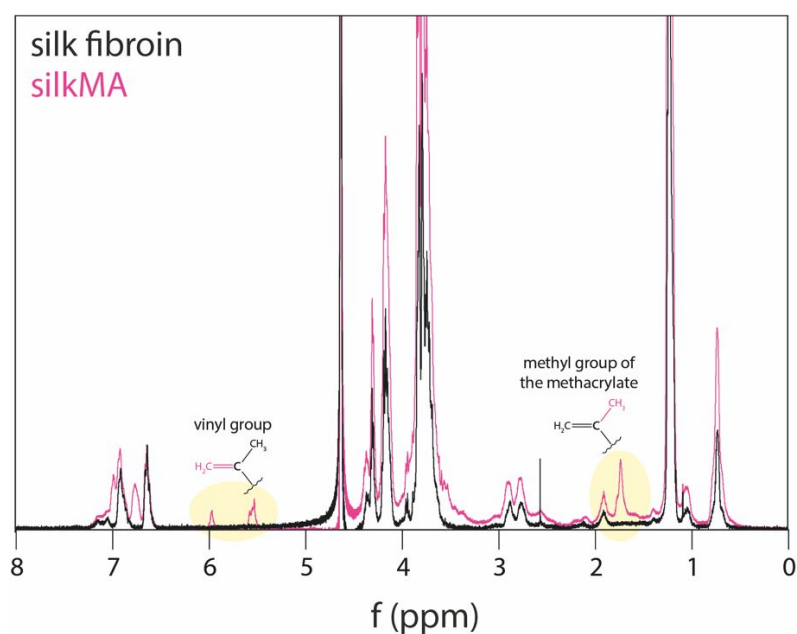


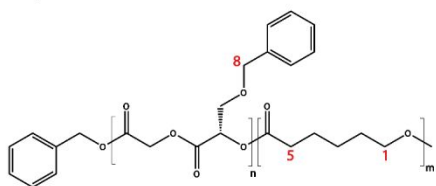
Covalent grafting of functionalized MEW fibers to silk fibroin hydrogels to obtain reinforced tissue engineered constructs

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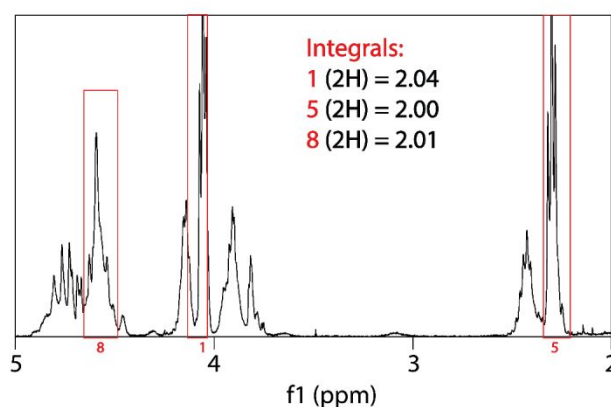


Supplemental figure 1. ¹H-NMR analysis of silkMA

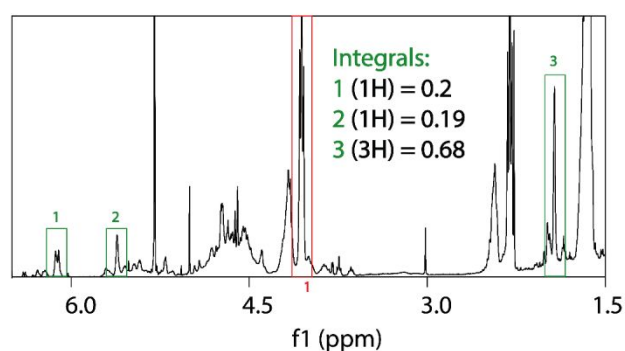
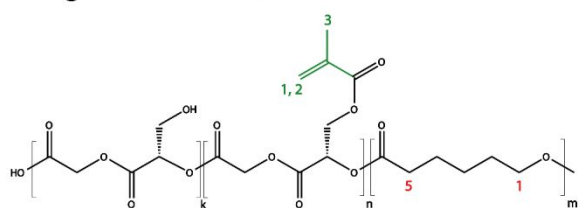
A. pBMGCL ¹H NMR



From this NMR we take as a reference the signals:
1CH₂ (4.0-4.1 ppm) and **5**CH₂ (2.2-2.3 ppm).
The integral of both these signals should be = 2
As a reference for the BMG we use the signal at 4.5
which correspond to **8**CH₂
The integral of that is also = 2, meaning that the ratio
between BMG and CL is 1:1 instead of 3:2.

