ADVANCED HEALTHCARE MATERIALS

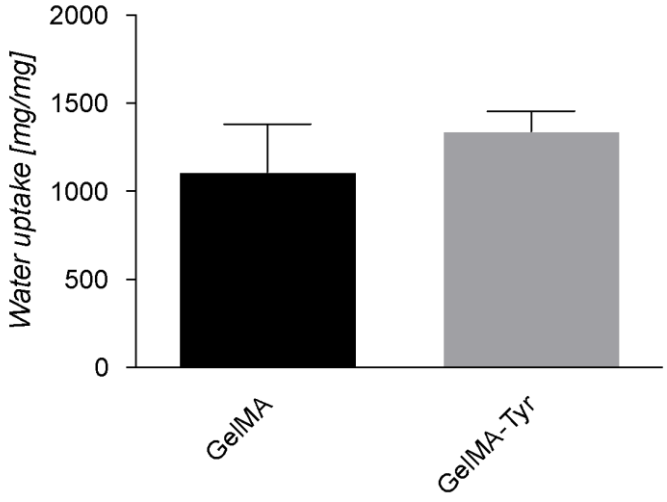
Supporting Information

for *Adv. Healthcare Mater.*, DOI: 10.1002/adhm.201901792

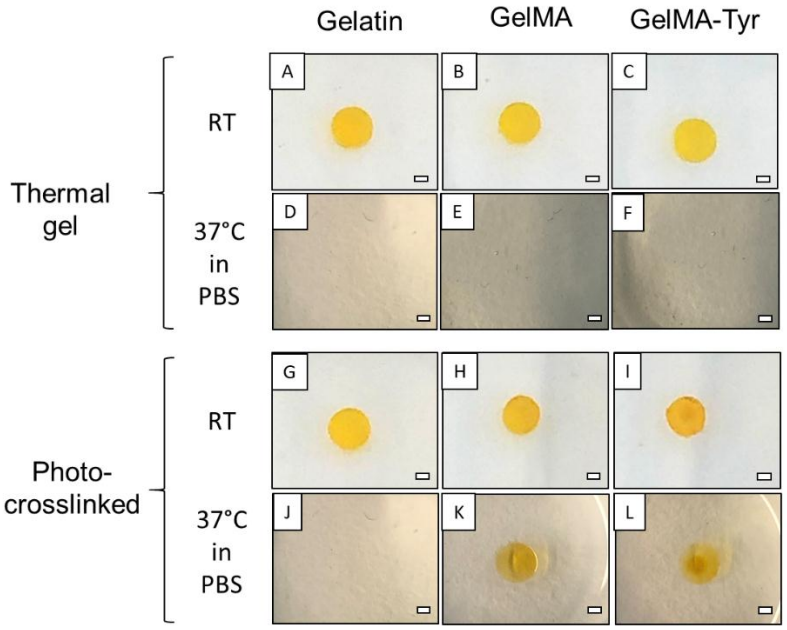
One-Step Photoactivation of a Dual-Functionalized Bioink as Cell Carrier and Cartilage-Binding Glue for Chondral Regeneration

Khoon S. Lim, Florencia Abinzano, Paulina Nuñez Bernal, Ane Albillos Sanchez, Pau Atienza-Roca, Iris A. Otto, Quentin C. Peiffer, Michiya Matsusaki, Tim B. F. Woodfield, Jos Malda, and Riccardo Levato**

