# Supporting Information

# Extracellular matrix mimetic supramolecular hydrogels reinforced with covalent crosslinked mesoporous silica nanoparticles

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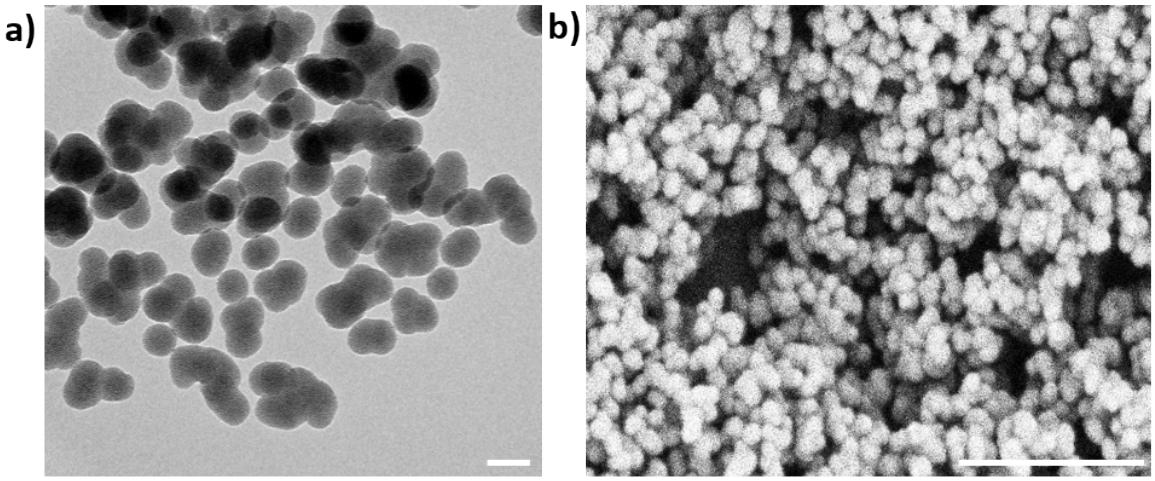


c)

| | Hydrodynamic Diameter (nm) | PDI | Zeta Potential (mV) |
|---|---|---|---|
| **MSN-OH** | 263.7 ± 2.0 | 0.12 | -29.2 ± 0.4 |

**Fig. S1:** Characterization of the synthesized MSN-OHs. (a) TEM imaging of MSN-OH (Scale bar: 50 nm); (b) SEM imaging of MSN-OH (Scale bar: 1 μM): (c) Hydrodynamic size and zeta potential of MSN-OHs.