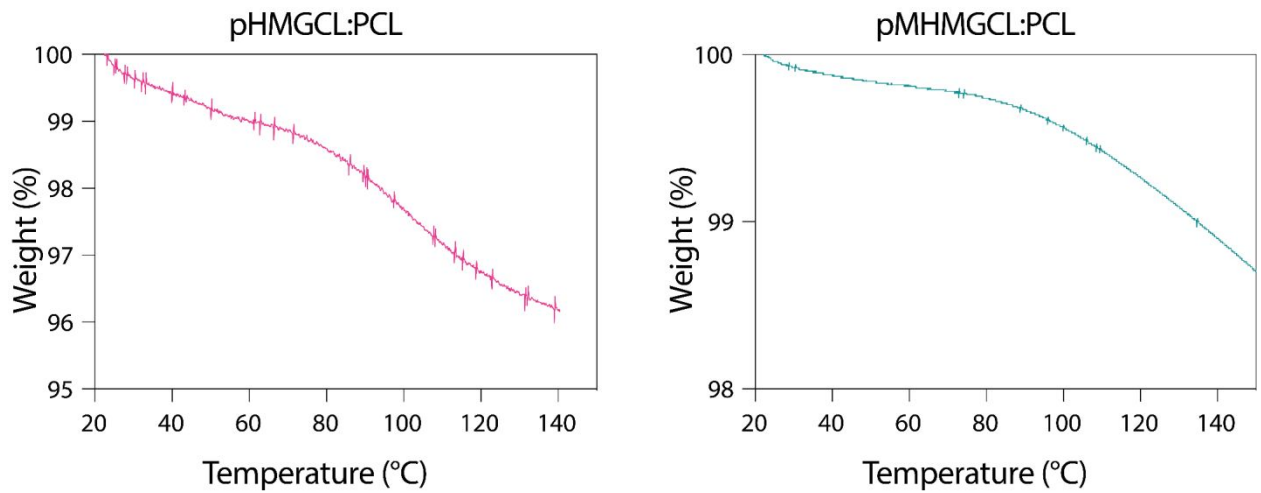


B. Degree of modification

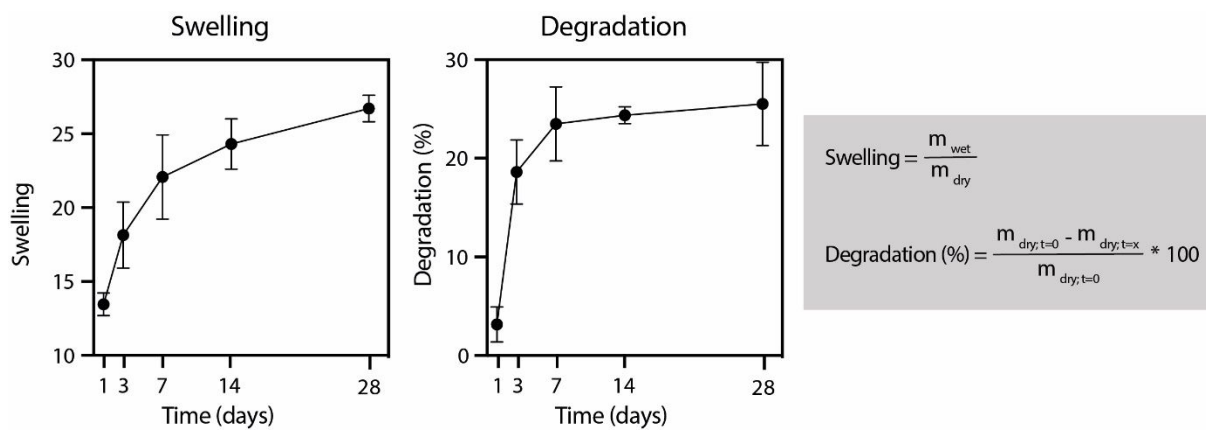


We use as reference peak always **1**CH₂ (4.0-4.1 ppm) which integrated is = 2. The signals of methacrylic anhydride are at 1.9 ppm, 5.6 ppm and 6.1 ppm. Since the integration of both peaks at 5.6 and 6.1 are around 0.2 and the one at 1.9 is 3 times higher we can conclude that we have almost 20% DoM in the BMG part of the polymer.