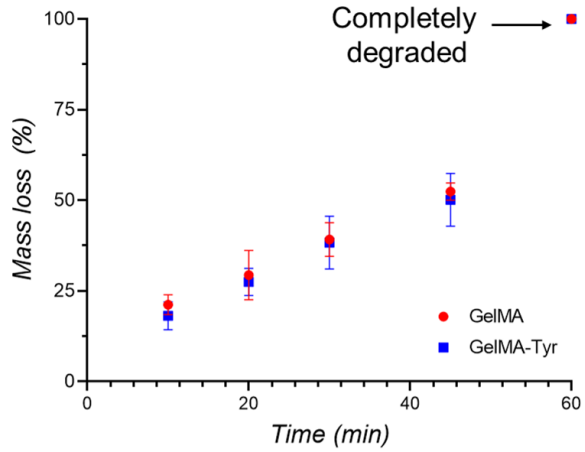
Supplementary figures



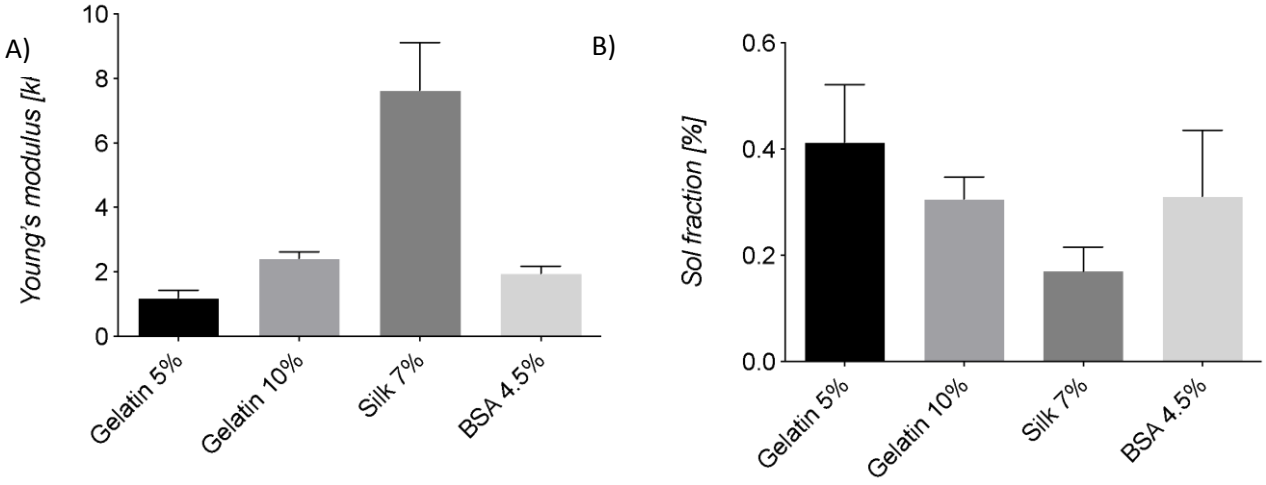
Supplementary figure S1: Water uptake in GelMA and GelMA-Tyr hydrogels.



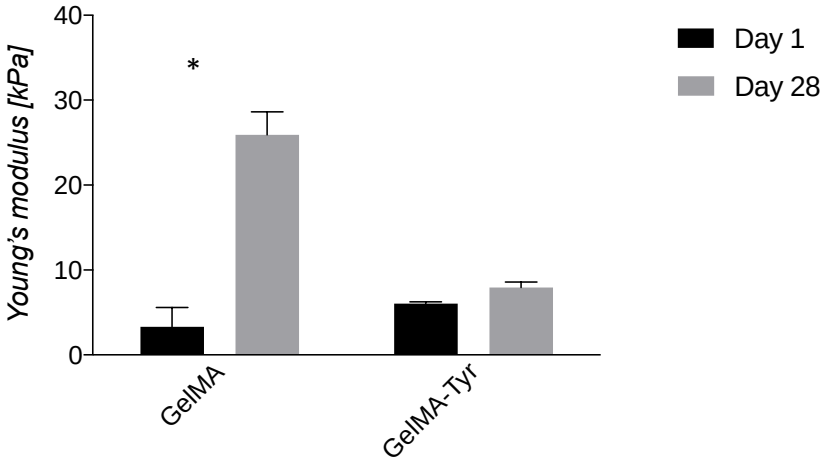
Supplementary figure S2: Thermally set or photo-crosslinked gelatin, GelMA and GelMA-Tyr hydrogels kept at RT or incubated in PBS at 37°C. Scale bar = 1mm.



