

## Supplementary Information

# Porous Silk Fibroin/Cellulose Hydrogels for Bone Tissue Engineering via a Novel Combined Process Based on Sequential Regeneration and Porogen Leaching

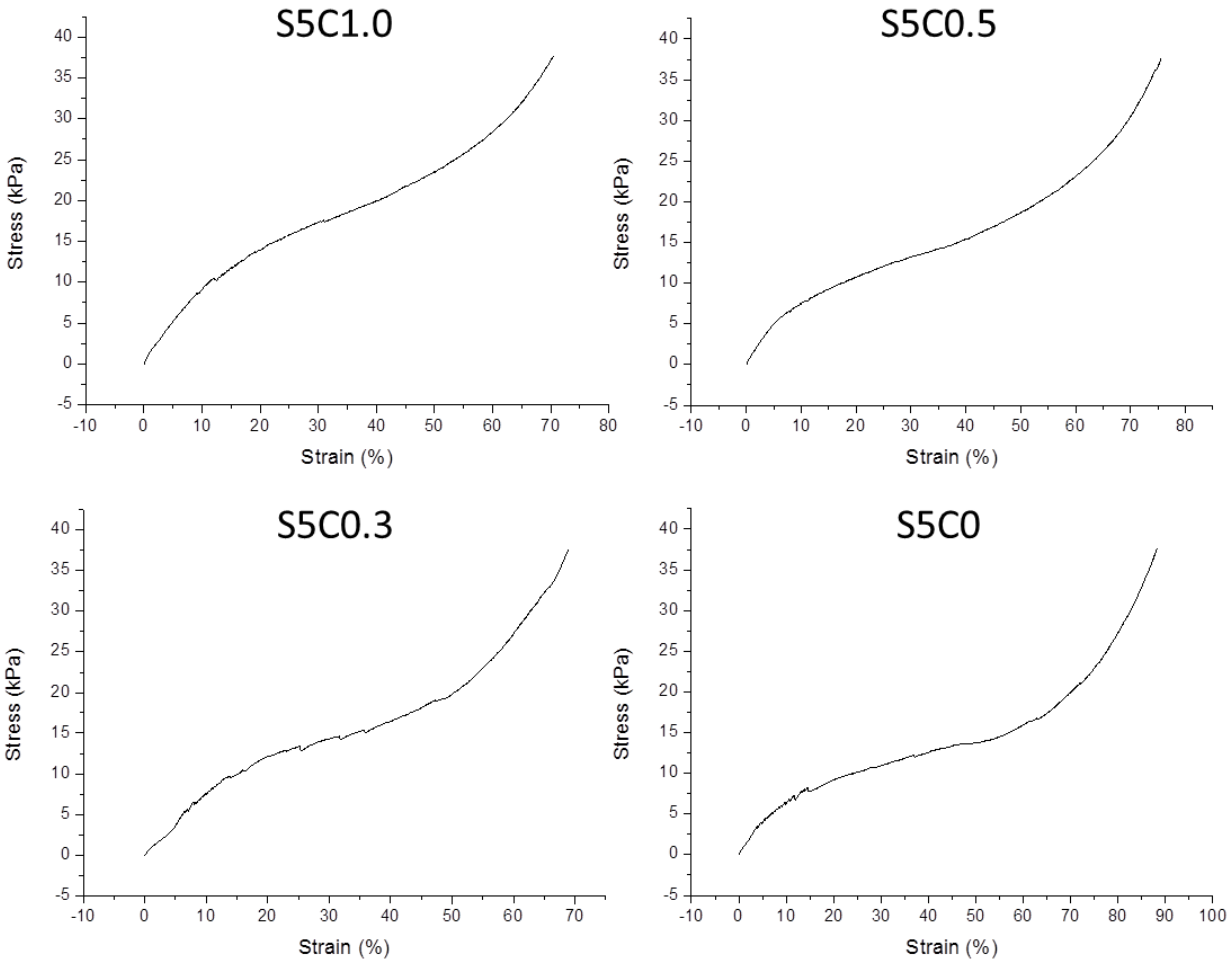
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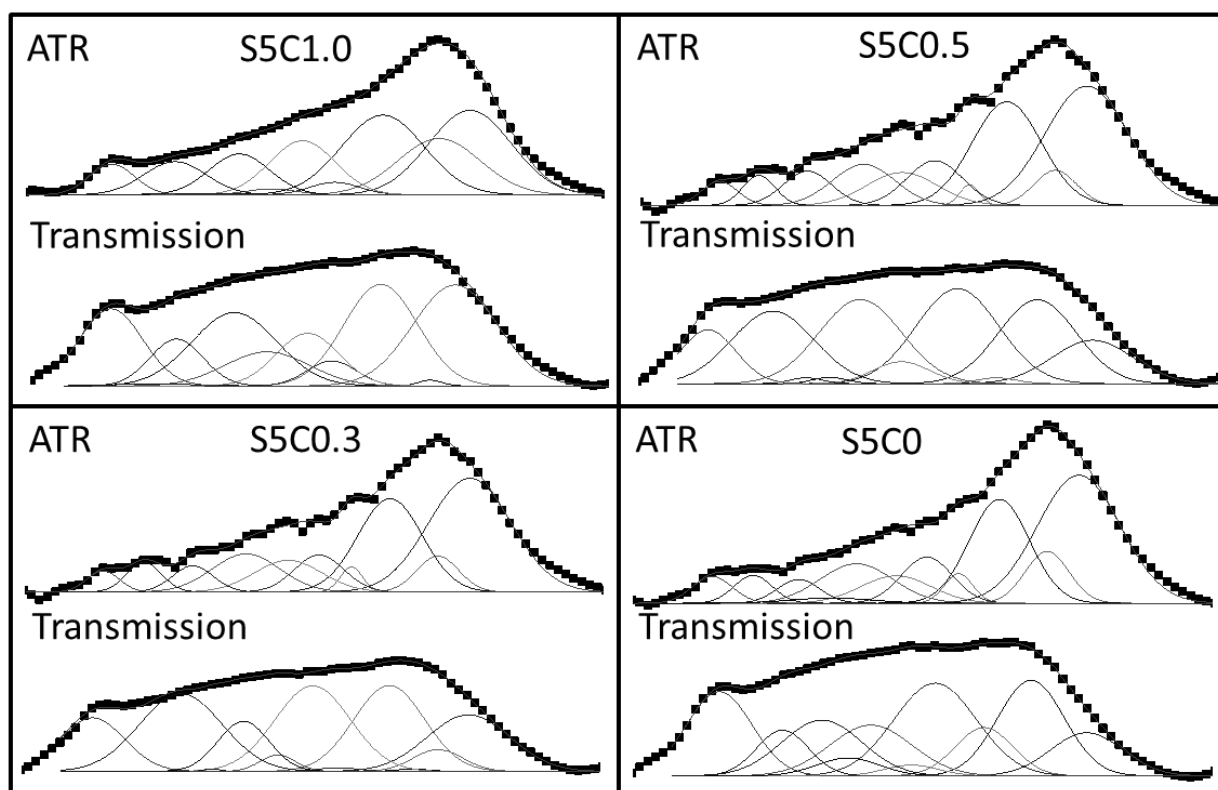
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**Compression test data**