Supplemental figure 2. NMR analysis A. BMG:CL ratio calculation; B. Degree of modification

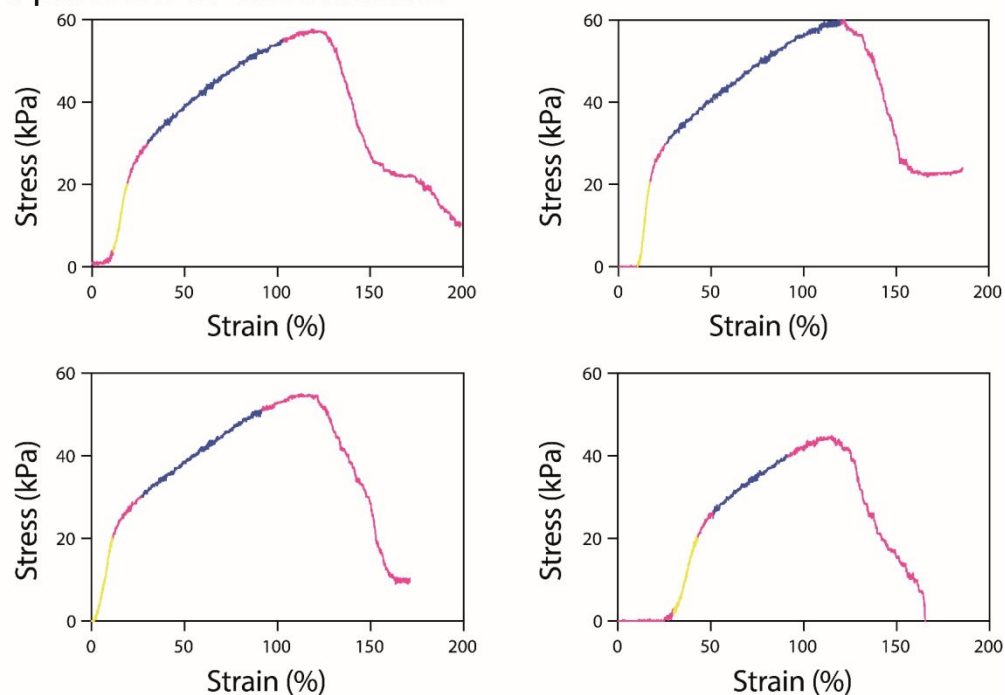


Supplemental figure 3. TGA analysis: degradation of pHMGCL:PCL and pMHMGCL:PCL polymer blends from RT to 140°C

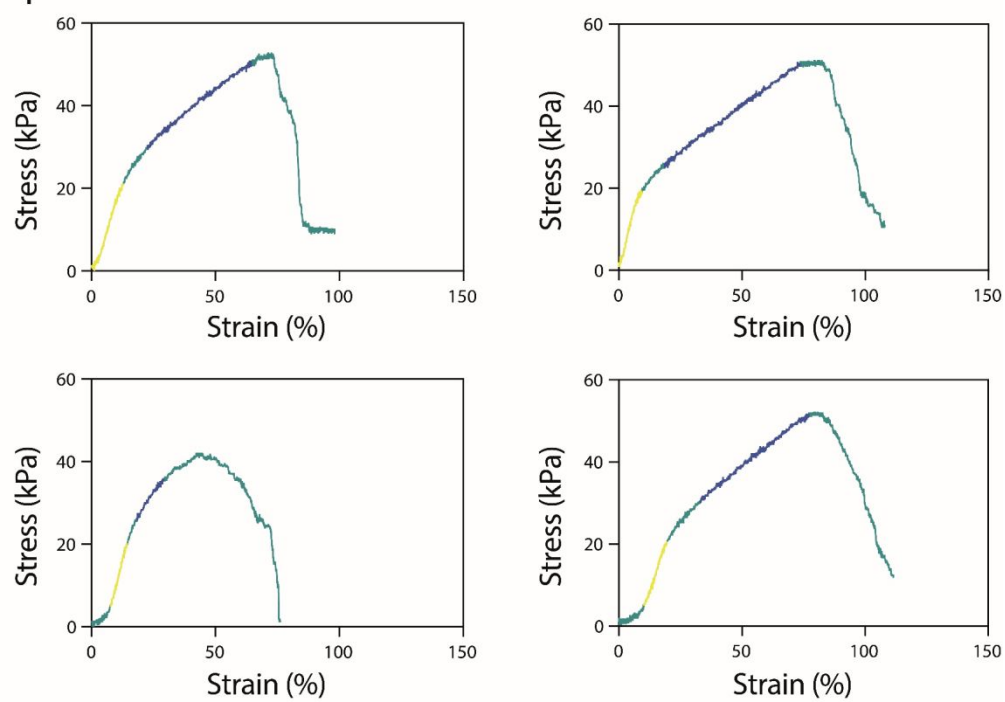


Supplemental figure 4. Swelling and degradation of a silkMA (7 % w v⁻¹) hydrogel with LAP (0.1 w v⁻¹) over 28 days (n = 3).

