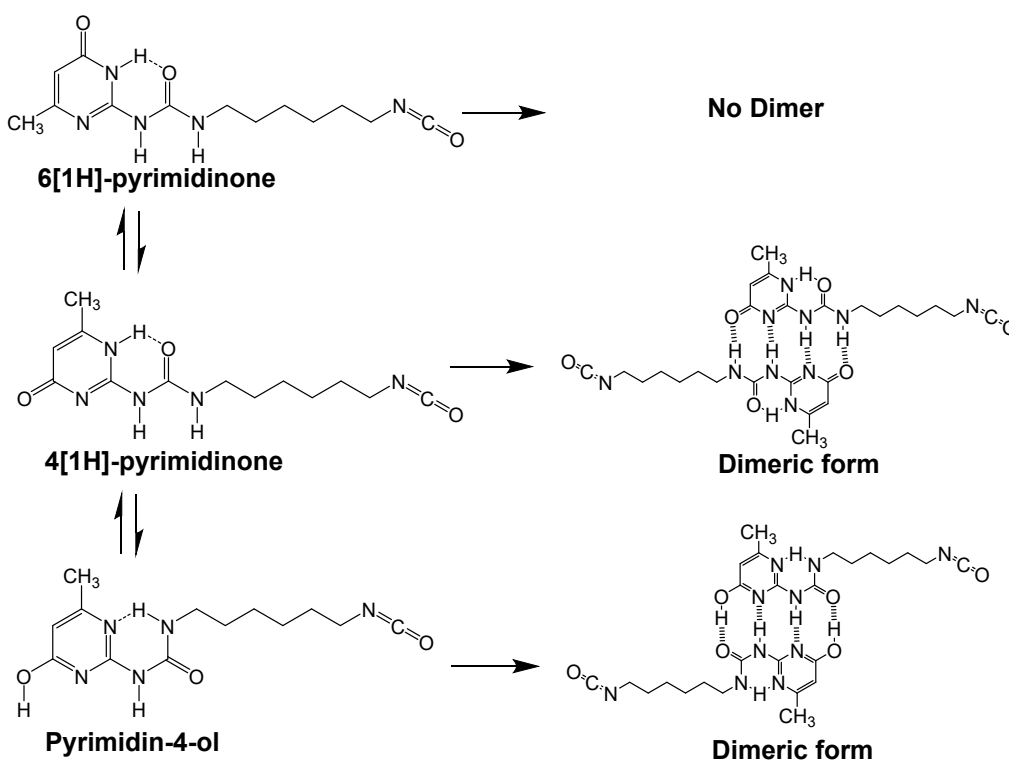


Supporting Information

Synthesis and fabrication of gelatin-based elastomeric hydrogels through cosolvent-induced polymer restructuring

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1. Tautomeric forms of ureidopyrimidinone



2. Synthesis of Ureidopyrimidinone-synthon

A typical procedure from an earlier report was used to synthesize ureidopyrimidinone or 2(6-isocyanatohexylaminocarbonylamino)-6-methyl-4[1H] pyrimidinone. 2g (.0159 mol) of 2-amino-4-hydroxyl-6-methyl pyrimidine and 21.5g (0.128 mol) of 1, 6-diisocyanatohexane was mixed in anhydrous pyridine (70ml, 0.906 mol) and heated under continuous stirring at 100°C for 16 hours in N₂ atmosphere. Further pentane (200ml) was added to precipitate the white powder, which was further filtered and washed with acetone three times. The obtained white powder was dried in vacuum at 50°C. Product was vacuum distilled to remove the excess 1, 6-diisocyanatohexane from the product [1].

¹H NMR δ 13.15 (s, 1H, CH₃CNH), 11.89 (s, 1H, CH₂NH(C=O)NH), 10.22 (s, 1H, CH₂NH(C=O)NH), 5.86 (s, 1H, CH=CCH₃), 3.33 (m, 4H, NH(C=O)NHCH₂ + CH₂NCO), 2.2 (s, 3H, CH₃C=CH), 1.62 (m, 4H, NCH₂CH₂), 1.44 (m, 4H, CH₂CH₂CH₂CH₂CH₂). ATR-FTIR 1667.5, 1699, 2267, 3216 cm⁻¹.

