

Supplementary Information

Silk scaffolds achieved using pickering high internal phase emulsion templating and ionic liquids

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Experimental method

Rheological Testing

The rheological tests were conducted with a Discovery HR-3 hybrid rheometer with a 40mm parallel plate to plate geometry at room temperature which was carefully controlled with a water bath. The frequency sweep test for the HIPES were carried out from the frequency 0.01 to 100 Hz at applied strain $\gamma = 1\%$. The strain sweep test was carried out from the strain 0.1 to 1000% at a set frequency $f = 1\text{Hz}$.

Dynamic mechanical analysis (DMA) Testing

The compressive properties of the freeze-dried porous scaffolds were measured on TA Q800 DMA testing machine in compression mode with a maximum load capacity of 18N. Compressive stress-strain curves for the porous scaffolds were obtained at room temperature at a controlled force rate of 0.1N/min. The average diameter and the height of the scaffolds measured to be 12 mm and 5.5 mm (automatically measured at 0.01N tare load) respectively. Measured compressive strain and strength values are obtained as the mean $\pm$ standard deviation (SD) from five samples.

Table S1a: single step HIPE preparation

Silk Oil:Water	1%	5%	10%
50 : 50	×	×	×
60 : 40	×	×	×
70 : 30	×	×	✓
75 : 25	×	×	✓
80 : 20	×	×	✓
85 : 15	×	✓	✓
90 : 10	×	×	×

1 step HIPE preparation: The required amount of silk particles was suspended in water and sonicated for 1 minute, the required amount of oil was then added and the sample was homogenised for one minute.

Table S1b: two step HIPE preparation

Silk Oil: Water	1%	5%	10%
70 : 30	×	✓	✓
80 : 20	×	✓	✓

2 step HIPE preparation: The required amount of silk particles was suspended in water and sonicated for 1 minute, the required amount of oil was then added to make an emulsion with oil:water ratio of 50:50 the sample was homogenized for one minute. Additional oil was than added and the emulsion was homogenized for one minute. × means no HIPE formation. ✓ means HIPE formation

Table S2: Internal volume fraction as a function of PIL concentration:

PIL Conc:	Internal volume % in Choline TA	Internal volume % in Choline LA	Internal volume % in EOA LA
0.0M	79.59%	79.65%	79.44%
0.5M	79.34%	79.02%	79.25%
1.0M	78.95%	76.74%	75.92%
2.5M	63.64%	60.14%	59.24%

The ionic liquid was added to the water prior to silk particle sonication. The internal volume fraction was systematically calculated as following equation;

$$Internal\ Volume\ \% = \frac{(Oil_{added} - Liquid_{excess})}{(Oil_{added} + Water_{added} - Liquid_{excess})} \times 100\%$$

Where;

Oil_{added} : Initially added amount of oil

Water_{added} : Initially added amount of water

Liquid_{excess} : Amount of liquid (oil and water mix) gathered on top of the HIPE after the stabilisation

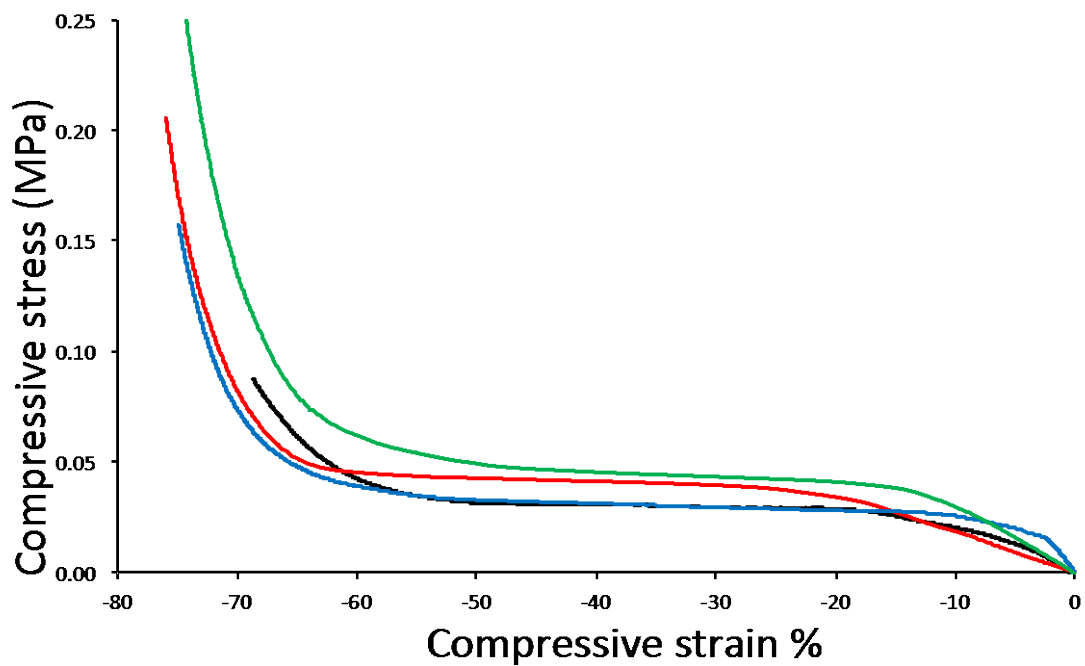


Figure S1: Stress – strain curves for HIEs where Black lines: 0.0 M, blue: 1.0 M EOALa, red: 1.0 M cholineLa and green line 1.0M cholineTa.

Table S3: Compressive stain and strength of freeze- dried silk scaffolds as a function of 1.0M ionic liquid.

Ionic liquid	Compressive strength (kpa)	Compressive strain at failure (%)
0.0 M	32 ± 0.012	56 ± 7.3
1.0 M Choline TA	48 ± 0.026	62 ± 6.1
1.0 M Choline LA	41 ± 0.036	65 ± 4.9
1.0 M EoA LA	34 ± 0.043	63 ± 6.4