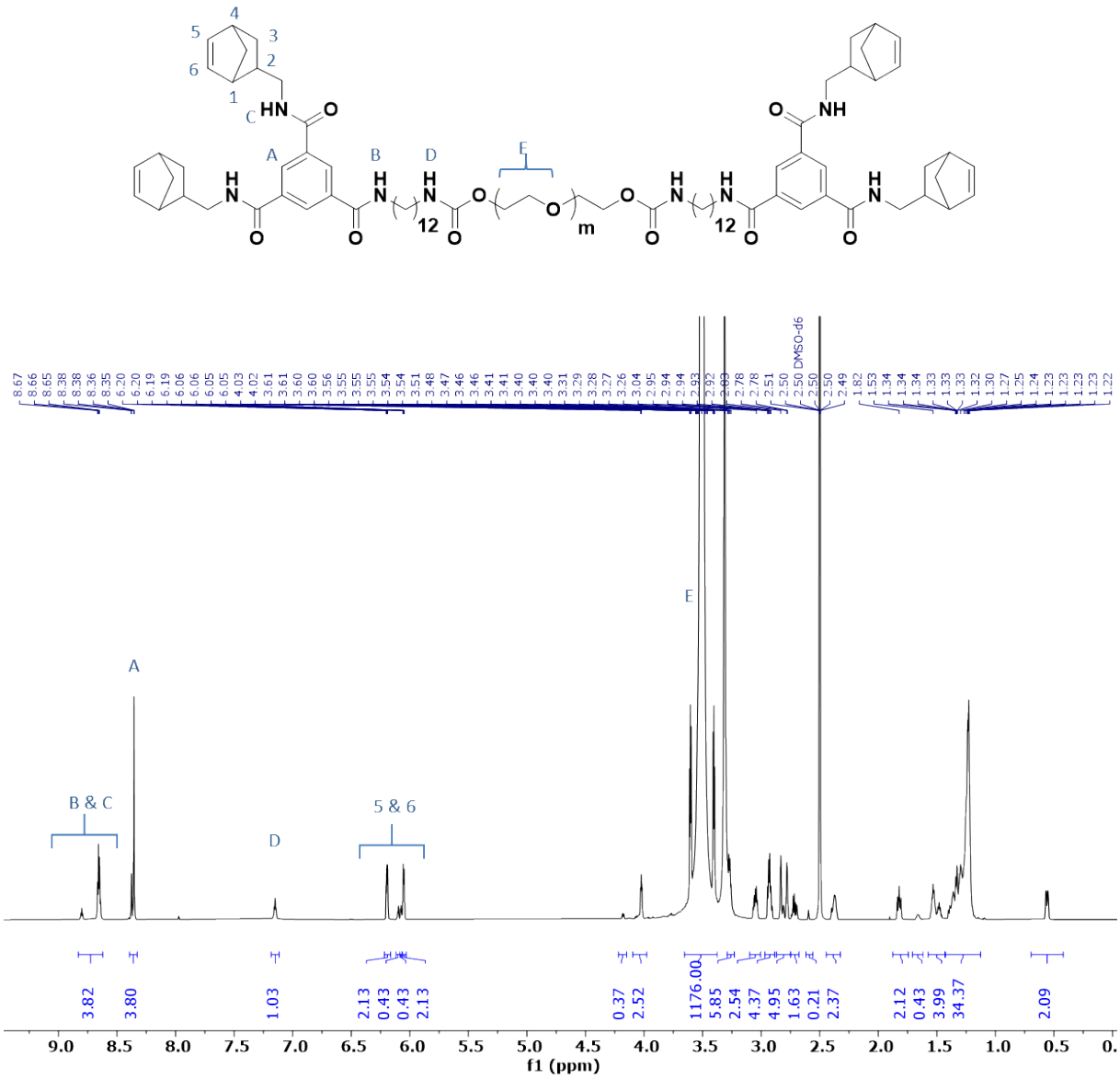


**Fig. S2** $^{1}$NMR data of NBTA macromonomers

*Table S1: Weight average (Mw), number average (Mn), and polydispersity (Đ) of the starting material (PEG 20K) and the final NBTA macromonomer.*

| Sample name | Mw (Daltons) | Mn (Daltons) | Polydispersity (Đ) |
|---|---|---|---|
| **PEG 20K** | 35368 | 31747 | 1.11 |
| **NBTA** | 47607 | 45583 | 1.04 |

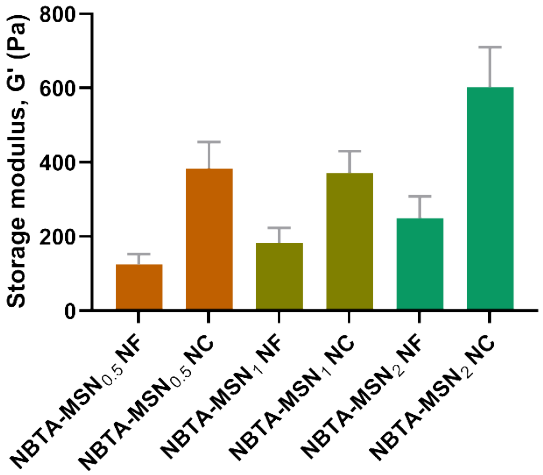


**Fig. S3** The storage modulus of NBTA-$MSN_{0.5}$, NBTA-$MSN_1$, and NBTA-$MSN_2$ hydrogels before UV light exposure where MSNs act as nanofillers (NF) and after UV light exposure as allowing MSNs to crosslink (NC)

