**Supplementary Information**

### 2.1 Preparation of native and crosslinked chitosan fibers

Preliminary rheological characterization was performed to determine fiber forming ability of chitosan. Gelation kinetics of chitosan in alkaline condition (5% w/v NaOH) was assessed using Bohlin CVO Rheometer (Malvern Instrument, UK) with parallel plate geometry (20 mm diameter spindle) maintaining a gap of 500 µm. Briefly, complex viscosity (η∗) of chitosan upon addition of NaOH solution (from Merck, India), pH 13, was evaluated with time sweep measurement in an oscillatory mode at constant amplitude (0.01%) and frequency (1Hz) at 25°C. Blends of chitosan with DHF (Sigma Aldrich) at three different concentrations (3, 5 and 10 v/v%) were placed on rheometer plate and time sweep viscosity measurement was carried out to evaluate influence of DHF concentration on gel forming ability at 25° C. Further, complex modulus measurement of chitosan–DHF blends were carried out as a function of temperature at constant frequency and shear stress of 1 Hz and 10 Pa, respectively, under linear visco-elastic region (LVR) upto 80 °C.

#### 2.2.2 Crosslinking density, Swelling and Biodegradation study

Ninhydrin (2, 2-Dihydroxyindane-1, 3-dione) assay was performed to determine degree of crosslinking of chitosan fibers. In this assay, crosslinked and uncrosslinked chitosan fibers were boiled with ninhydrin (SRL Pvt. Ltd., India) solution and optical absorbance of the solution was recorded at 595 nm with a spectrophotometer (Recorders and Medicare Systems, India). Glycine (Merck, India) at various known concentrations was used as standard. Crosslinking density of the fibers was calculated as per the following equation.

$$\% \text{ Crosslinking density} = [(Cb-Ca)/Cb] \times 100,$$

Where, Cb, Ca are the optical absorbance at 595 nm for chitosan fibers before and after crosslinking, respectively.

Swelling behavior of chitosan fibers was measured in terms of change in weight as a function of immersion time. Dry scaffolds were weighed and soaked in phosphate-buffered solution, PBS (pH 7.4) at 37°C. They were taken out after 24 h and blotted dry with filter paper and again weighed. The percentage of swelling was calculated using the following formula-

$$\% \text{ Swelling} = [(\text{wet weight} - \text{dry Weight})/ \text{dry weight}] \times 100$$

Three replicates were performed and averaged.

*In vitro* degradation of crosslinked and uncrosslinked fibers was performed in PBS, pH 7.4 at 37 °C containing 1.5 µg/ml lysozyme (hen egg-white, Sigma-Aldrich) at specified time intervals (7, 14, 21 and 28 days).

### 3.1 Preparation of native and crosslinked chitosan fibers

The complex viscosity of 6 wt % chitosan upon addition of 5% w/v NaOH solution increased instantaneously and reached a plateau in 400 s (S1a). Time sweep viscosity measurements of chitosan in presence of DHF with constant shear rate (10 $s^{-1}$) at 25 °C demonstrated absence of gelation which was also evident by insignificant change in viscosity (data not shown). In stark contrast, temperature sweep measurement revealed rapid increase in viscosity due to progressive liquid-gel transition at elevated temperature. Complex modulii of the blends increased significantly with increasing temperature and reached to maxima beyond 80 °C. Further, complex modulus was found to be strongly dependent on DHF concentration. However, complex modulii values were marginally higher for gels formed in 10% v/v DHF in comparison to 5% v/v during reaction progression. The increase in complex modulus with rise in temperature and concentration of crosslinker was mainly due to high degree of crosslinking of polymer by butenedial formed in situ through furan ring opening under acidic condition. Similar result was reported by Johnson et al. during gelation of alumina slurries using 1.5% chitosan solution and 10-100 mM DHF.[12] To assess gelation kinetics, elastic (G′) and viscous (G″) modulus of the chitosan blend using 5% v/v DHF was measured as shown in inset. The study clearly indicated that the G′ values increased significantly in comparison to G″ values mainly due to gel formation**.**

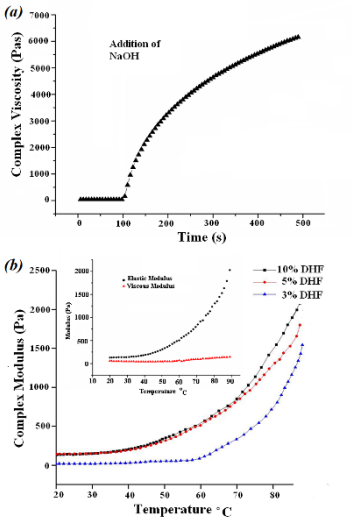


Fig. S1a Complex viscosity of 6 wt% chitosan after addition of 5% w/v NaOH at 25 ℃ as a function of time. Fig. S1b Complex modulus of chitosan-DHF blends under temperature sweep (0-80 ℃) at a constant frequency (1Hz) and shear stress (10Pa). b) Inset shows elastic and viscous modulus of 6 wt% chitosan with 5% v/v DHF blend as a function of temperature.

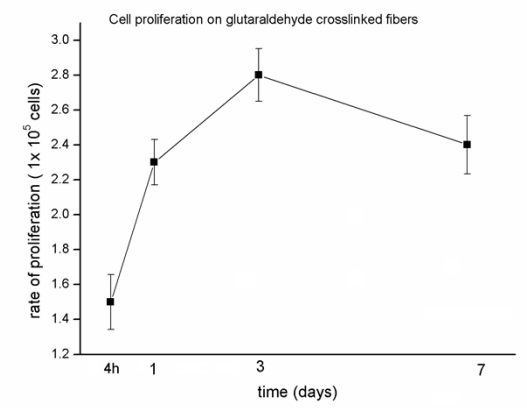


Fig. S2 MTT of hMSCs on chitosan fibers crosslinked with glutaraldehyde.