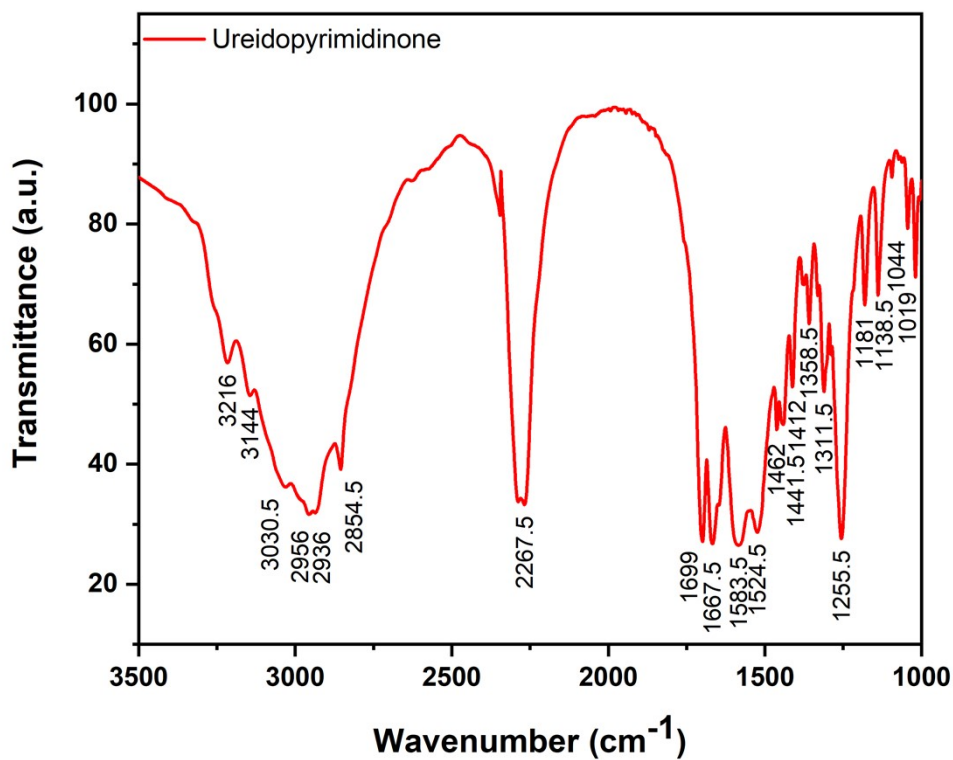


Figure S1. FTIR spectrum of Ureidopyrimidinone-synthon

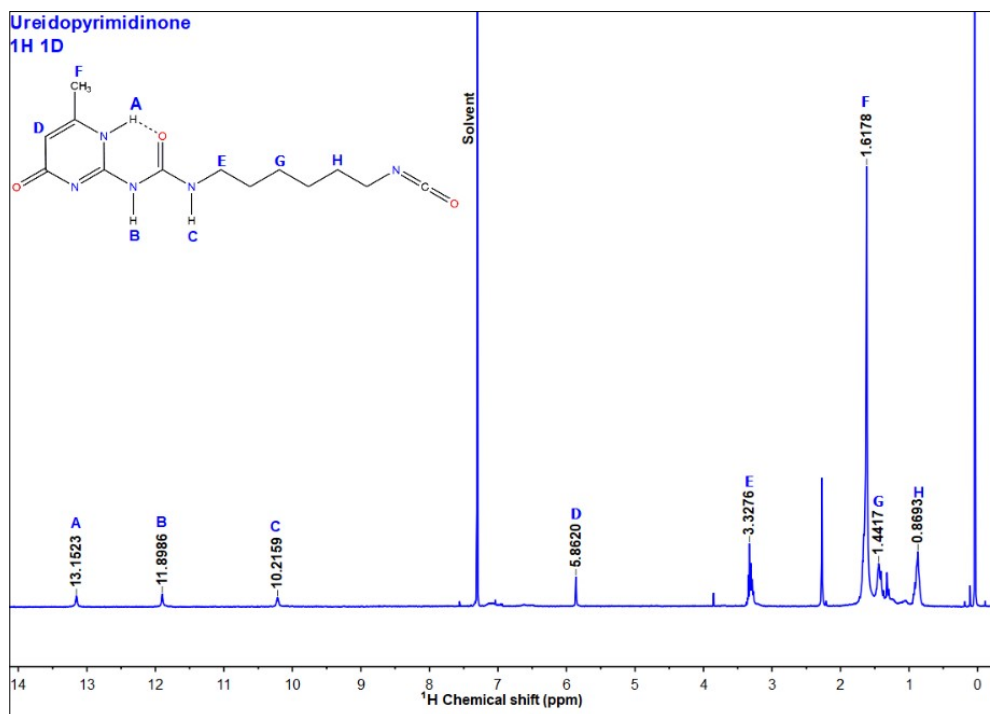


Figure S2. ¹H NMR spectrum of ureidopyrimidinone (Upy-synthon)

