

Figure S1. ^1H NMR was used to determine the successful synthesis of Cat-8PEG and determine the functionality ratio. Characteristic peaks at $\delta = 6.59$ ppm, 6.54 ppm and 6.40 ppm are representative of the DOPA catechol group functionalization. Cat8PEG was obtained as a solid (3.35 g) with 74 % functionalization. ^1H NMR (Cat8PEG, 300 MHz, DMSO-d_6 (*)) δ : 8.73 (s, 8H, OH), 8.63 (s, 8H, OH), 7.20 (m, 8H, NH), 6.59 (m, 8H, C_6H_4), 6.53 (m, 8H, C_6H_4), 6.39 (m, 8H, C_6H_4), 4.10-4.02 (m, 32H, $\text{CH}_2\text{CH}_2\text{OCONH}$), 3.85-3.35 (m, PEG protons). 3.15-3.05 (m, 32H, NHCH_2CH_2). Cat-8PEG was stored under vacuum.

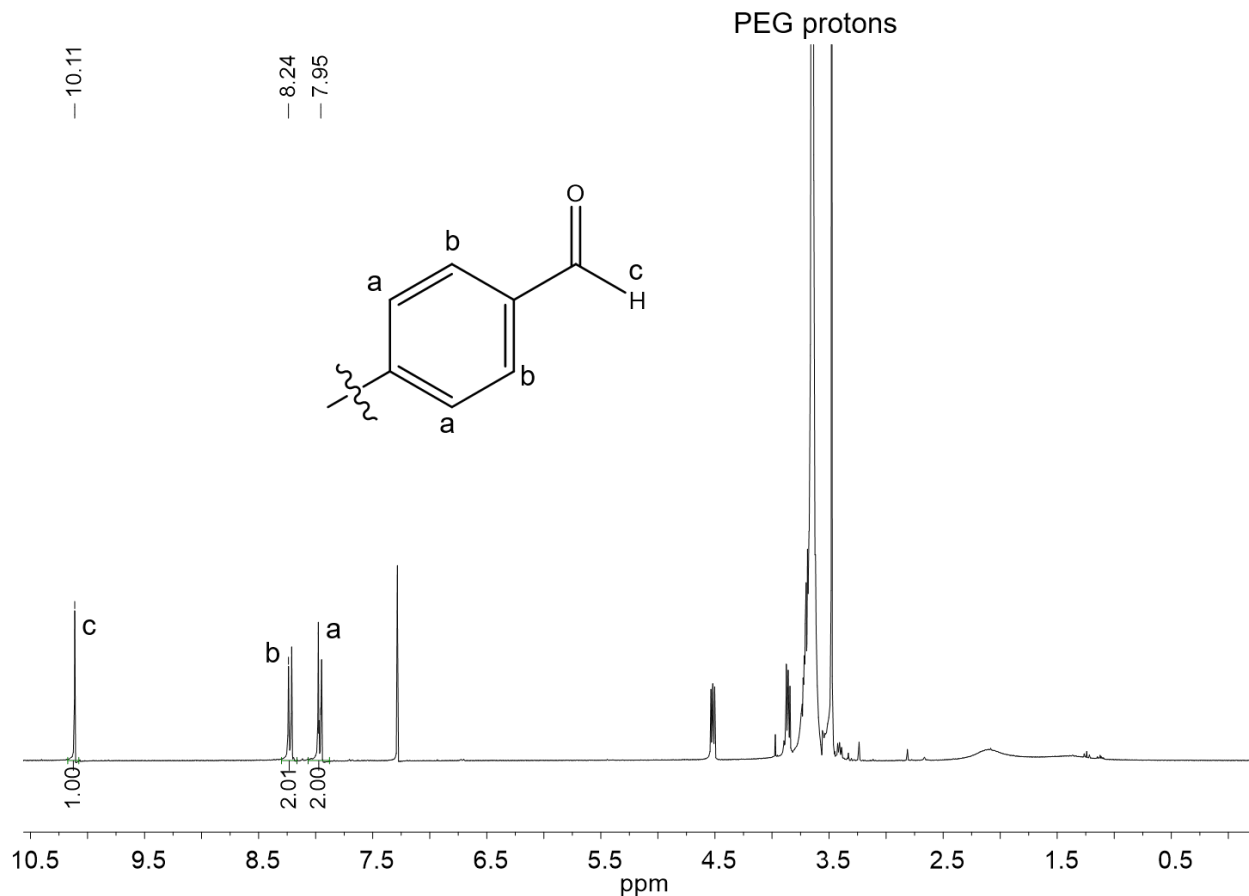


Figure S2. ¹H NMR was used to determine the successful synthesis of Ald-8PEG and determine the functionality ratio. Characteristic peaks at $\delta = 10.11$ ppm, 8.24 ppm and 7.95 ppm are representative of the benzaldehyde functionality. Ald-8PEG was obtained as a white solid with 97 % functionalization. ¹H NMR (Ald-8PEG, 300 MHz, CDCl₃) δ : 10.11 (s, 8H, CHO), 8.24 (m, 16H, C₆H₄), 7.95 (m, 16H, C₆H₄), 4.49 (m, 16H, CH₂OCHO), 3.85-3.35 (m, CH₂CH₂OCHO and PEG protons). Ald-8PEG was stored under vacuum until use.