A. pHMGCL:PCL - silkMA scaffold

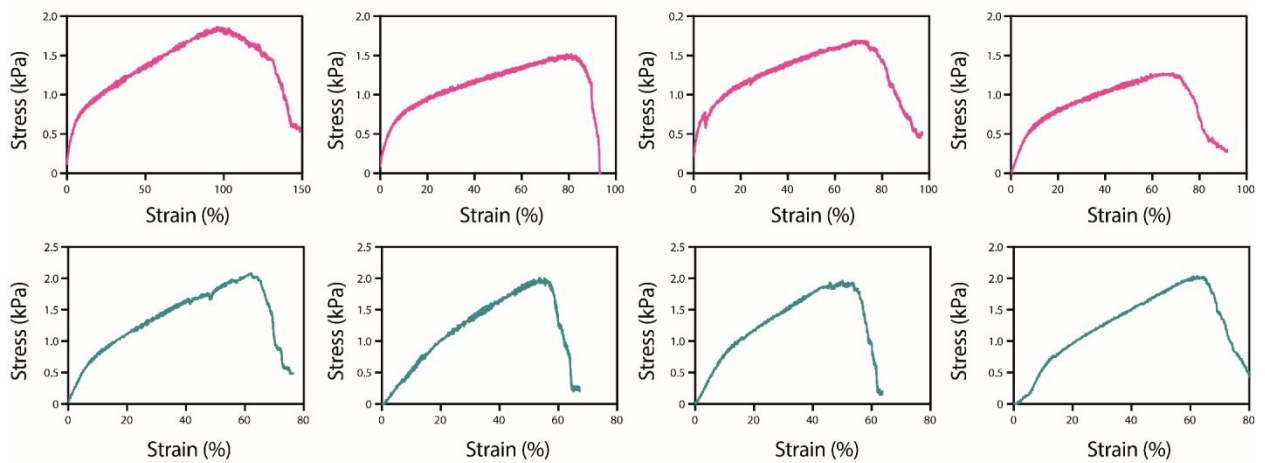


B. pMHMGCL:PCL - silkMA scaffold

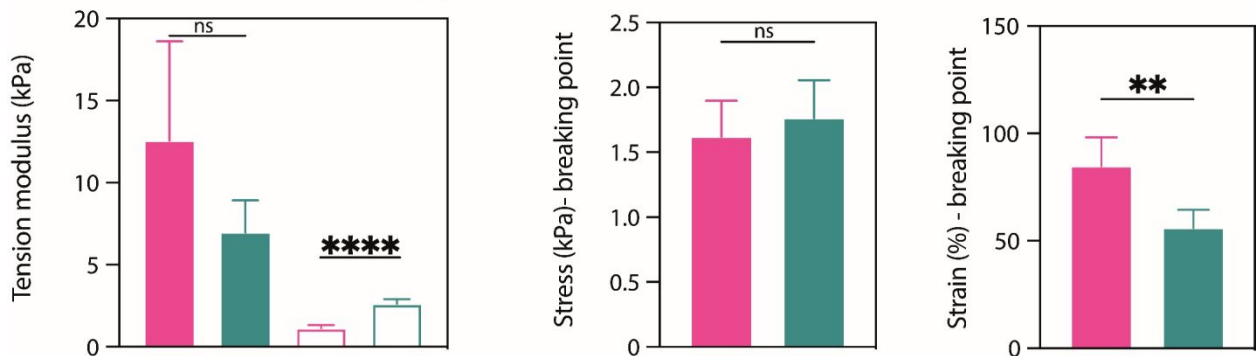


Supplemental figure 5. Mechanical characterization: curves for the uniaxial tensile tests of A. pHMGCL:PCL-silkMA (pink) and B. pMHMGCL:PCL-silkMA (green) scaffolds.