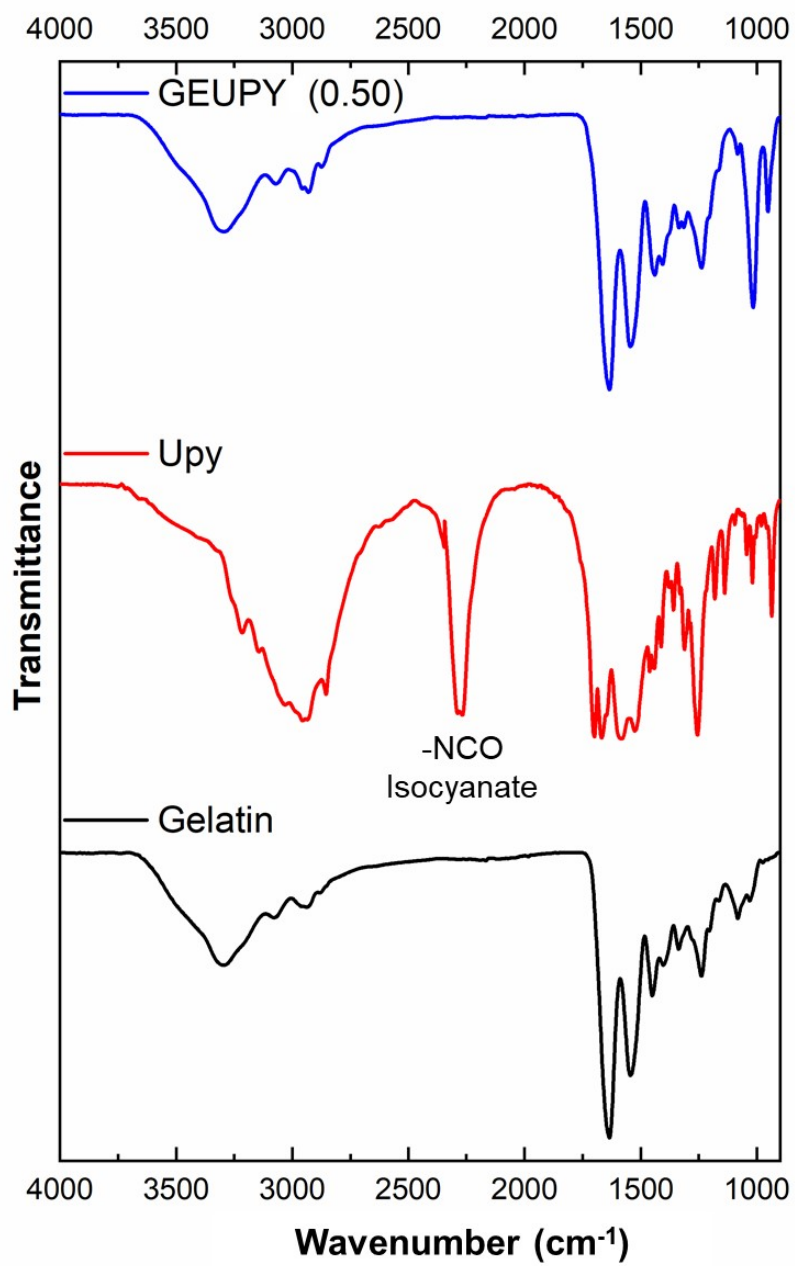


Figure S3a. Complete FTIR spectrum of GEUPY (0.50), Upy-synthon and gelatin

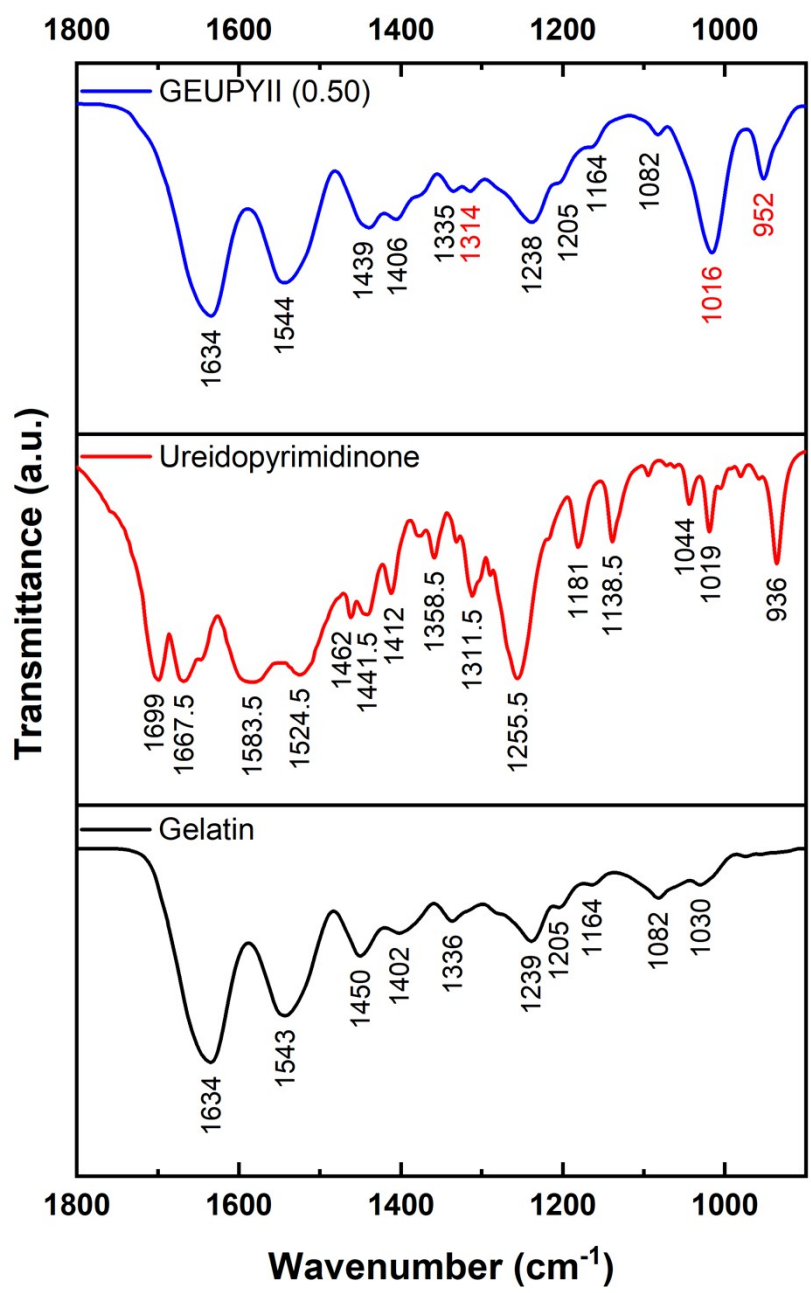


Figure S3b. FTIR spectrum of GEUPY (0.50), Upy-synthon and gelatin

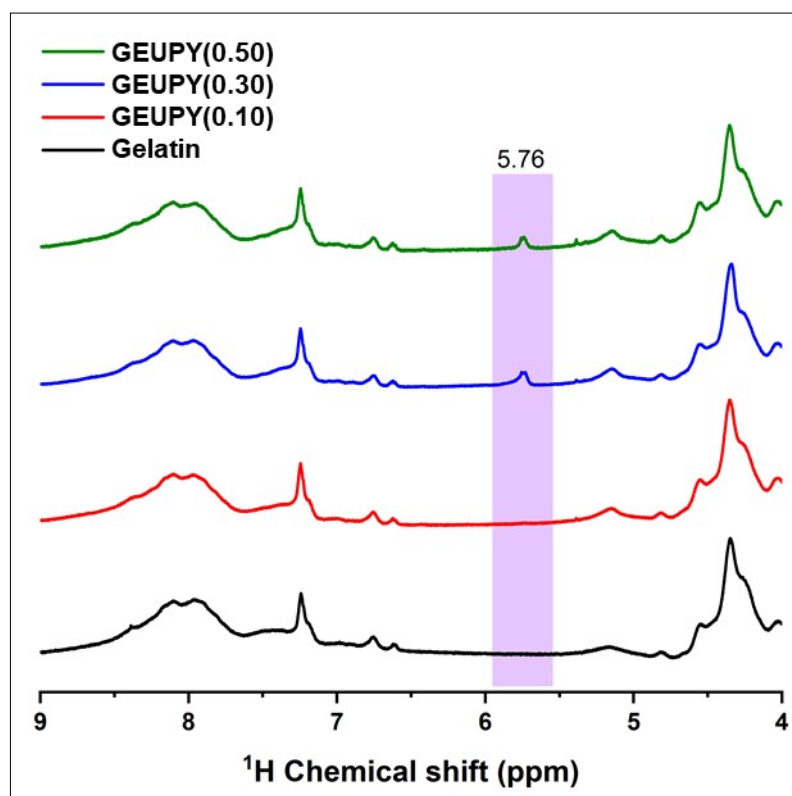


Figure S4. ^1H NMR spectrum of gelatin and Upy substituted gelatin derivatives (GEUPY (0.10), GEUPY (0.30) & GEUPY (0.50))

