



**Supplemental Figure 1.** The overall functionality of the integrated, engineered cartilage increased after culture *in vitro* or *in vivo* for 5 weeks. The elastic modulus of cellular hydrogel grafts significantly increased and the elongation property of those samples was also dramatically improved as a function of cellular activity. As shown in Fig.5, the mechanical property of acellular hydrogel grafts consistently demonstrated an energy-storing characteristic with very low elastic modulus (< 200 KPa) and minimal elongating capacity (< 5%). In contrast, the cellular hydrogel grafts, after 5 weeks culture *in vitro* or *in vivo*, increased tensile elastic moduli to approximately 1.3 MPa, approximately 6-9 fold greater than Day-0 values or acellular controls. In parallel, the elongation of the cellular hydrogel grafts was also enhanced to 45-55%, contributed mostly by a non-energy-storing deformation. Along with this mechanical evolution, including elevated elastic modulus and plastic elongation, ideal viscoelastic stiffness was

produced in the integrated, engineered implants. This mechanical evolution demonstrates the competence and longevity of the CS-primed hydrogel-cartilage integration, *in vitro* and *in vivo*, and also confirms the translation of scaffold-tissue bonding to integration and development of functional new tissue with the host tissue.

## Supplemental Methods.

**Calibration of Sputter Rate.** To calibrate the rate at which the CS-adhesive was removed by argon ions a calibration curve was generated by sputtering a gold sample coated with a thin (< 100 nm) film of CS-adhesive. In these experiments the gold sample was first sputter cleaned. The cleaned Au substrate was then immersed into a solution containing the CS-adhesive solution (0.25% w/v, in deionized water) for 2 minutes. Excess solution was removed with a Pasteur pipette before the CS-coated Au sample was placed within the vacuum chamber. The CS-adhesive was then removed by using Ar<sup>+</sup> ions at 1.5kV. Changes in the film thickness during sputtering were determined by monitoring the increase in the Au (4f) XPS peak intensity as a function of Ar<sup>+</sup> sputter time using the following equation:

$$I_{(t=0)} = I_{(t)} \exp\left(\frac{-d}{\lambda \cos \theta}\right)$$

d – CS-adhesive thickness (nm)

$I_{(t)}$  – Au(4f) intensity after Ar<sup>+</sup> sputter time t

$I_{(t=0)}$  – Au(4f) intensity after CS-primer deposition

$\lambda$  - inelastic mean free path of Au(4f) photoelectrons (1.65 nm)

$\theta$  - detection angle (45°)

Cell viability of hydrogel-encapsulated chondrocytes and the cells in the adjacent cartilage explants was qualitatively evaluated by fluorescent Live (calcein, 0.5  $\mu$ mol/mL) - Dead (ethidium, 4  $\mu$ mol/mL) cell assay (Molecular Probes, Eugene, OR).

**Film Characterization using X-ray photoelectron spectroscopy in conjunction with Sputter Depth Profiling.** In depth profiling experiments, the CS-adhesive layer was removed using argon ions generated from a PHI 04-303 ion gun (Ion energy 1.5kV, operating at an emission current of 15 milliamps and 20 millipascals) while XPS followed the evolution of the near surface

region as a function of Ar<sup>+</sup> sputter time. Analysis of XPS spectra was performed with commercially available software<sup>21,35</sup>.

**MRI-T<sub>2</sub> mapping and scanning.** T<sub>2</sub> maps were generated using a multi-echo sequence, utilizing cross-coil operation with: field strength of 1.9 T, TE = 14.06 ms, TR= 2000 ms, Number of echoes = 100, matrix size = 256×256, FOV = 3 cm, based on which weighted T<sub>2</sub> images and curves of T<sub>2</sub> distribution were plotted. Images were obtained 5 weeks after gel implantation.

**Histology and immunohistochemistry.** Constructs were fixed in 4% paraformaldehyde (Sigma Aldrich, St. Louis, MO) solution at 4 °C overnight, paraffin embedded according to standard histological technique and stained with Safranin-O and Masson's Trichrome. Immunohistochemistry for types I and II collagen (antibodies from Research Diagnostics, Flanders, NJ) were performed using Histostain-SP (Zymed, South San Francisco, CA) according to manufacturer's protocol.