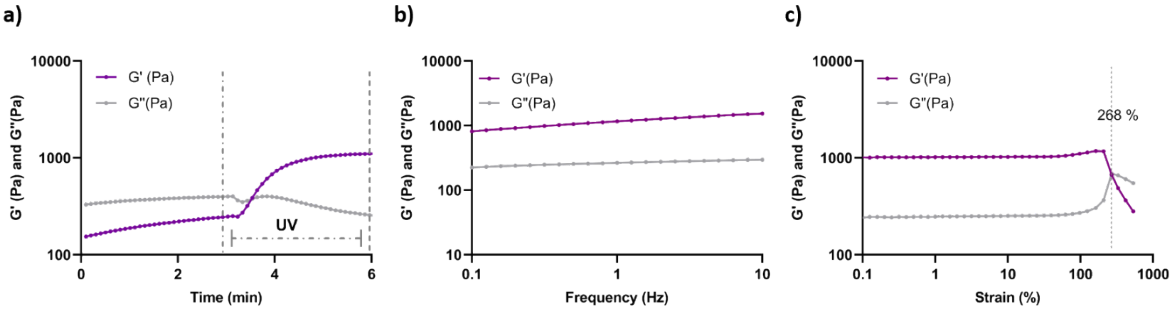


**Fig. S4** *In situ* photorheological characterization of NBTA-MSN-$CaP_1$ supramolecular nanocomposites. (a) Time sweep test of NBTA-MSN- $CaP_1$; (b) Frequency sweep test of NBTA-MSN- $CaP_1$; (c) Strain sweep test of NBTA-MSN-$CaP_1$

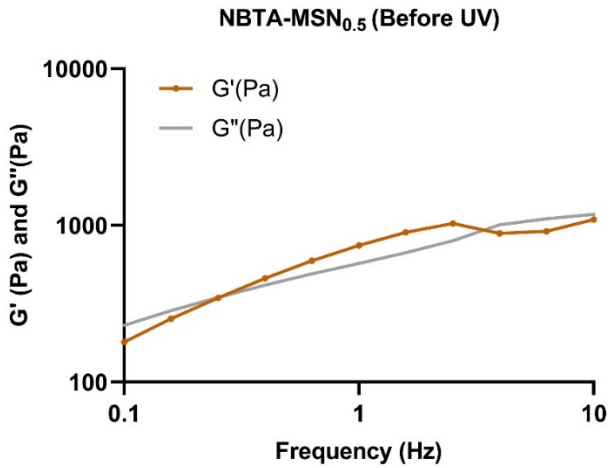


**Fig. S5** Frequency sweep of NBTA-$MSN_{0.5}$ supramolecular nanocomposite hydrogel before crosslinking. The frequency sweep test showed a typical viscoelastic behavior.