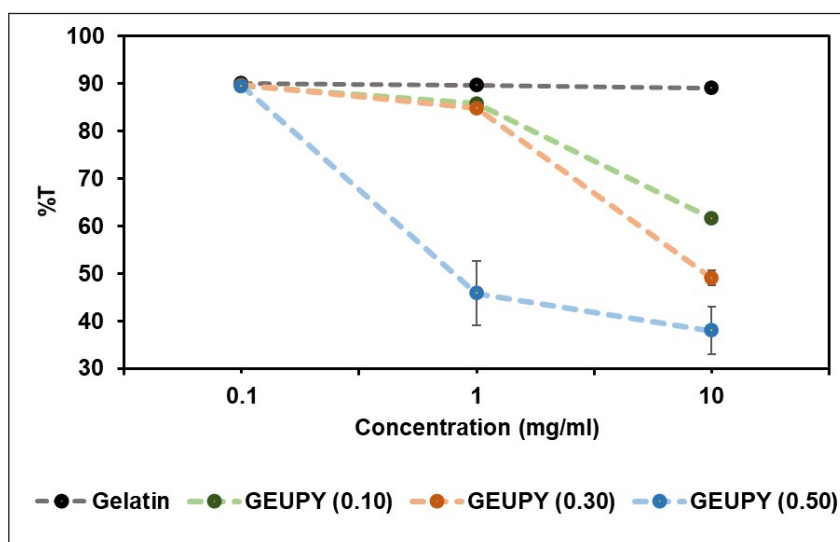


Figure S5 Concentration dependent transmittance analysis of gelatin and GEUPY samples

3. Microscopical image analysis of GEUPY samples

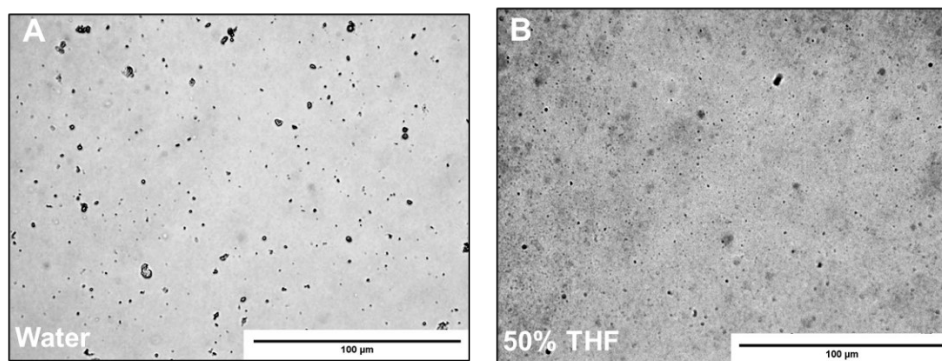
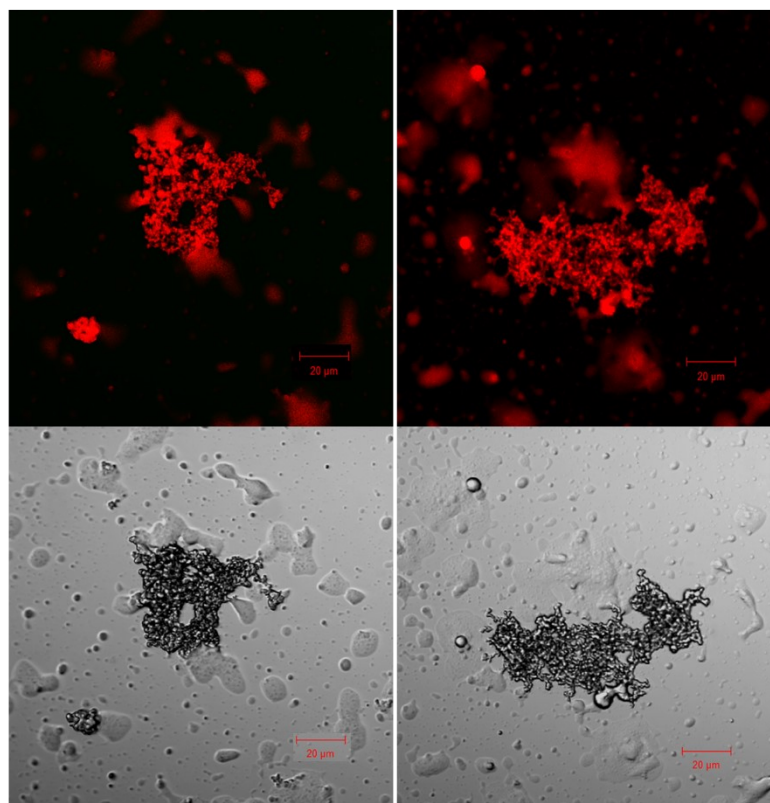
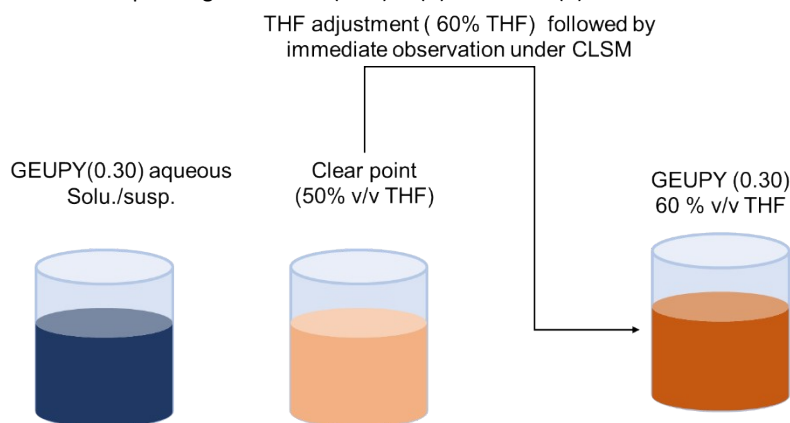