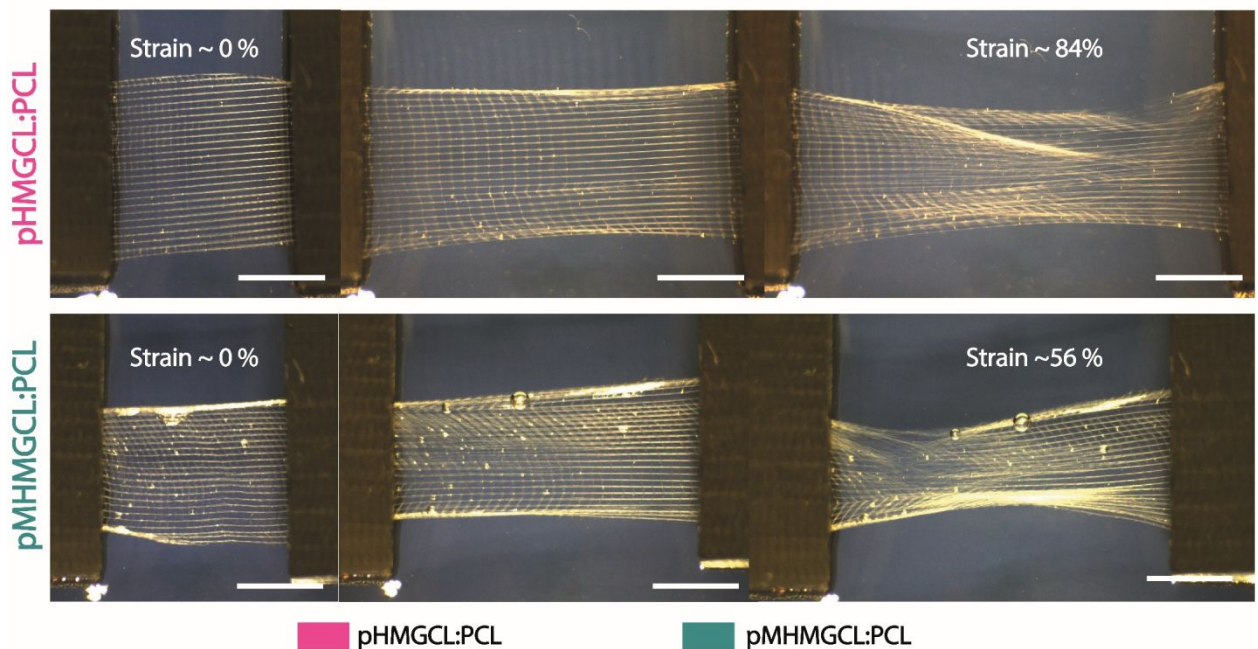
A. Stress-Strain curves for pHMGCL:PCL and pMHMGCL:PCL scaffolds