**Fig. S6.** Injectability of NBTA-$MSN_1$ nanocomposite hydrogels through the 19-Gauge syringe needle

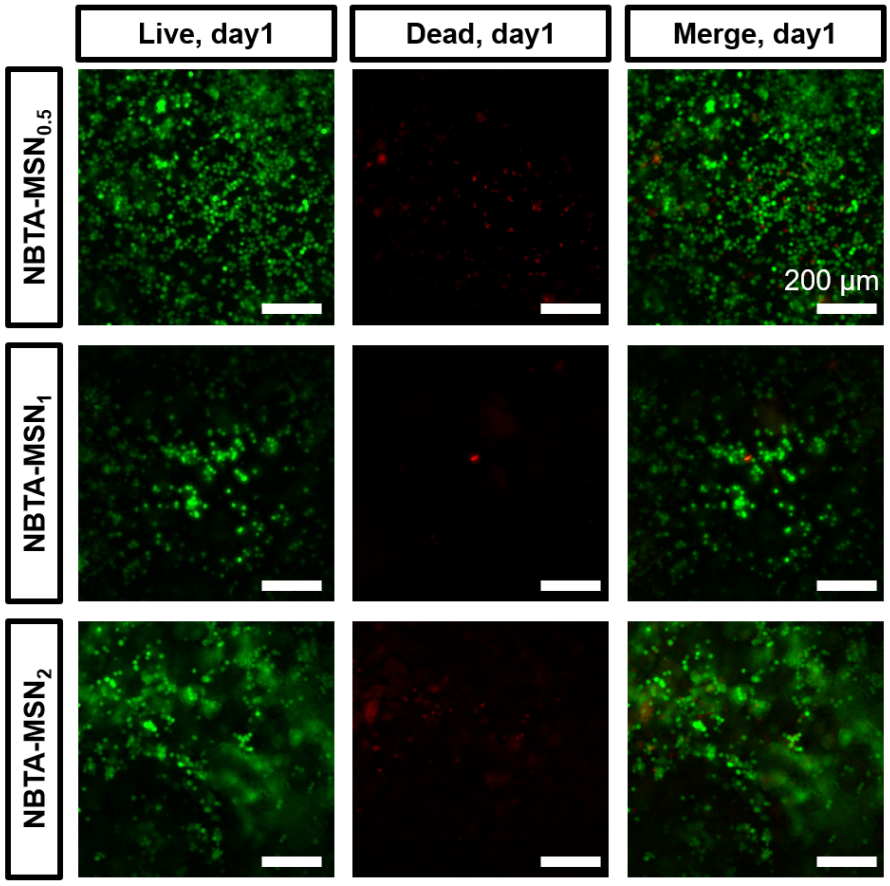


**Fig. S7** Viability of encapsulated MG63 cells inside NBTA-$MSN_{0.5}$, NBTA-$MSN_1$ and NBTA-$MSN_2$ hydrogels at day 1. The representative images of encapsulated cells stained with calcein AM (Green = live cells) and ethidium homodimer (EthD-1, red = dead cells). Scale bar: 200 µm

