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Supporting Information

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3D-Bioprinted Osteoblast-Laden Nanocomposite Hydrogel Constructs with Induced Microenvironments Promote Cell Viability, Differentiation, and Osteogenesis both In Vitro and In Vivo

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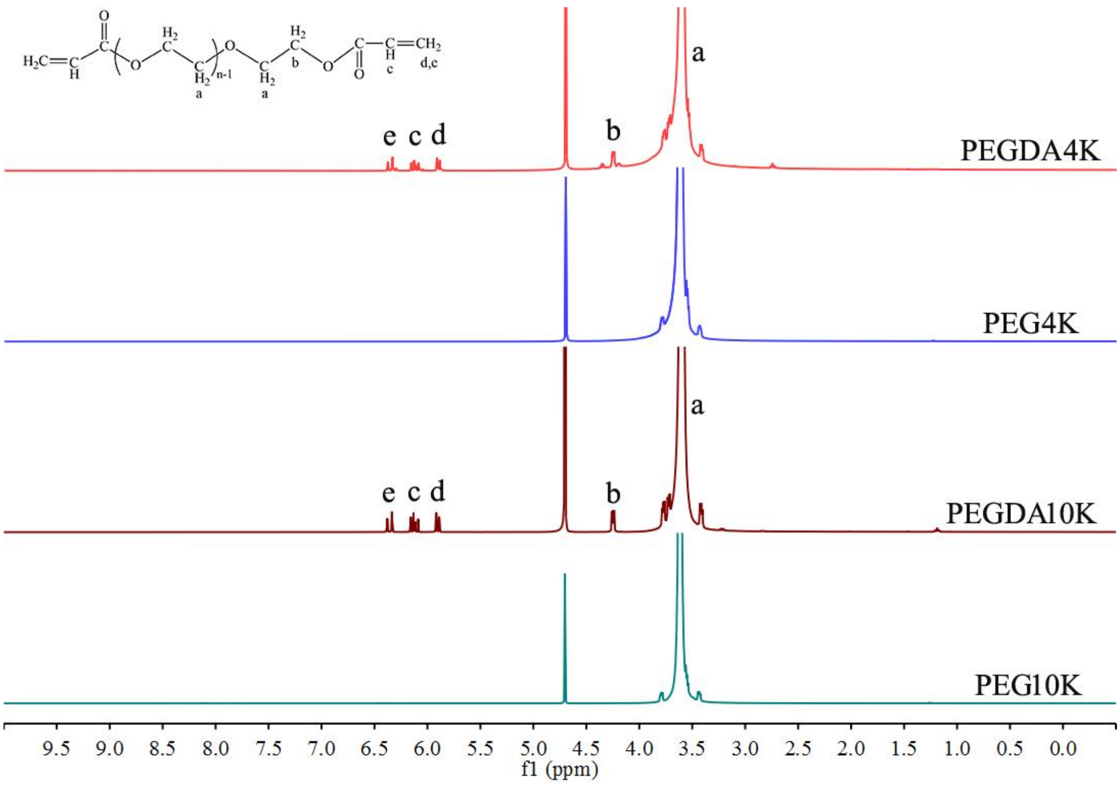

Figure S1. ^{1}H NMR spectra of PEG4K, PEG10K, PEGDA4K and PEGDA10K.

^{1}H NMR (D_2O): δ=6.35 (d, 2H, *cis* CH_2=CH), δ=5.80 (d, 2H, *trans* CH_2=CH), δ=6.10 (m, 2H, ROOCC*H*=CH$_2$), δ=4.25 (t, 4H, CH$_2$=CHCOOCH_2-CH$_2$O-), δ=3.5–3.6 (m, 360H, -CH_2CH_2O-).[1]