B. Tension moduli and breaking points



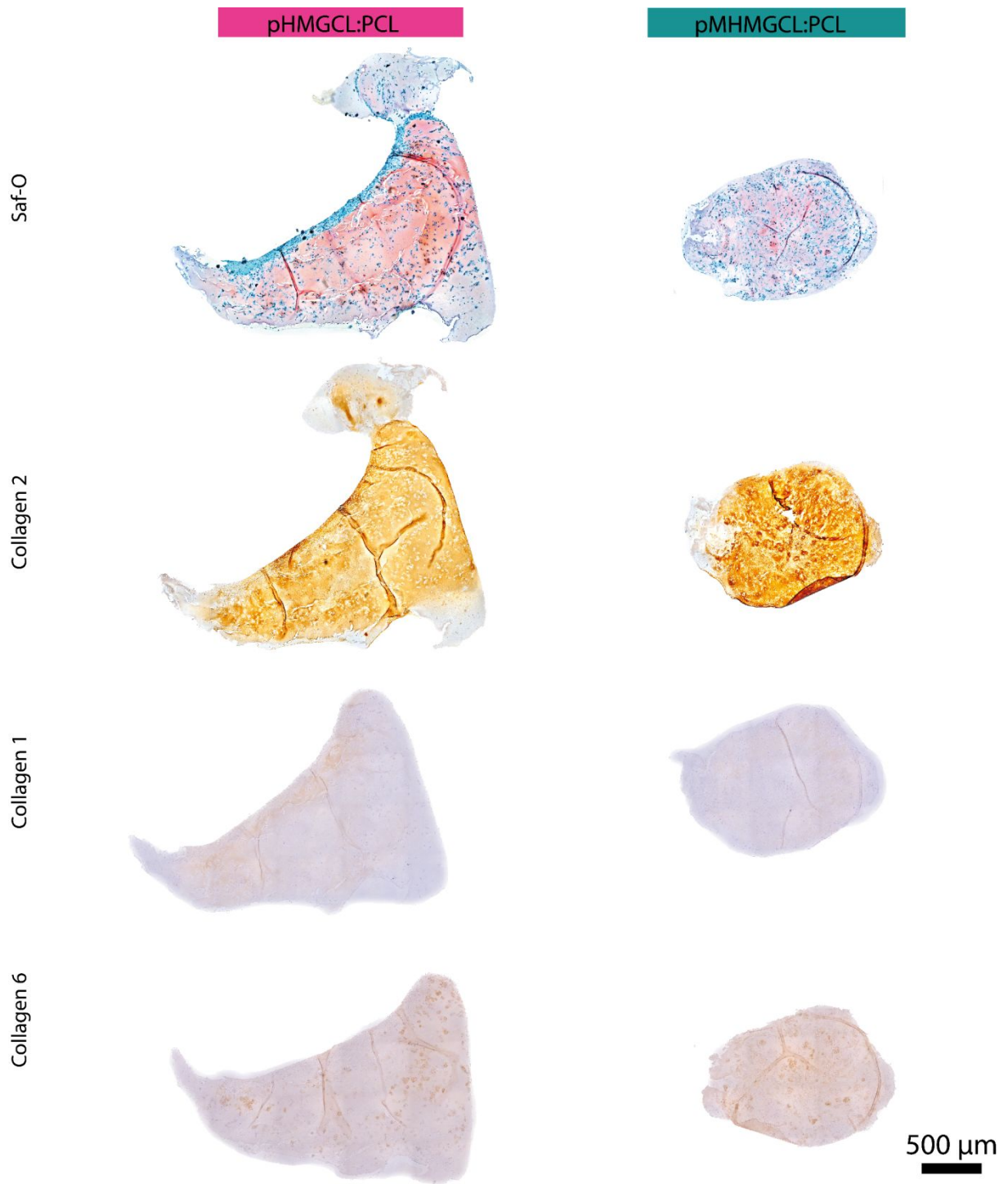
C. Top-view photographs of the uniaxial tensile testing setup



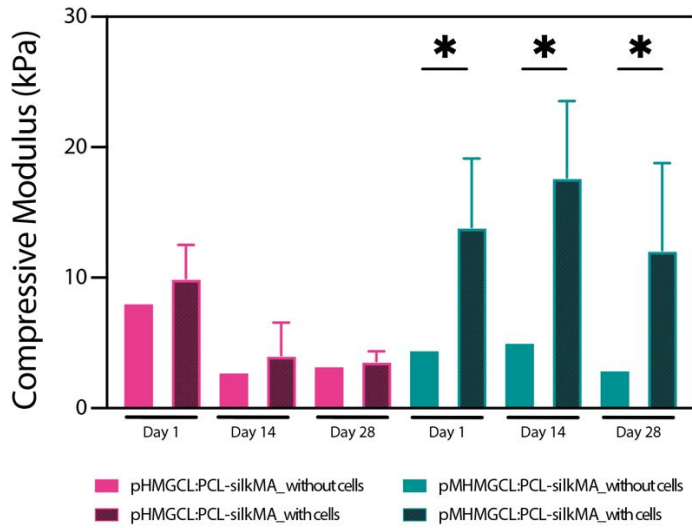
Supplemental figure 6. Mechanical characterization: A. curves for the uniaxial tensile tests of pHMGCL:PCL (pink) and pMHMGCL:PCL (green) scaffolds. B. Tensile modulus calculated using the

first and second slope of the curves, values of the stress and the strain at the breaking point for pHMGCL:PCL and pMHMGCL:PCL scaffolds (n=4), significance values: ** $p \leq 0.01$, **** $p \leq 0.0001$.

C. top-view photographs of the uniaxial tensile testing setup for pHMGCL:PCL and pMHMGCL:PCL: starting point, maximum elastic elongation and breaking point. Scale bar = 5 mm.



Supplemental figure 7. Overview (tile scans of whole samples) of histological analysis of samples at D28 *in vitro*. Representative image shown. n = 3. Scale bar = 500 μm.



Supplementary figure 8. Compressive analysis testing of samples prior to digestion showing compressive modulus. Data shown as mean \pm standard deviation ($n = 5$). The statistics show non-significant differences between samples unless indicated otherwise.

