Supporting Information

## Catechol Functionalized Elastin-like Polypeptides as Tissue Adhesives

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Materials:

Protein modification -- dimethyl sulfoxide (DMSO) – Fisher Scientific, BP231-100; N,Ndimethilformamide (DMF) – Fisher Scientific, D119-500; dopamine hydrochloride – Sigma, H8502; N-(3-dimethylaminopropyl)-N'-ethylcarbodiimide hydrochloride (EDC) – Sigma-Aldrich, E7750; N-hydroxysuccinimide (NHS) – Aldrich, 130672; hydrochloric acid – VWR, VW3110-3, triethylamine (TEA) – TCI America, T0424

Mechanical testing -- cyanoacrylate glue – Permabond 105; sodium periodate – Sigma-Aldrich, S1878

Cell substrate preparation -- (3-aminopropyl)triethoxysilane (APTES) – Aldrich, 440140; sulfuric acid – Fisher Scientific, A300C-212; hydrogen peroxide – Fisher Scientific, H325-100; glass coverslips – Hampton Research, HR3-277; Toluene – Fisher Scientific, T290-1

Cell culture -- DMEM with 4.5 g/L glucose, L-glutamine and sodium pyruvate – Corning, 10-013-CV; HyClone Calf Serum – GE Healthcare, SH30073.03; penicillin-streptomycin – Gibco, 15140-122; DPBS without calcium and magnesium – Corning, 21-031-CV; 0.25% trypsin with 0.1% EDTA – Corning, 25-053-CI; Bovine Albumin (BSA) – Affymetrix, J10857-18; Phalloidin-iFluor 488 – Abcam, ab176753; DAPI – Invitrogen, D21490; Triton X-100 – Fisher Scientific, BP151-500

**Supplementary Figures:** 

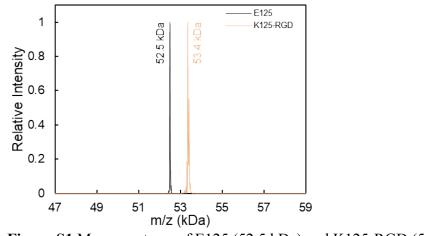
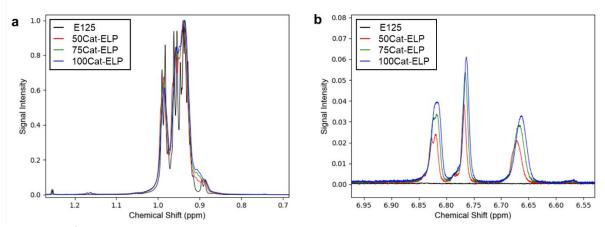
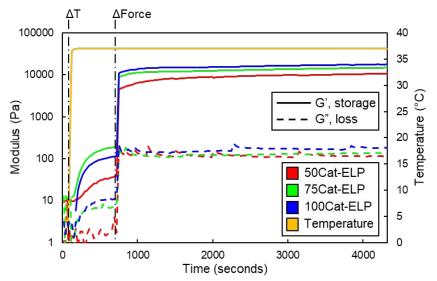


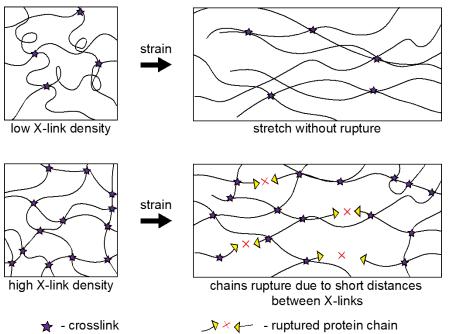
Figure S1 Mass spectrum of E125 (52.5 kDa) and K125-RGD (53.4 kDa)



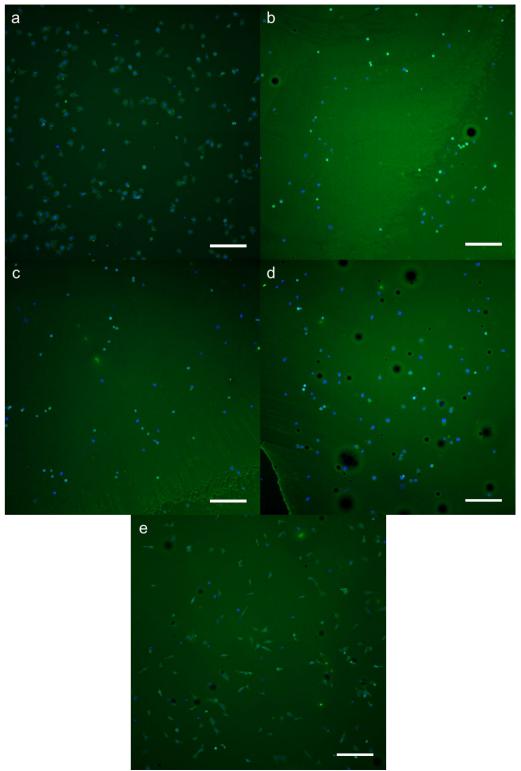
**Figure S2** <sup>1</sup>H NMR signal from valine methyl groups is used to quantify dopamine: a) Signal from protons on valine methyl groups; b) Signal from catechol functional groups. 50Cat-ELP showed 90% reaction yield, while 75 Cat-ELP showed 86% and 100 Cat-ELP showed 79% reaction yield



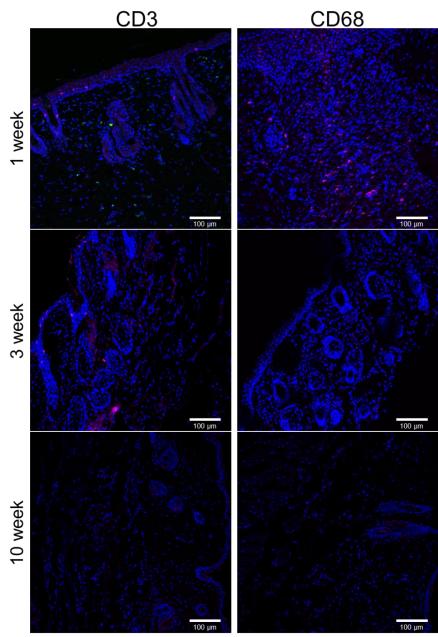
**Figure S3** Representative rheology test data; Rheology tests for Cat-ELP hydrogels initially show negligible moduli at 4°C. The moduli increase slightly when the temperature is ramped ( $\Delta$ T) to 37°C; however, because Cat-ELP deswells upon crosslinking, it has poor contact with the probe until the application of 0.2 N normal force ( $\Delta$ Force). As shown, after the normal force is applied, the storage and loss moduli values reflect the hydrogel properties



**Figure S4** Schematic showing the effects of crosslink density and strain. 50Cat-ELP hydrogels show higher flexibility because a larger portion of the polymer chain is available for stretching compared to highly crosslinked 100Cat-ELP



**Figure S5** Representative images of fibroblasts on coated glass substrates: a) Control - APTES coated; b) 50Cat-ELP; c) 75Cat-ELP; d) 100Cat-ELP; e) 100Cat-RGD. Nuclei are stained blue with DAPI and cell cytoskeleton is stained with phalloidin-iFluor 488 (Scale bars: 100 µm)



**Figure S6** Representative images of tissue sections from sham controls at weeks 1, 3 and 10. Similar to the 100Cat-ELP and 100Cat-RGD samples, the control samples were stained for CD3 and CD68. CD68 signal is observed at the 1 week, which indicates the presence of macrophages.