Figure S6 Optical microscopic images of GEUPY(0.50) in (A) water and (B) THF



FigureS7. CLSM images of Nile red stained sample of GEUPY (0.50) in wet state at 60% THF (1mg/ml)

4. Film analysis

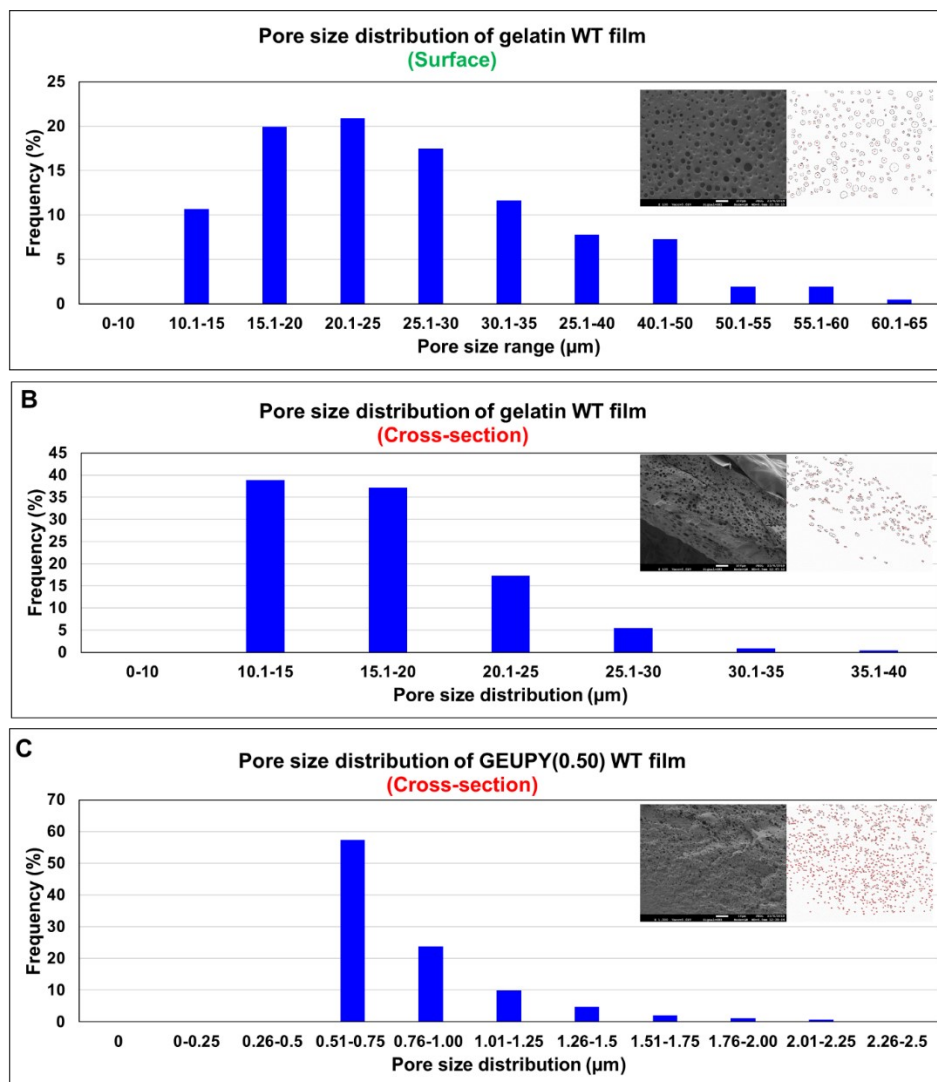


