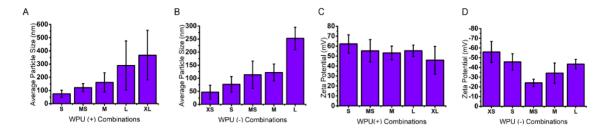
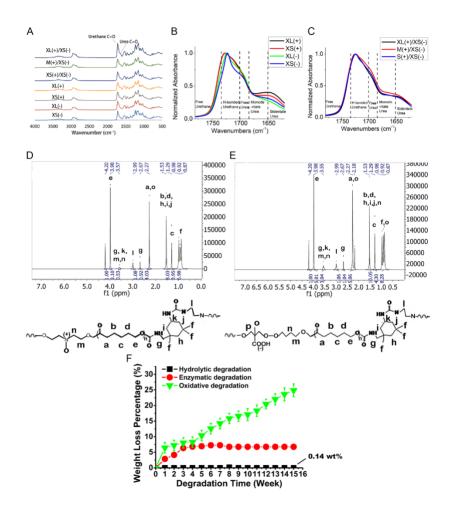


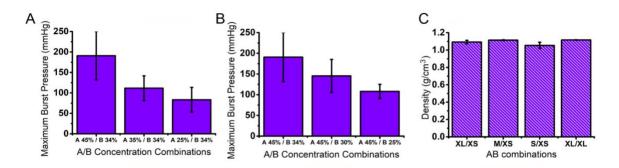
Figure S1 Tilt test of WPU (+), WPU (-), and B-Seal, showing the fast-gelling of B-Seal within 10s after mixing WPU(+) and WPU(-).



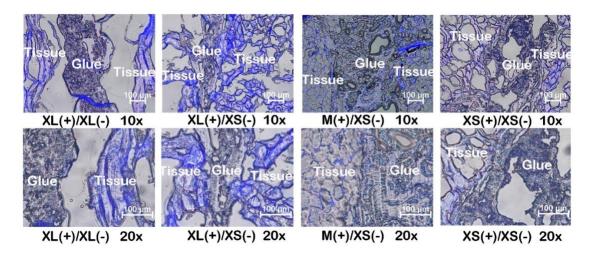
**Figure S2** Average particle sizes of (A) WPU (+) and (B) WPU (-) and average zeta potential of (C) WPU (+) and (D) WPU (-) for B-Seals nanodispersions characterized by Zetasizer. (n=3)



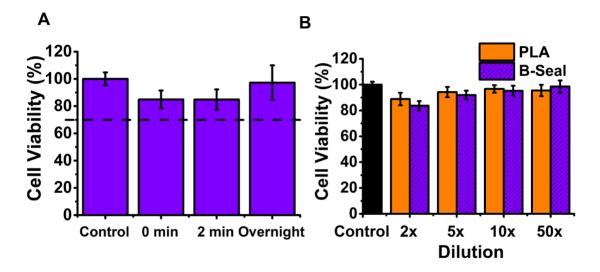
**Figure S3** Characterization of B-Seal. (A) The FTIR spectra of XL and XS WPU (+) films, XL and XS WPU (-) films, and B-Seal films by mixing XL, M, S WPU (+) with XS WPU (-) films respectively. (B, C) FTIR spectra in the carbonyl region for (B) single phase WPU films and (C) B-seal films formed by various size pairs of WPU (+) and WPU (-) nanodispersions. NMR spectra of (D) WPU (+) and (E) WPU (-). (F) The in vitro degradation profiles of B-Seal under hydrolytic (in PBS), enzymatic (in PBS containing 1 U/mL of cholesterol esterase) and oxidative (in PBS containing 20 wt% hydrogen peroxide (H2O2) and 0.1 M cobalt chloride (CoCl2)) degradation at 37 °C.



**Figure S4** Burst strength and density of B-Seal. (A, B) Burst strengths of B-Seal with various concentrations of (A) WPU (+) and (B) WPU (-). (C) Density measurements of various B-Seal film. (n=5)



**Figure S5** Microscopic observations at the interface of porcine tissue and B-Seal formed with mismatched particle sizes of WPU(+) and WPU(-) nanodispersions.



**Figure S6** *in vitro* Cytotoxicity of B-Seal. (A) Cell viability of L929 fibroblasts cultured with MEM elution of B-Seal for 24 h according to ISO10993-5. MEM elution were collected by incubating B-Seals gelled at 0 min (right after mixing), 2 min, and overnight with MEM for 24 hours; (B) Cell viability of L929 fibroblasts cultured with B-Seal degradation products at various dilutions according to ISO10993-5. B-Seal samples were degraded in 0.2 M NaOH, followed by pH adjustment to neutral using 1M HCI. (n=5)

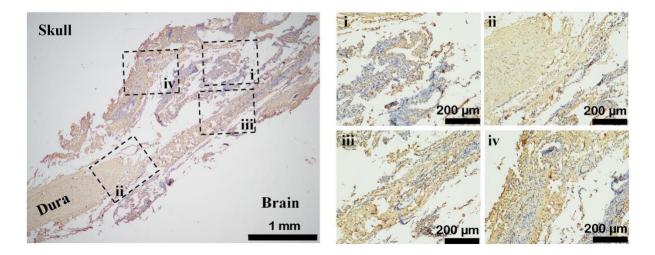


Figure S7 Type I collagen staining of the harvested dura mater after 12 weeks of B-Seal implantation.