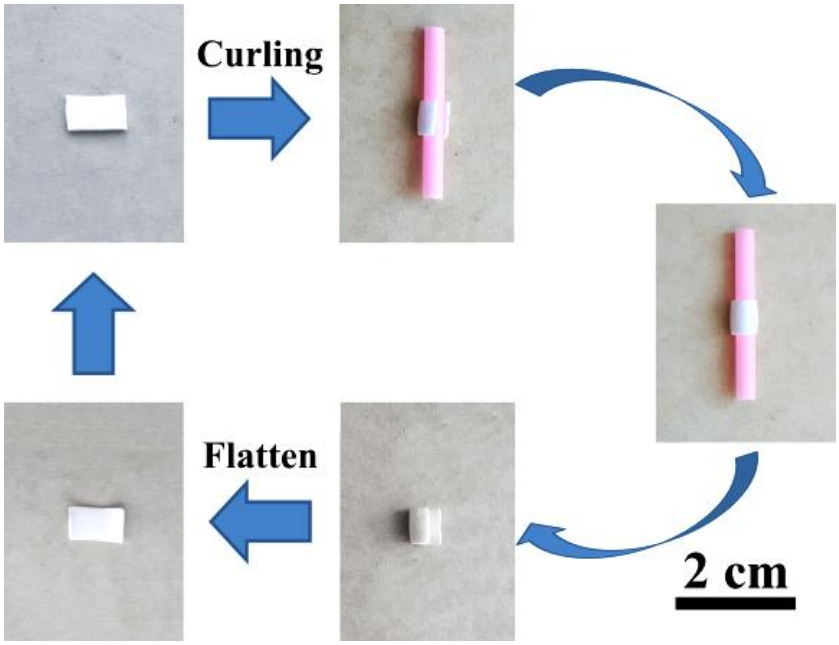

Fig. S8. Demonstration of biofilm flexibility with curling and spreading tests.

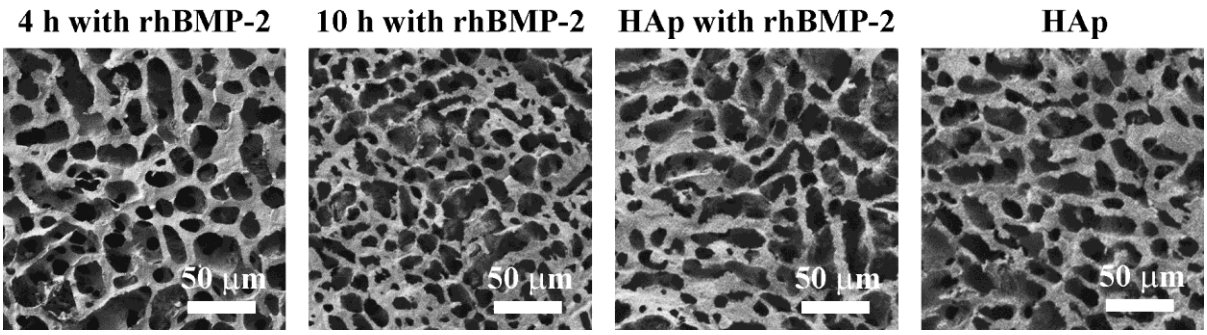

Fig. S9. SEM images of the films.

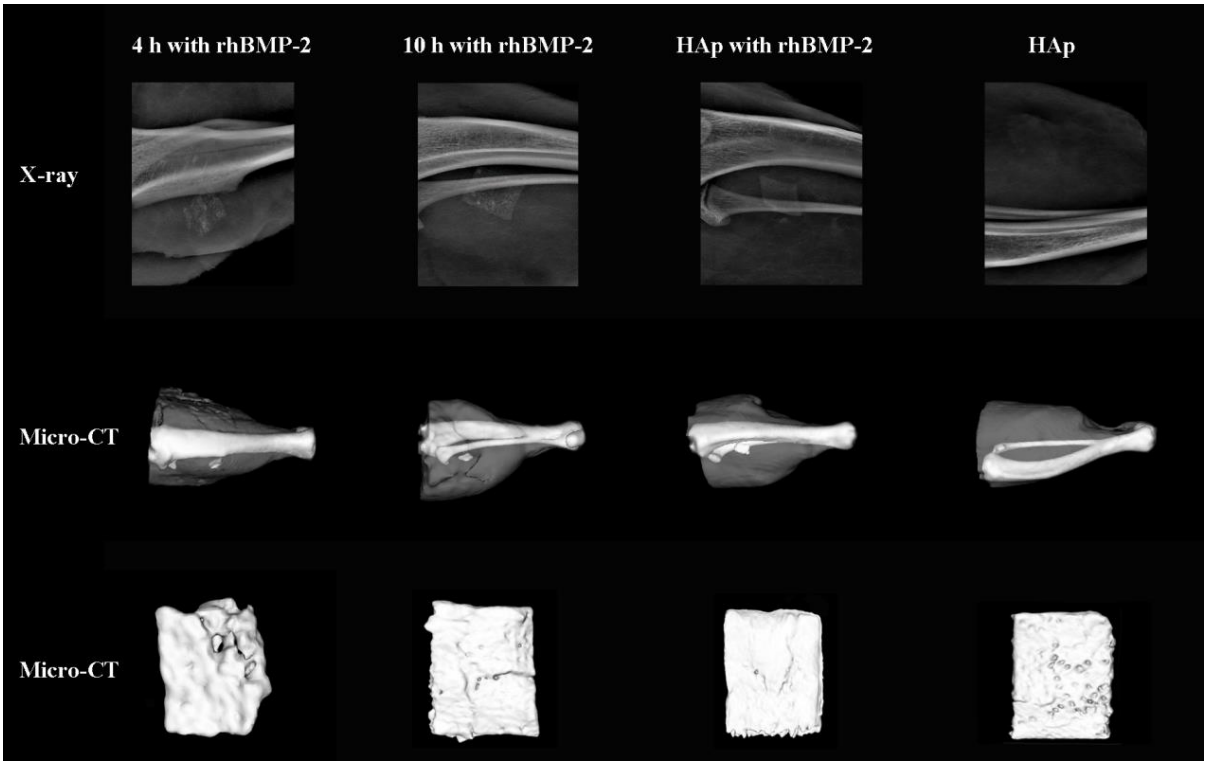

Fig. S10. X-ray and micro-CT characterization of ectopic bone formation after a 6-week post-implantation of (from left to right) the composite film doped with rhBMP-2 and nanogel/SiO_2/PAA/HA NPs from a 4 h gelation core (left), its 10 h equivalent, PLGA/HA film loaded with rhBMP-2, and PLGA/HA film alone.