Figure S8 – Pore size distribution of gelatin and GUEPY(0.50) 80%THF film

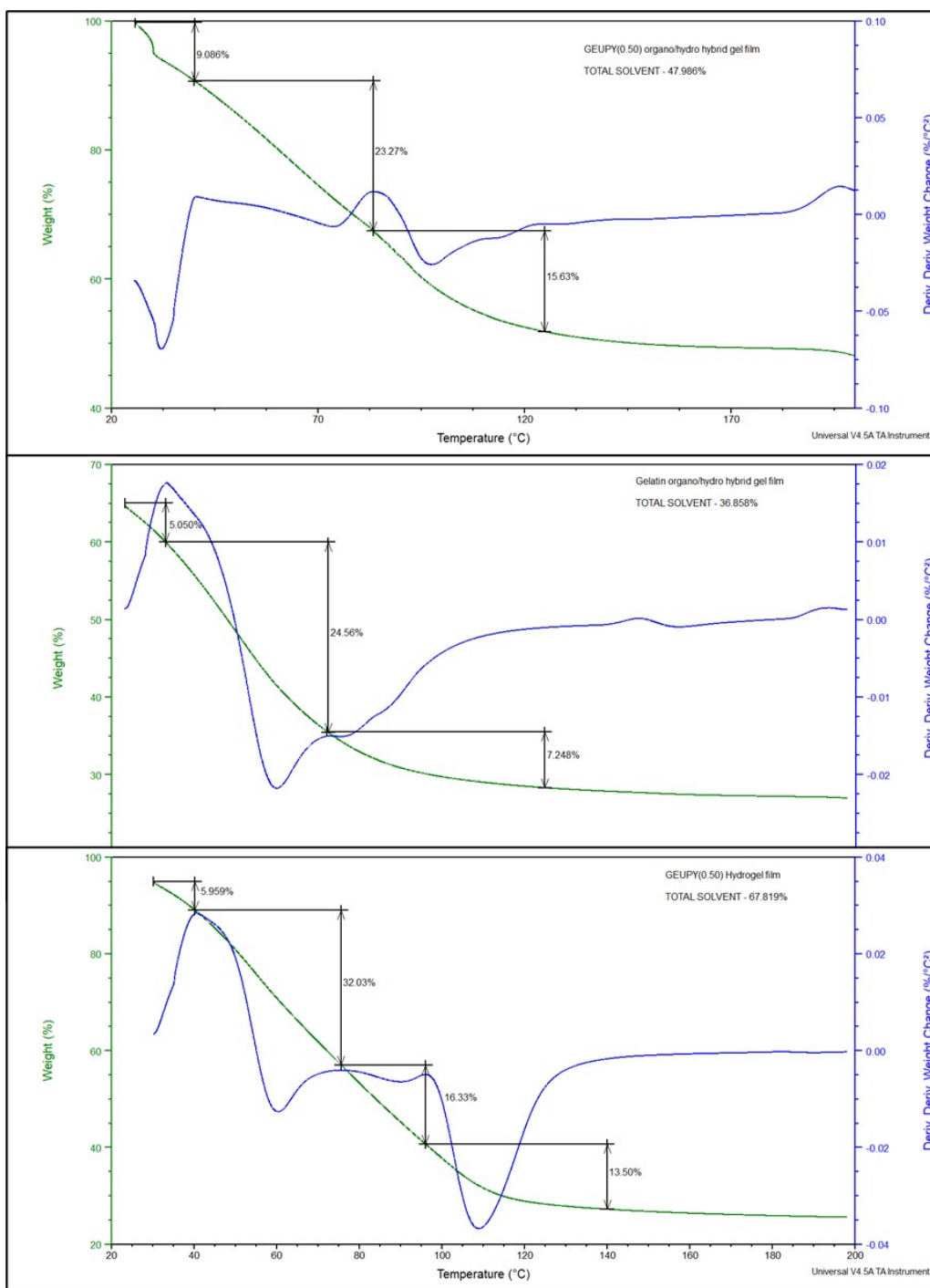


Figure S9a. Thermal gravimetric analysis of GEUPY(0.50) organo/hydro hybrid gel films, gelatin organo/hydro hybrid gel films and GEUPY(0.50) hydrogel films

