Figure S1. Typical ATR-FTIR spectra of silk fibroin cryogel networks formed at various TEMED contents indicated. The original data are shown by the filled circles while solid curves are the results of curve fitting for the original spectrum (thick curve) and hidden peaks (thin curves). $T_{\text{prep}} = -18^\circ$C. $C_{\text{SF}} = 4.2$ wt %. EGDE = 20 mmol/g.
**Figure S2.** Stress-strain curves of fibroin scaffolds formed at various temperatures $T_{\text{prep}}$ and EGDE contents indicated. $C_{\text{SF}} = 4.2$ wt %. TEMED = 0.50 %.

**Figure S3.** The compressive modulus $E$ (open symbols) and the compressive stress $\sigma_{\text{comp}}$ (filled symbols) of fibroin scaffolds shown as a function of $T_{\text{prep}}$ (A) and EGDE content (B).
Figure S4. SEM images (A, B) and stress-strain curves (B) of fibroin scaffolds. Drying conditions: At 24°C under vacuum (A) and after freeze-drying (B). $T_{prep} = -18^\circ$C. TEMED = 0.50 %. EGDE = 20 mmol/g. Scaling bars in the images (from left to right): 1 mm, 100 µm and 50 µm.