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Supplementary Information for

Instant tough bioadhesive with triggerable benign detachment

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Movies S1 to S2



Fig. S1. Schematic illustrations for the different timescales of adhesion and the corresponding requirement for triggerable benign detachment of the bioadhesive.



Fig. S2. Chemical schemes for the synthesis of functional monomer.



Fig. S3. ¹H NMR spectra for synthesized NHS ester functionalized monomer with a disulfide bond.



Fig. S4. The number of adhered fluorescent microbeads on the bioadhesive 5 min after incubation in varying solutions in Fig. 2*C-F*. Values represent the mean and the standard deviation (n = 4). *P* values are determined by a Student's *t*-test; ns, not significant (p > 0.05); * $p \le 0.05$; ** $p \le 0.01$.



Fig. S5. Mechanical testing setups for interfacial toughness measurements based on the standard 180-degree peel test (ASTM F2256).



Fig. S6. Effect of pH on the adhesion performance. (*A*) Various pH values in human body. (*B*) Interfacial toughness between the bioadhesive and wet porcine skin tissues incubated in various pH-adjusted PBS. Values in (B) represent the mean and the standard deviation (n = 3). *P* values are determined by one-way ANOVA and Tukey's multiple comparison test; ns, not significant (p > 0.05).



Fig. S7. The efficiency of the triggerable detachment of the bioadhesive. (*A*) Interfacial toughness between the bioadhesive and wet porcine skin tissues without triggering and 1, 5, 10, and 30 min after the application of the triggering solution. (*B*) Representative force/width vs. displacement curves for the 180-degree peel tests. Values in (A) represent the mean and the standard deviation (n = 4). *P* values are determined by a Student's *t*-test; ns, not significant (p > 0.05); * $p \le 0.05$.



Fig. S8. (*A*-*C*) Representative force/width vs. displacement curves for the 180-degree peel tests of short-term (A), intermediate-term (B), and long-term (C) adhesion in Fig. 2*H*-*J*.



Fig. S9. Effect of triggerable detachment and re-application of bioadhesive on the adhesion performance. (*A*) Schematic illustrations for triggerable detachment and re-application of the bioadhesive. (*B*) Interfacial toughness between wet porcine skin tissues and the bioadhesive originally applied and re-applied on the same tissue after triggerable detachment. Values in (B) represent the mean and the standard deviation (n = 3). *P* values are determined by a Student's *t*-test; ns, not significant (p > 0.05).

Movie S1. Triggerable benign detachment and re-application of bioadhesive for sealing of *ex vivo* porcine lung.

Movie S2. Triggerable benign detachment of bioadhesive device from beating *ex vivo* porcine heart.