Electronic supplementary information for:

Chemical characterisation and fabrication of Chitosan-Silica hybrid scaffolds with 3-glycidoxypropyl trimethoxysilane

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Figure S1 – Solution state ¹H NMR of chitosan and GPTMS in D₂O adjusted to pH 4 with 2M DCl at 5 min, 24 h, 72 h and after 10 days. The methanol peak, resulting from the hydrolysis of the trimethoxysilane groups was observed at all time points at δ^{1} H 3.19 ppm. After 5 min, the multiple peaks around δ^{1} H 0.53 ppm were due to incomplete hydrolysis of the trimethoxysilane groups. After 10 days, the lines become broader, indication condensation of the silane groups and reduced molecular mobility.



Figure S2 – ${}^{1}H$ - ${}^{13}C$ HSQC spectrum of fully hydrolysed GPTMS after 72 h in D₂O/DCl at pH 2.



Figure S3 – 1 pulse ²⁹Si MAS NMR of functionalised chitosan showing only Tⁿ species, as expected. Chitosan-GPTMS at pH 4 functionalised for (a) 5 min, (b) 4 h (c) 24 h and at pH 2 for (d) 5 min (e) 4 h and (f) 24 h.

Sample		Silica species			
рН	Reaction time	T ³	T ²	T1	D _c
4	5 min	48.1±2.6	39.0±2.0	12.9±2.2	78.4
	4 h	44.4±4.3	45.3±3.4	10.2±2.4	78.0
	24 h	37.2±4.0	51.9±4.9	11.0±3.1	75.5
2	5 min	64.8±5.0	35.2±3.6	-	88.3
	4 h	44.2±2.6	55.8±3.9	-	81.4
	24 h	60.5±3.8	39.5±2.9	-	86.8

Table S1 – Quantified ²⁹Si MAS NMR of functionalised chitosan showing highly condensed Tⁿ species with the degree of condensation remaining approximately constant over time.



Figure S4 - TGA traces showing mass loss due to combustion of the organic component of 50 wt% organic scaffolds at 0, 72, 168, and 672 h in SBF. The relative organic composition of the hybrids remains constant throughout the timescale of the study.