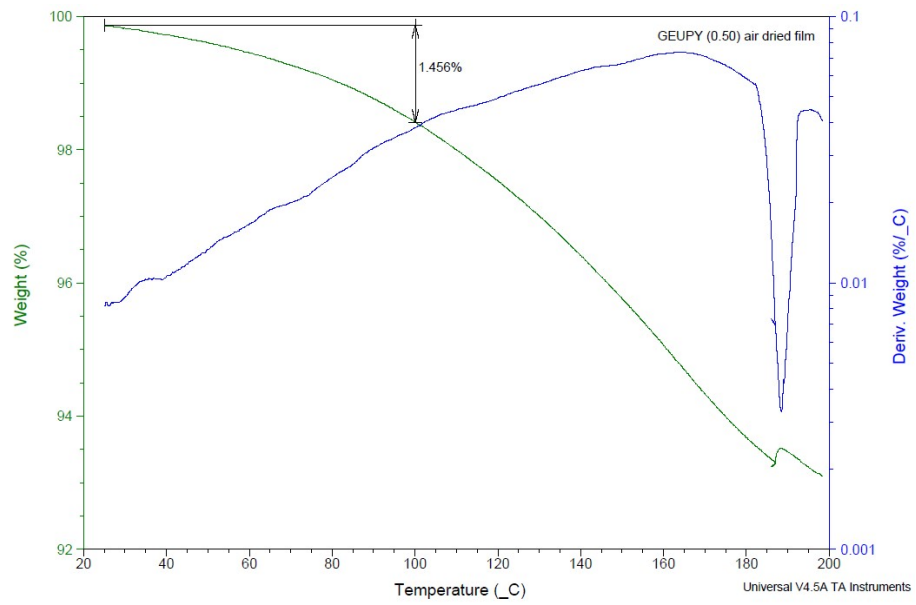


Figure S9b. TGA analysis of air-dried films for solvent content

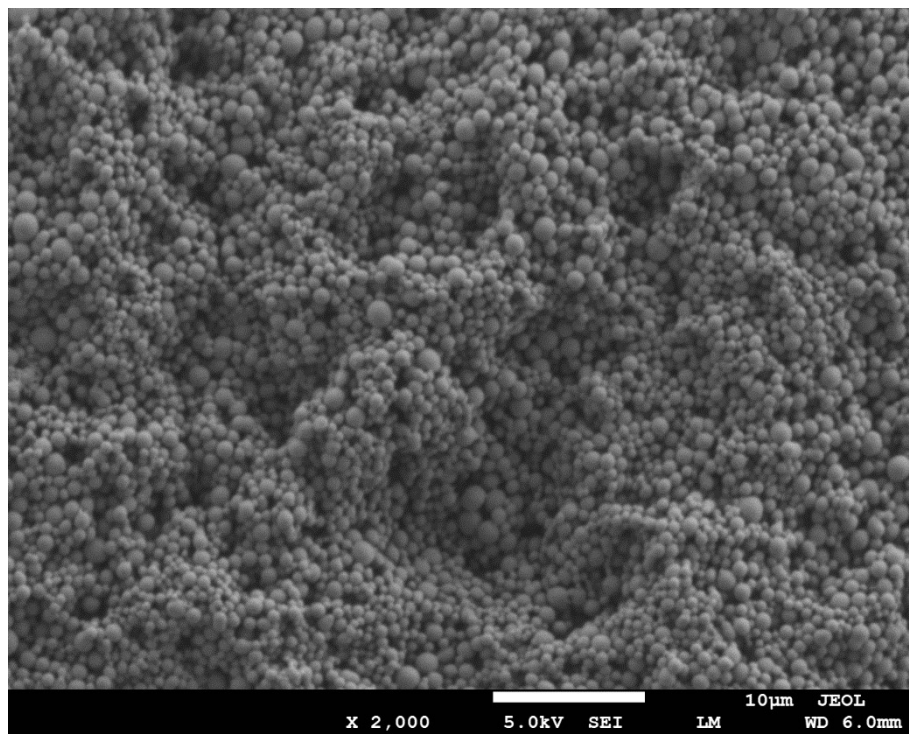


Figure S10. SEM image of gelatin at 80% ethanol

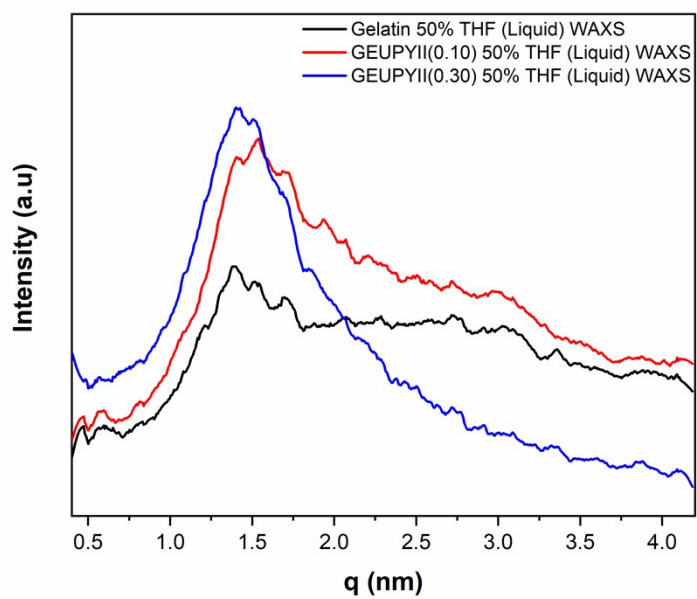


Figure S11 – WAXS of gelatin, GEUPY (0.10), GEUPY (0.30) at 50% THF

1. Folmer, B.J.B., et al., *Supramolecular Polymer Materials: Chain Extension of Telechelic Polymers Using a Reactive Hydrogen-Bonding Synthron*. *Advanced Materials*, 2000. **12**(12): p. 874-878.