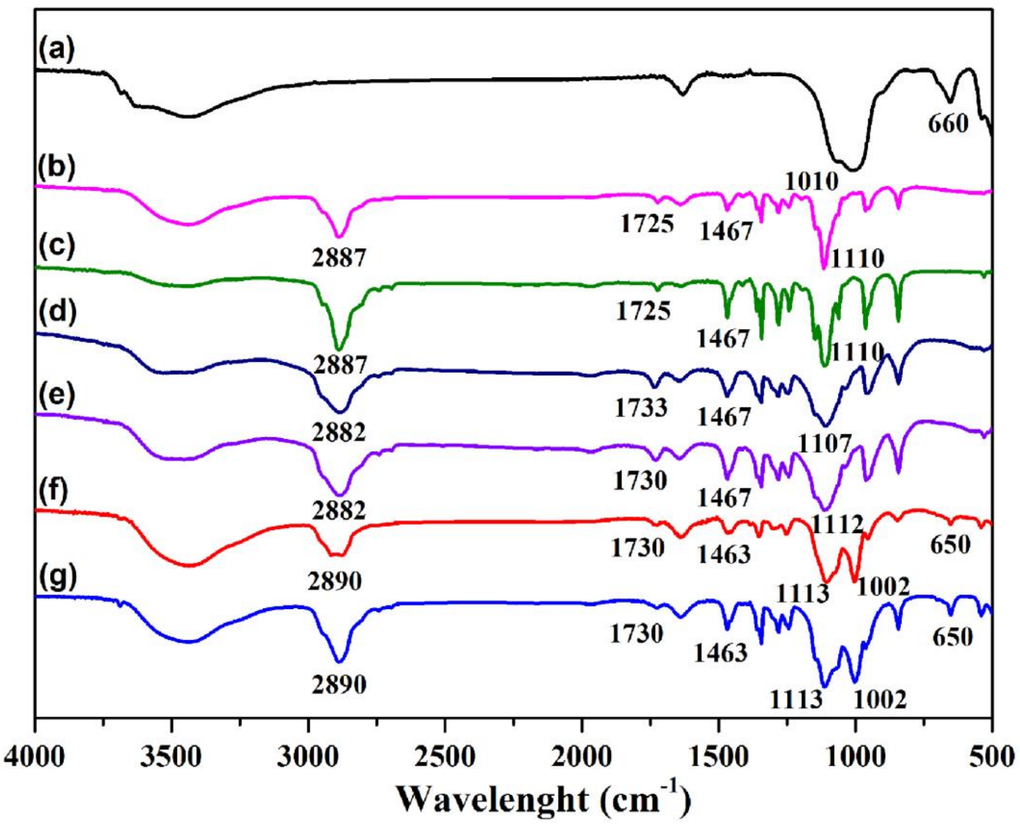

Figure S2. FTIR spectra of nanoclay (a), PEGDA4K (b), PEGDA10K (c), PEG4K hydrogel (d), PEG10K hydrogel (e), 20%PEG4K-7%Clay hydrogel (f) and 20%PEG10K-7%Clay hydrogel (g).

As shown in the figure, besides Si-O stretching and bending bands appearing at 1002 cm^{-1} and 650 cm^{-1}, PEG-Clay hydrogel also shows C-H stretching bands at 2890 cm^{-1}, C=O stretching vibration at 1730 cm^{-1}, C-O-C stretching bands at 1113 cm^{-1} and -CH_2- bending vibration at 1463 cm^{-1}, which can also be found out in PEGDA crosslinker and pure PEG hydrogel. This suggests the formation of hybrid hydrogel.[1, 2]