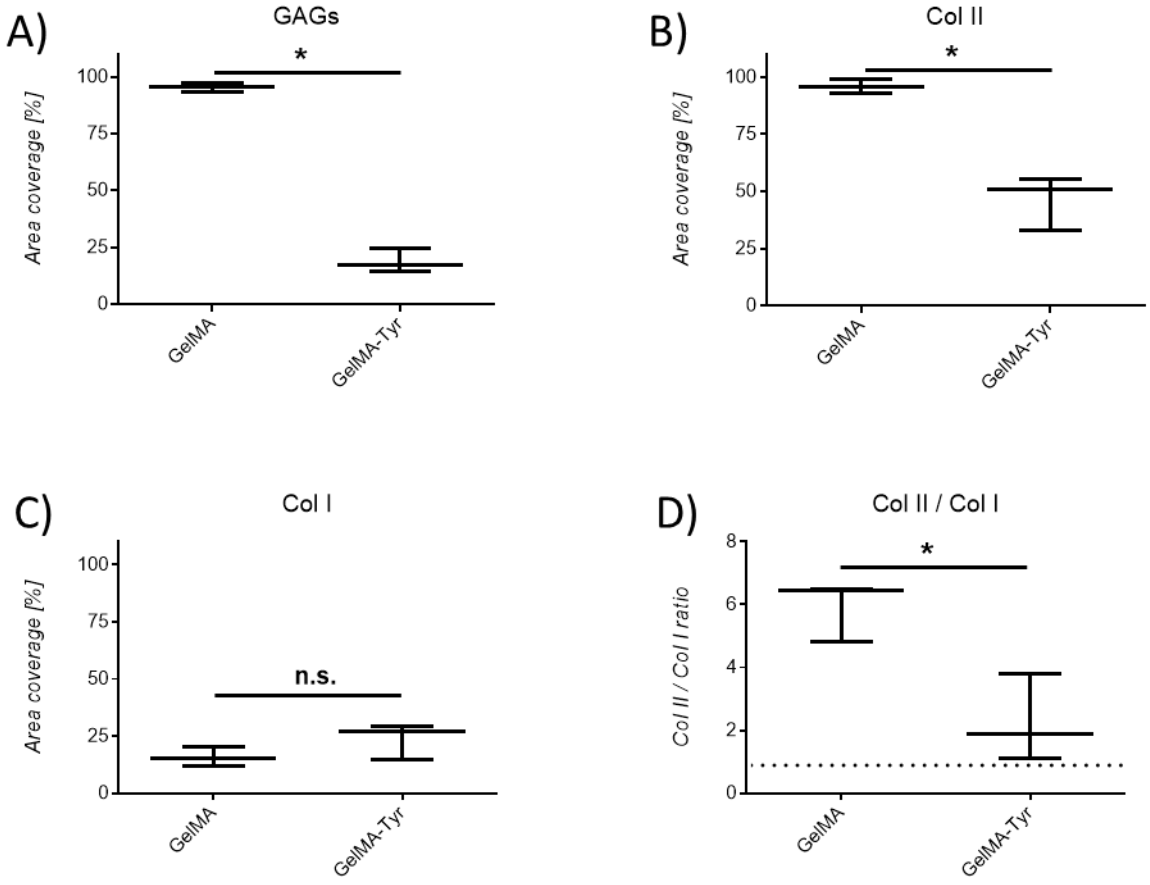
Supplementary figure S3: Mass loss of GelMA and GelMA-Tyr hydrogels in collagenase. A mass loss of 100% indicates complete degradation.



Supplementary figure S4: Ru/SPS can be used to crosslink any protein that contains tyrosine residues, creating hydrogels without the need for further modification. Here, 0.5/5mM Ru/SPS was used to crosslink gelatin, silk and bovine serum albumin (BSA); (A) sol-fraction and (B) Young's modulus were calculated to confirm the formation of the hydrogel networks.

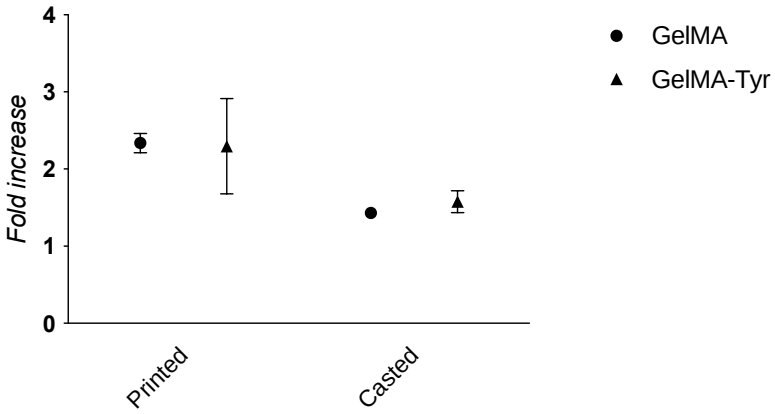


Supplementary figure S5: The compressive modulus of GelMA was significantly increased with culture, while no increment could be detected in GelMA-Tyr hydrogels.

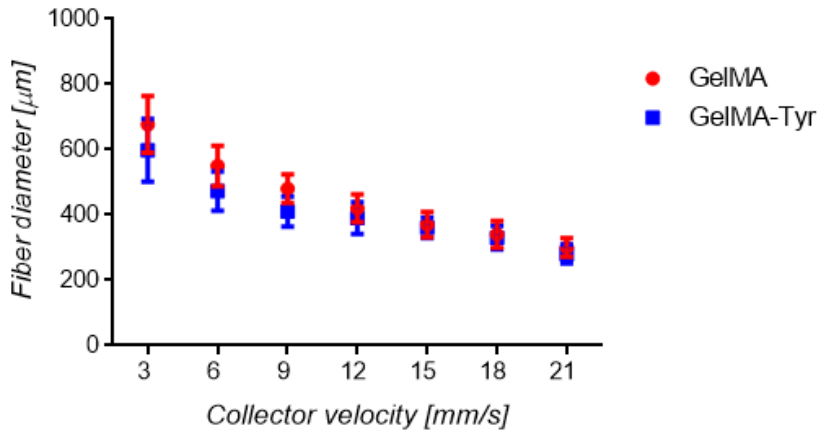


Supplementary figure S6. Distribution of different cartilage ECM components as observed in the histological slides. Image analysis performed on the stained slides confirmed the qualitative observations, showing a more homogenous distribution of A) GAGs and B) collagen type II in the GelMA samples, compared to GelMA-Tyr, where no significant difference was found in the area coverage of (C) collagen type I. Finally, for both GelMA and GelMA-Tyr D) collagen type II was