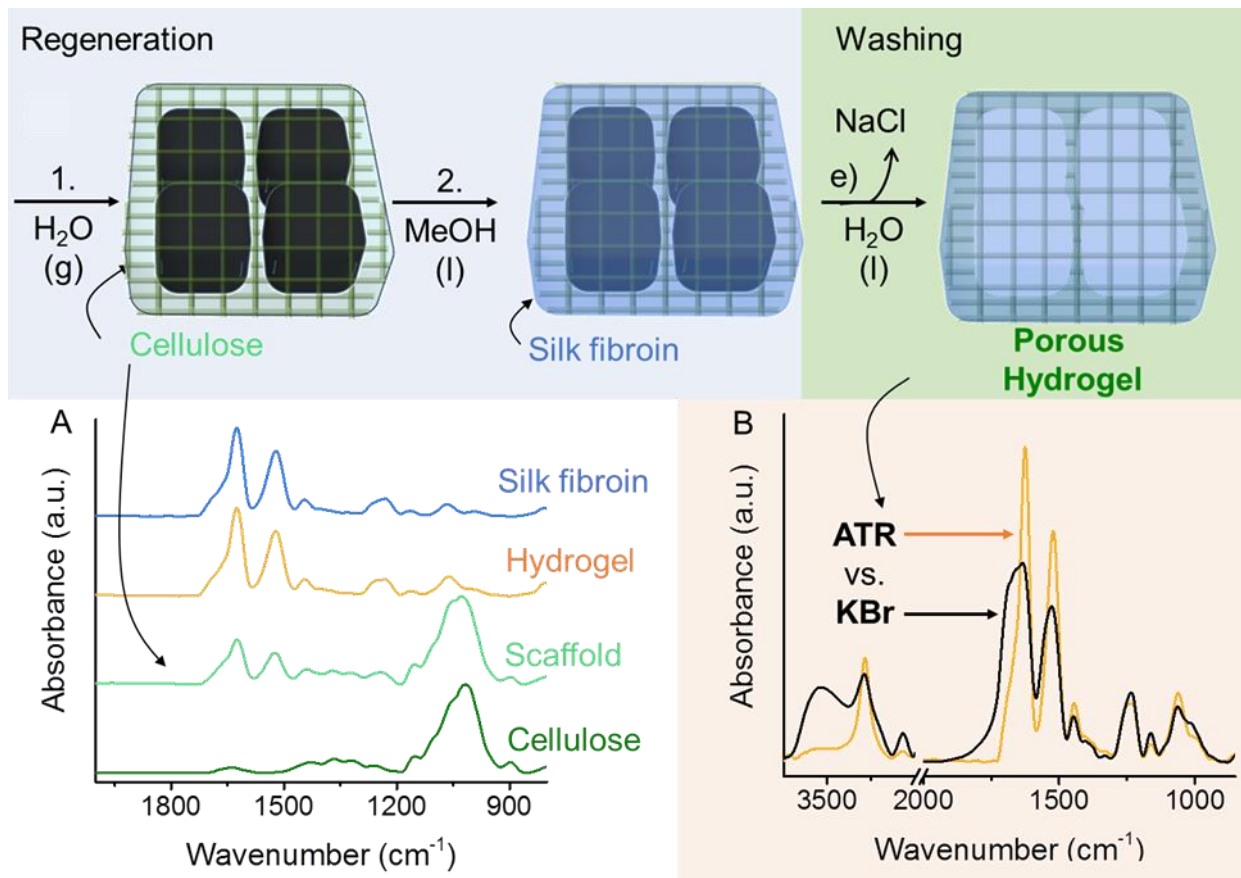
**Figure S1:** Stress-strain curves of all samples and control. Number of samples measured: S5C1.0 n=5; S5C0.5 n=5; S5C0.3 n=6; S5C0 n=4.