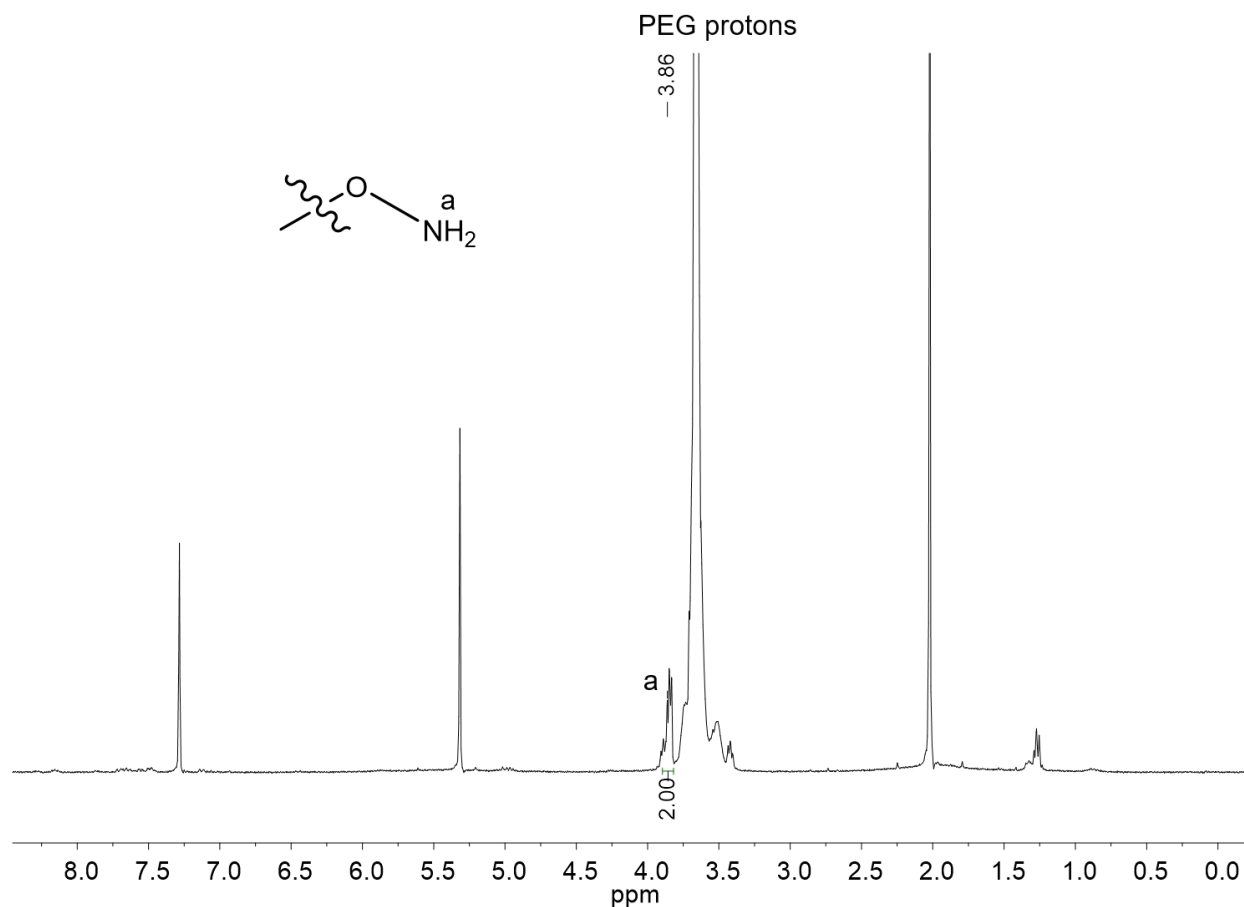


Figure S3. ¹H NMR was used to determine the successful synthesis of AO-8PEG and determine the functionality ratio. The characteristic peak at $\delta = 3.86$ ppm represents the aminoxy functionality. AO-8PEG was obtained as a white solid with 83 % functionalization. ¹H NMR (AO-8PEG, 300 MHz, D₂O) δ : 3.86 (m, 2H, CH₂ONH₂), 3.8-3.4 (m, PEG protons). AO-8PEG was stored under vacuum until use.