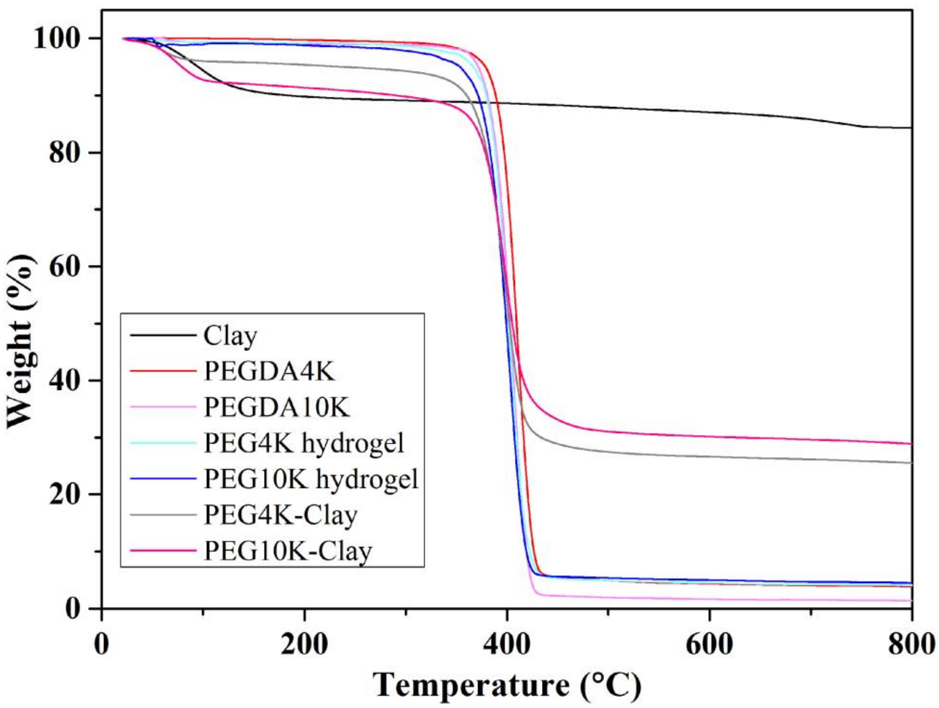

Figure S3. TGA curves of nanoclay, PEGDA4K, PEGDA10K, PEG4K hydrogel, PEG10K hydrogel, 20%PEG4K-7%Clay hydrogel and 20%PEG10K-7%Clay hydrogel.

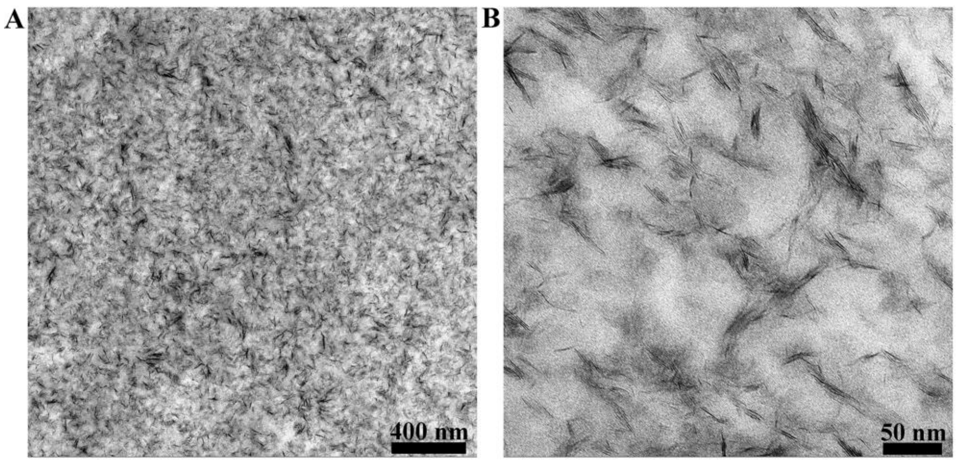

Figure S4. TEM images showing the exfoliation and dispersion of clay in the PEG4K-Clay hydrogel, where the dark platelets represent the exfoliated clay.