## Deconvolution results

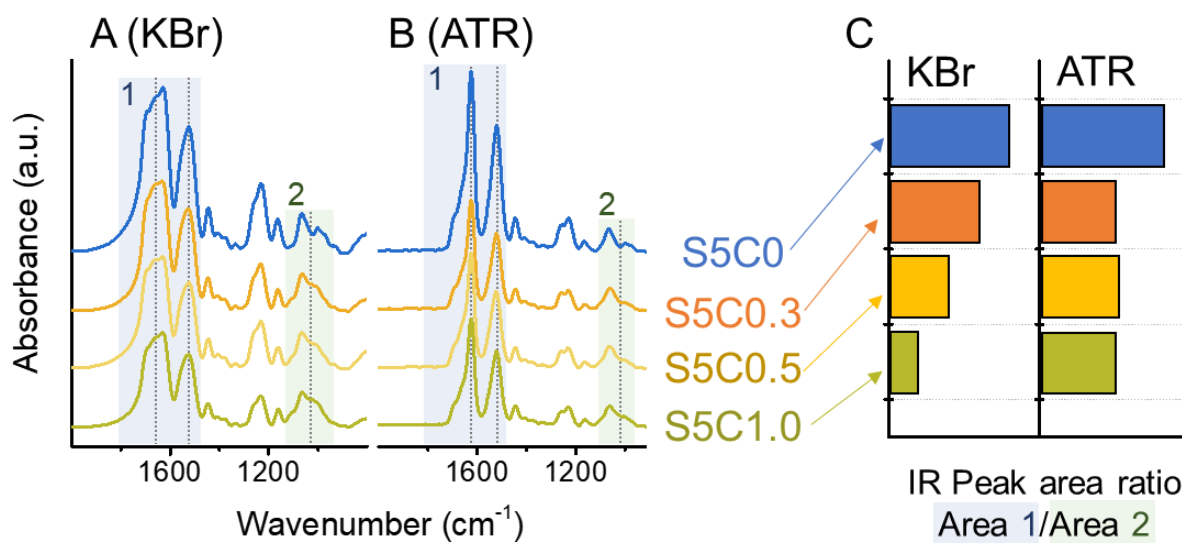


**Figure S2:** Diagrams of deconvoluted peaks in amide I band region. Bands for peak allocation to secondary structure are:  $\beta$ -sheet: 1611  $\text{cm}^{-1}$ , 1619  $\text{cm}^{-1}$ , 1626  $\text{cm}^{-1}$ , 1699  $\text{cm}^{-1}$ .  $\alpha$ -helix/random coil: 1631  $\text{cm}^{-1}$ , 1641  $\text{cm}^{-1}$ , 1649  $\text{cm}^{-1}$ . Turns and bends: 1659  $\text{cm}^{-1}$ , 1668  $\text{cm}^{-1}$ , 1674  $\text{cm}^{-1}$ , 1682  $\text{cm}^{-1}$ .

## SF matrix /cellulose scaffold composite structure



**Figure S3:** Cellulose framework structure embedded in a SF matrix. (A) Cellulose framework: By omitting methanol treatment of samples, a cellulose-rich, very stable hydrogel formed (“Scaffold” spectrum); that sample was compared with regenerated cellulose, degummed silk fibroin and lyophilized SF/cellulose hydrogel by ATR FTIR. (B) SF matrix: ATR and transmission spectra (KBr) normalized to the cellulose-specific band area  $1100\text{ cm}^{-1}$  to  $960\text{ cm}^{-1}$  of a lyophilized hydrogel. The larger amide band area in ATR mode, indicates a relatively increased SF content in the surface-near region compared to the bulk.



**Figure S4:** Different bulk and surface composition. Ratios of silk fibroin (SF) specific amide I and II band area and cellulose-specific band area at 1100 cm<sup>-1</sup> to 960 cm<sup>-1</sup> (respectively area 1 and 2 in A and B) of all samples decrease with increasing cellulose content in transmission mode (KBr) (C). In ATR mode ratios of samples do not differ, but show a lower ratio compared to the SF control. This indicates a richer SF structure in the surface regions of the hydrogel compared to bulk composition.