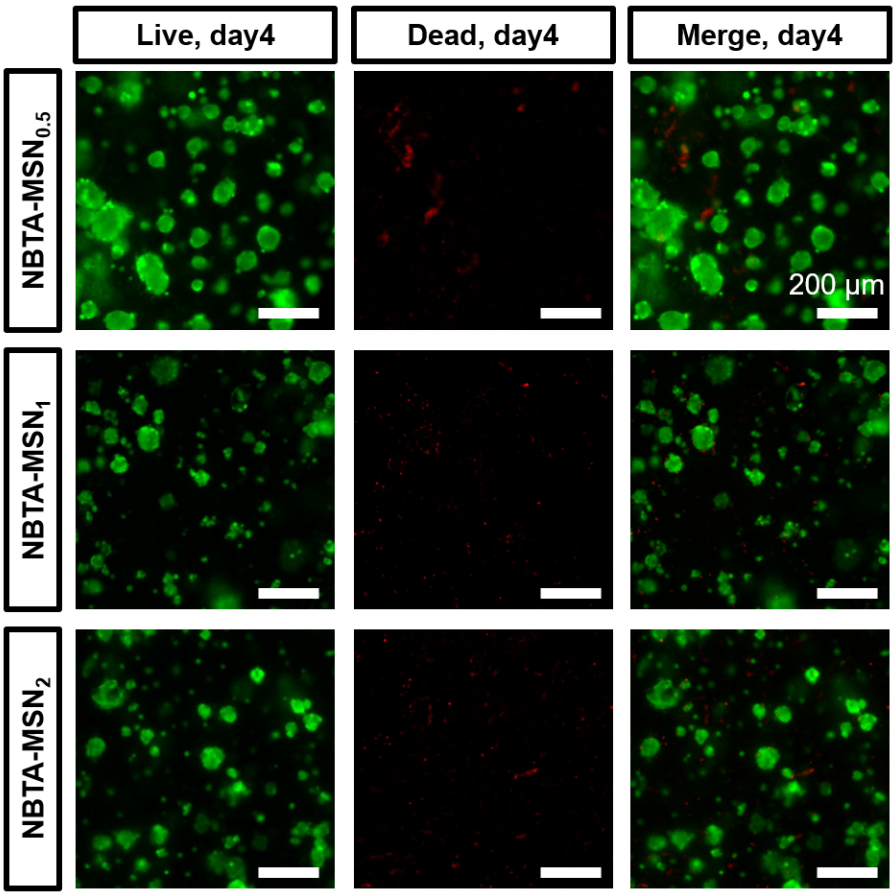


**Fig. S8** Viability of encapsulated MG63 cells inside NBTA-$MSN_{0.5}$, NBTA-$MSN_1$ and NBTA-$MSN_2$ hydrogels at day 4. The representative images of encapsulated cells stained with calcein AM (Green = live cells) and ethidium homodimer (EthD-1, red = dead cells). Scale bar: 200 µm

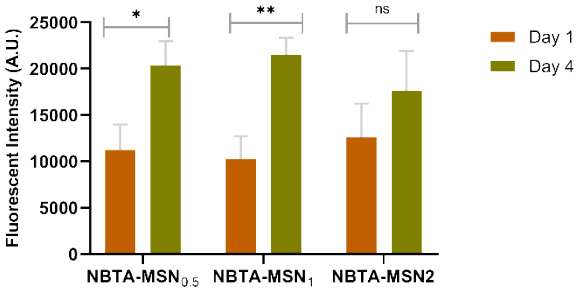


**Fig. S9** Metabolic activity of MG63 cells encapsulated NBTA-$MSN_{0.5}$, NBTA-$MSN_1$, NBTA-$MSN_2$ hydrogels during 4 days incubation in cell culture media. The data are presented as the mean ± SD for 3 replicates. Two-way ANOVA followed by Turkey's multiple comparison test was used, * P; *p =0.296, **=0.0067