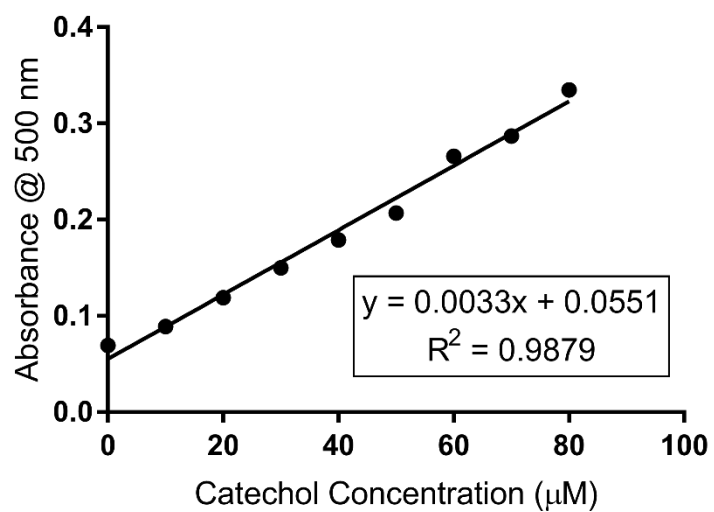


Figure S4. A standard curve was generated using known concentrations of 3,4-dihydroxy-L-phenylalanine for quantifying catechol functionalization in Cat-8PEG. Absorbance was recorded at 500 nm.

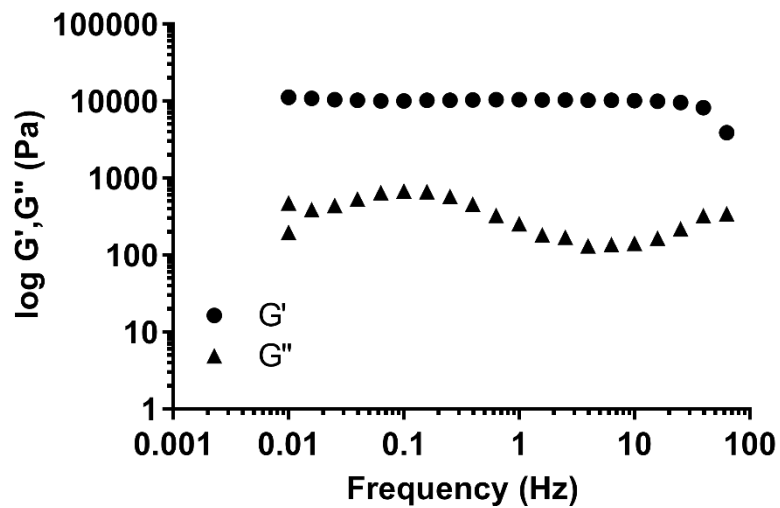


Figure S5. Representative G' and G'' across a frequency sweep. This graph is representative of the viscoelastic behavior of both the Ald-AO and Ald-AO-Cat gels which crosslinked and gelled within seconds of combining the two solutions prior to rheometry.