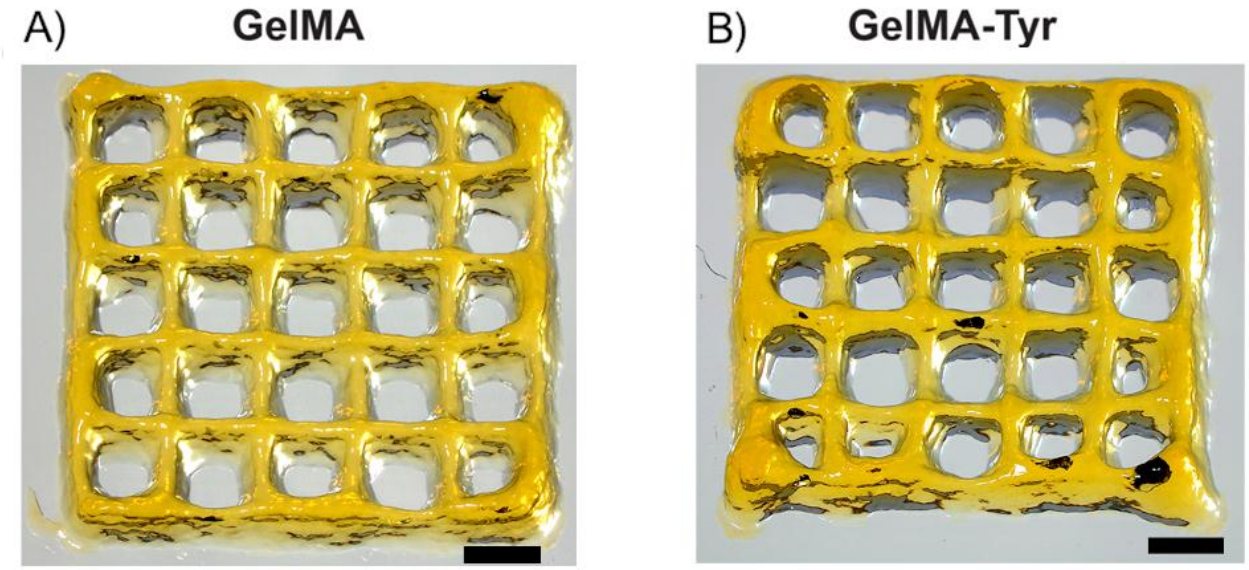
found to be predominant over collagen type I in terms of distribution throughout the hydrogels (Col II/Col I ratio > 1, a threshold indicated by the dotted line), an indicator of cell differentiation towards an articular cartilage-like phenotype. Data are represented as minimum, average and maximum value; * indicates $p<0.05$, n.s. indicates no statistically significant differences.



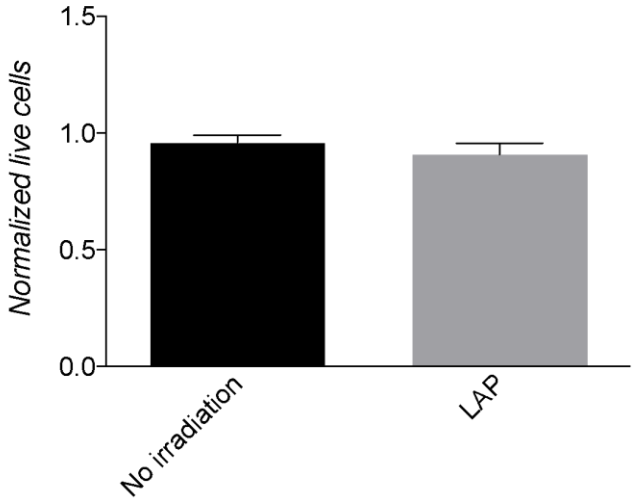
Supplementary figure S7: Metabolic activity of both printed and casted GelMA and GelMA-Tyr hydrogels showing no significant differences, further demonstrating the viability of the encapsulated cells (AlamarBlue assay, calculated as fold increase of values at day 7 over day 1).



Supplementary figure S8: Influence of the collector speed on the diameter of printed GelMA and GelMA-Tyr filaments.



Supplementary figure S9: A) GelMA and B) GelMA-Tyr printed square grids (5x5 mm, designed line spacing = 1 mm), as obtained extruding with a 27G nozzle. Scale bar represents 1 mm.



Supplementary figure S10: Viability of native cartilage cells after exposure to LAP and blue light irradiation (405 nm), showing no measurable cytotoxic effect on the cartilage surrounding the chondral defect in which the LAP-laden GelMA-Tyr precursor was injected.