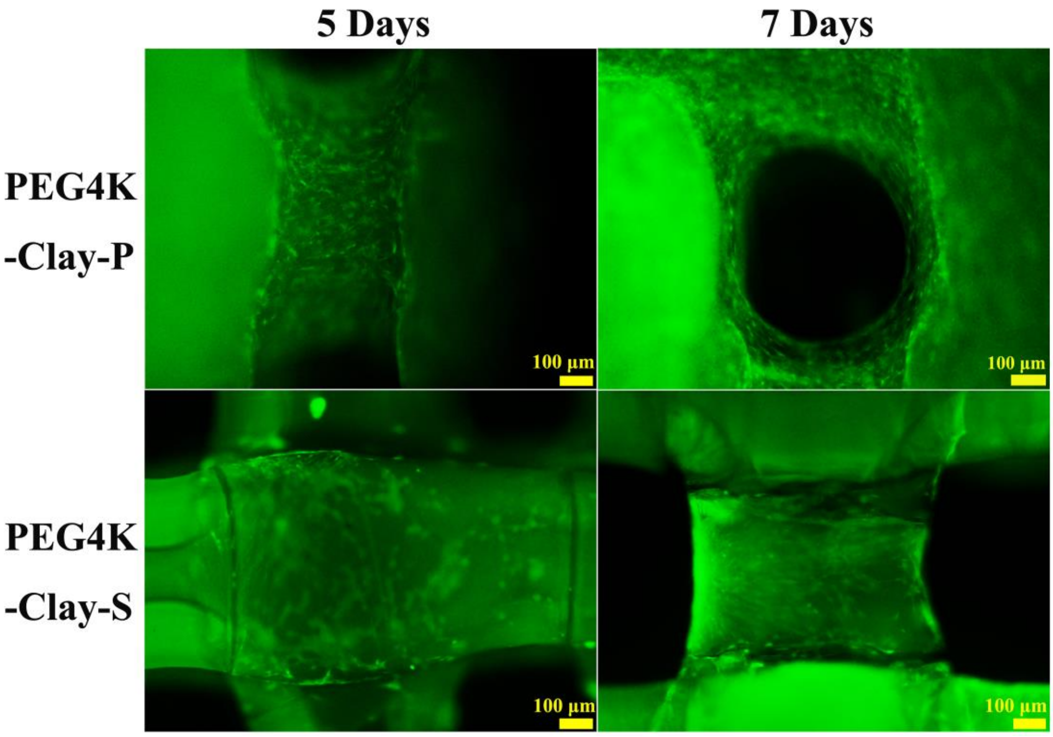

Figure S5. The status of ROBs after 3D-bioprinting (PEG4K-Clay-P) or traditional seeding (PEG4K-Clay-S) on PEG4K-Clay scaffolds after culturing for 5 and 7 days under 100× magnification.

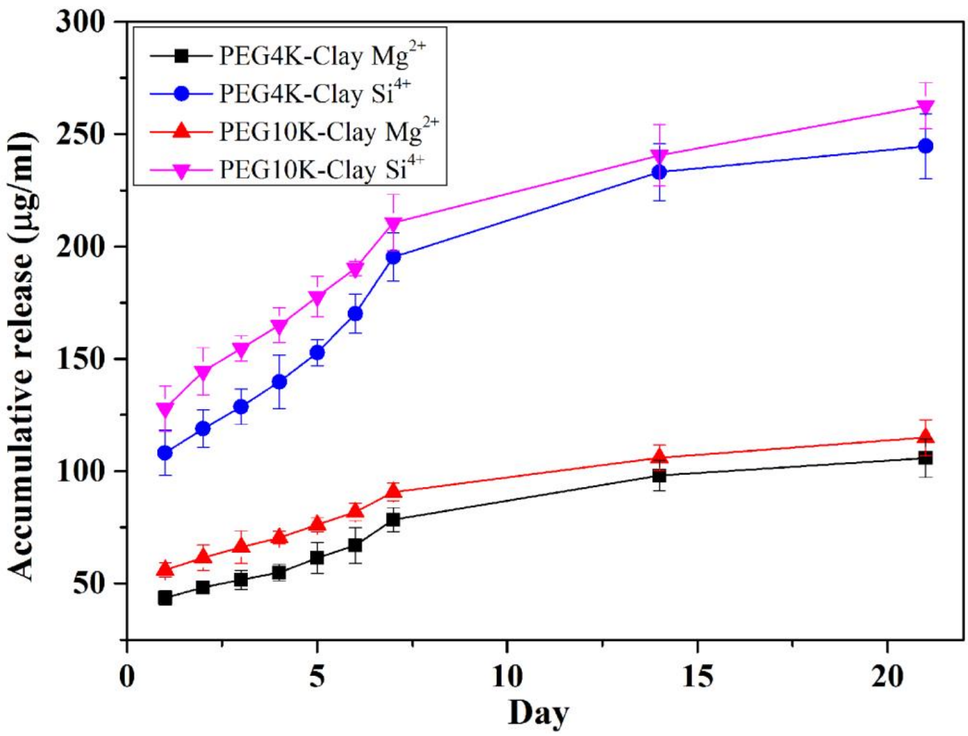

Figure S6. Accumulative release of magnesium ions and silicon ions from PEG-Clay scaffolds as a function of time.

Table S1. Preparation of PEG-Clay and PEG pre-hydrogel solutions with varied monomer concentrations.

Sample	PEGDA (g)	Laponite XLG (g)	H_2O (μL)
20%PEG4K-3%Clay	0.20	0.03	1000
20%PEG4K-5%Clay	0.20	0.05	1000
20%PEG4K-7%Clay	0.20	0.07	1000
20$PEG4K-10%Clay	0.20	0.10	1000
20%PEG10K-3%Clay	0.20	0.03	1000
20%PEG10K-5%Clay	0.20	0.05	1000
20%PEG10K-7%Clay	0.20	0.07	1000
20%PEG10K-10%Clay	0.20	0.10	1000
PEG4K	0.20	0	1000
PEG10K	0.20	0	1000

References

[1] J. Zhang, N. Wang, W. Liu, X. Zhao, W. Lu, *Soft Matter* **2013**, *9*, 6331

[2] H. Li, R. Wu, J. Zhu, P. Guo, W. Ren, S. Xu, J. Wang, *J. Polym. Sci., Part B: Polym. Phys.* **2015**, *53*, 876.