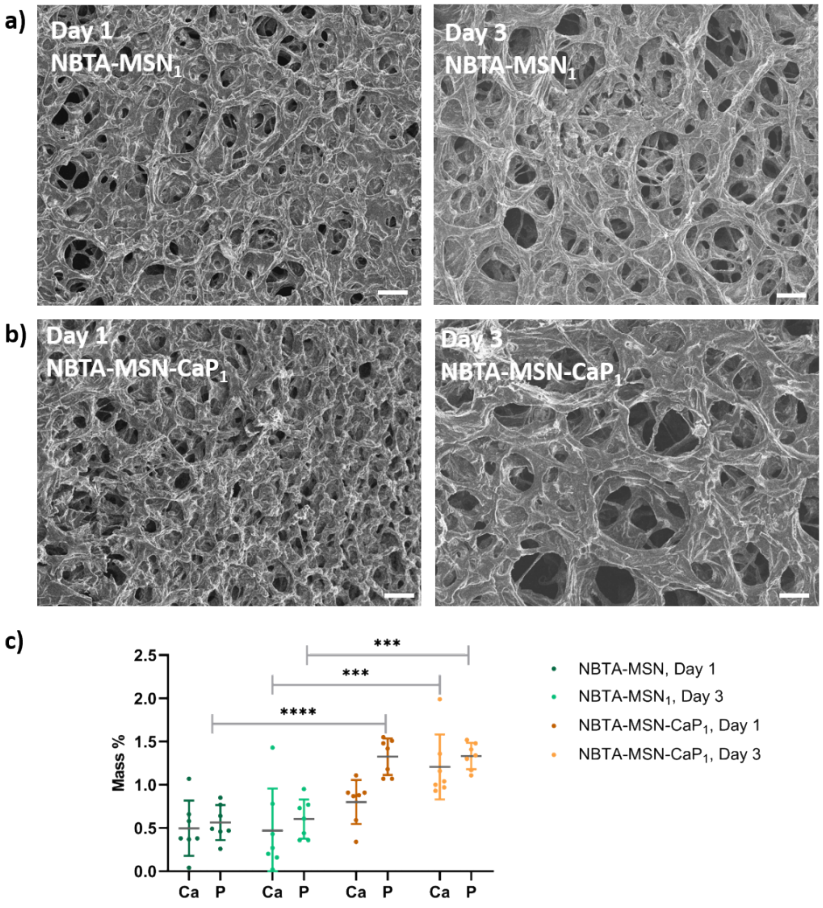


**Fig. S10** In vitro mineralization of NBTA-$MSN_1$ and NBTA-MSN-$CaP_1$ hydrogels in m-SBF buffer for 1 day and 3 days. (a) SEM image of freeze-dried NBTA-$MSN_1$ hydrogels after 1 day (left) and 3 days (right) of incubation in m-SBF buffer. (b) SEM images of freeze-dried NBTA-MSN-$CaP_1$ hydrogels after 1 day (left) and 3 days (right) of incubation in m-SBF buffer. (c) EDX analysis of Ca and P ions within freeze-dried NBTA-$MSN_1$ and NBTA-MSN-$CaP_1$ hydrogels (n=7). Scale bar for SEM images: 100µm on the left and 10 µm on the right. The data are presented as the mean ± SD for 7 sample areas. Two-way ANOVA followed by Turkey's multiple comparison test was used, * P; ***p =0.002, ****p<0.0001

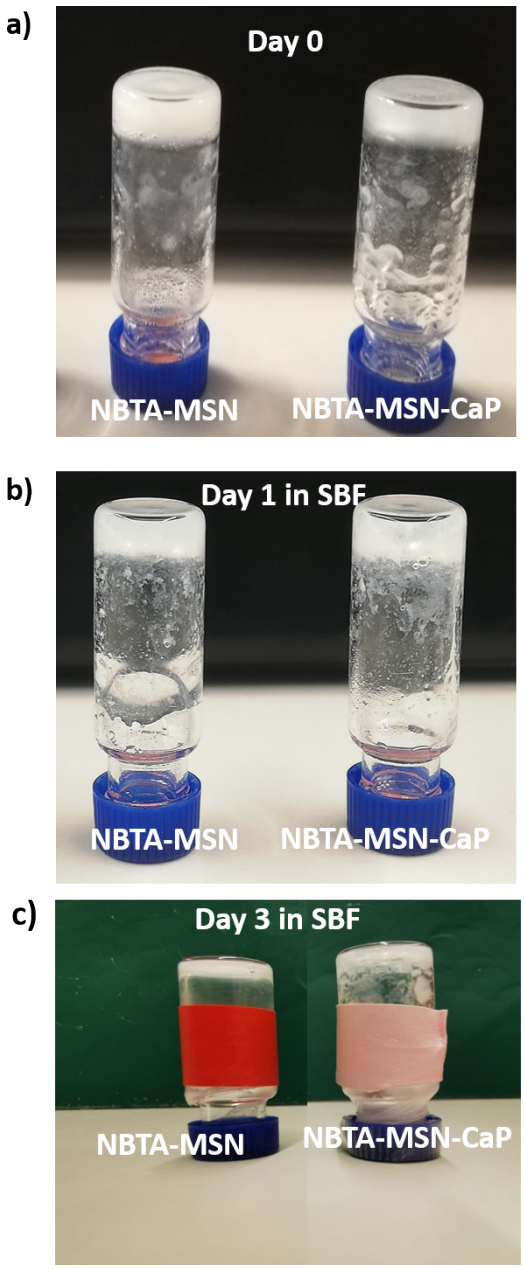


**Fig. S11.** Photographs of NBTA-$MSN_1$ and NBTA-MSN-$CaP_1$ nanocomposites (a) before SBF incubation, (b) after 1 day of incubation, and (c) 3 days of incubation in SBF buffer.