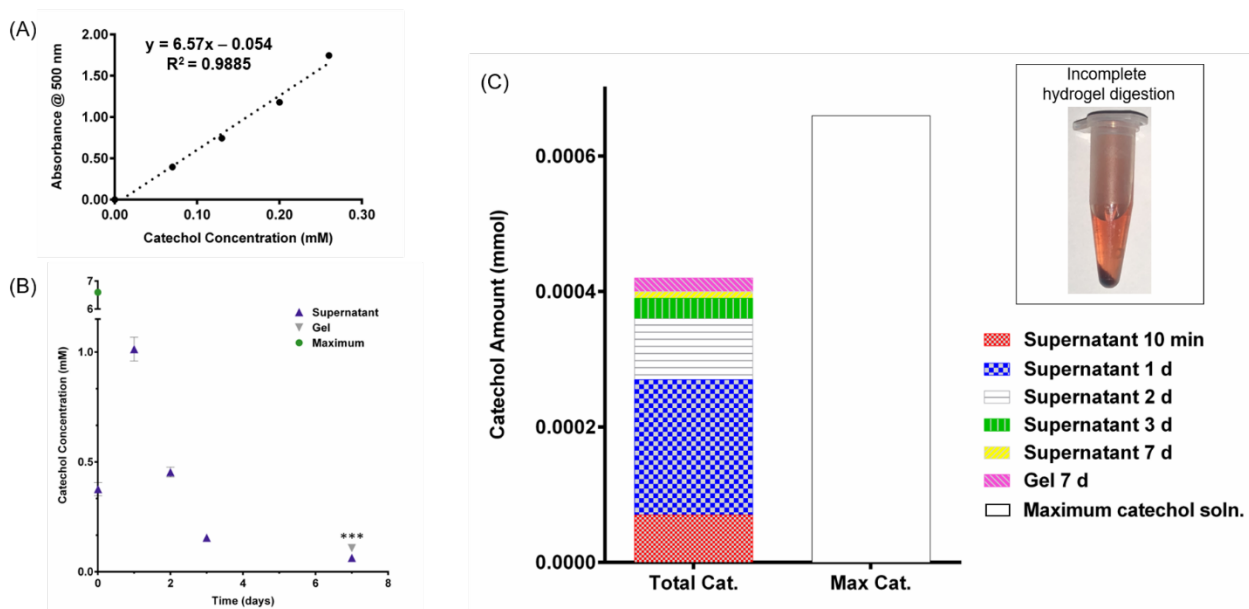


Figure S6. Catechol is retained in the Ald-AO-Cat as indicated by the time-dependent DOPA-nitration assay. (A) Release of Cat-8PEG was detected in gel supernatant at 10 min, 1 day, 2 days, 3 days, and 7 days post-gelation. An initial release of Cat-8PEG was observed at 10 min, corresponding to the release of Ald-8PEG and AO-8PEG observed in the ex vivo retention assay (Figure 2F). This is indicative of the release of unreacted or non-physically trapped materials. A burst release of Cat-8PEG was detected, similar to the release of Ald-8PEG and AO-8PEG, which was expected with maximum swelling of the oxime hydrogel in aqueous conditions. A time-dependent, decrease in Cat-8PEG release was calculated up to day 7, with a significantly larger amount of Cat-8PEG physically trapped in the gel at 7 days post gelation. (B) The summation of Cat-8PEG released in the supernatant and retained in the gel at day 7, was compared to the total Cat-8PEG concentration in the original gel formulation, with 63 % of the total Cat-8PEG accounted for. The remaining Cat-8PEG can be explained by the incomplete digestion of the gels at day 7 resulting in incomplete release of Cat-8PEG for detection.

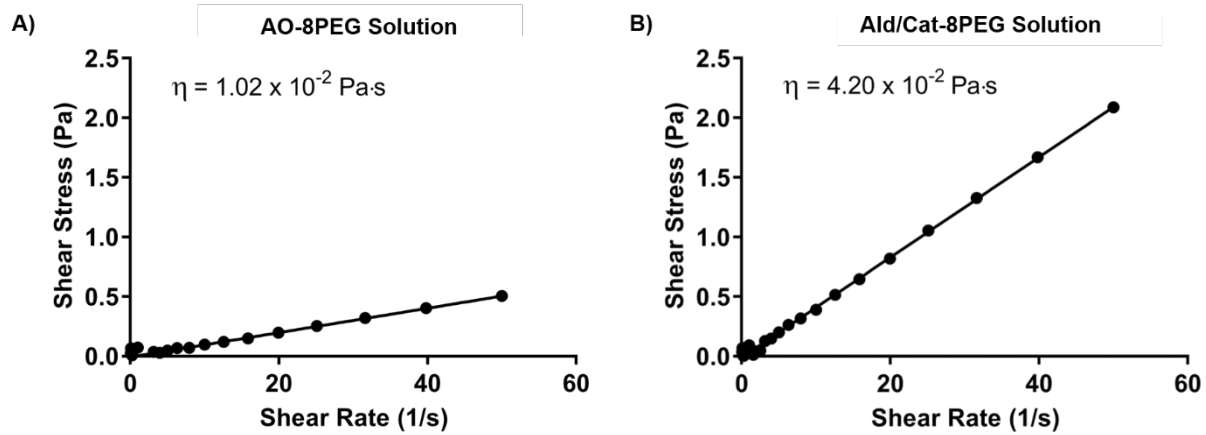


Figure S7. Parallel plate rheometry was used to measure the viscosity (η) of the Ald-8PEG/Cat-8PEG (A) and AO-PEG (B) solutions.

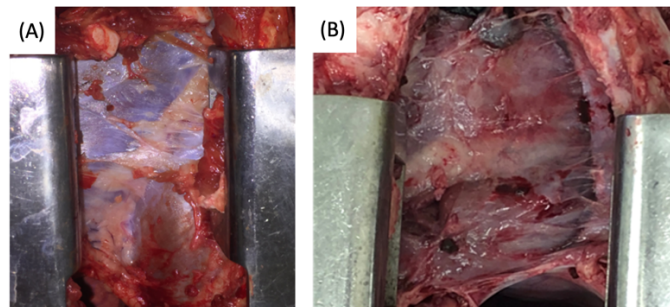


Figure S8. Representative images from pilot pig study showing robust adhesions in the control group (A) that required sharp dissection, whereas Ald-AO-Cat treated pigs (B) had milder adhesions that required only manual/blunt dissection. B displays Ald-AO-Cat treated pig euthanized at 3 weeks after application.