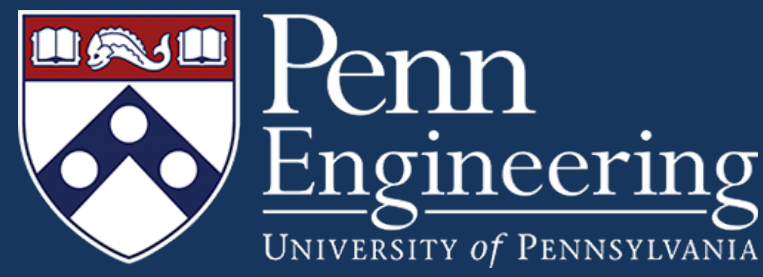


A chronically stable polymer adhesive for bioelectronics

J. Brock Horton, Yewei Huang, Yaxin Fan, Yuanwen Jiang
hortonjb@seas.upenn.edu



Abstract

In biomedical applications, strong, long-term adhesion onto wet tissue is necessary for device-to-biological tissue positioning. Current methods often employ the use of crosslinked hydrogels that swell and lose mechanical and adhesion properties over time during *in vivo* conditions. To overcome the above limits of hydrogel biointerfaces, this poster aims to design a chemically-stable polymer adhesive to achieve a chronically stable and strong adhesive for biomedical device applications. The long-term stability of this polymer adhesive is not compromised by either dehydration or excess swelling, which are the two main shortcomings of conventional hydrogels. Therefore, it shows a promising future for long-term implantable applications in biomedical devices.

Background

- Tissue adhesion is challenging to be achieved due to the existence of interfacial water layer and high water content of the tissue
- Long-term stability and toughness of wet-tissue adhesion are important for device-to-biological tissue placement.
- Adhesion occurs through absorbing water around the tissue to achieve adhesion [1] and creating bonds to the tissue through intermolecular and/or intramolecular bonds [2].
- Hydrogel systems fail over time due to over-swelling, leading to the degradation of mechanical integrity and loss of adhesion [3].

Proposed solution for adhesion

- Conventional hydrogels match the elastic moduli to the underlying tissue and provide adhesion, but are unstable under dynamic wet *in vivo* conditions.
- Polymer matrices like polydimethylsiloxane (PDMS) are chemically-stable, but are not as soft and adhesive as hydrogels.
- By tuning the modulus of PDMS and modifying the surface of the PDMS elastomer with a layer of "hydrogel skin", we managed to achieve an "all-in-one" polymer adhesive tape with tissue-like softness, wet tissue adhesion, and long-term stability.

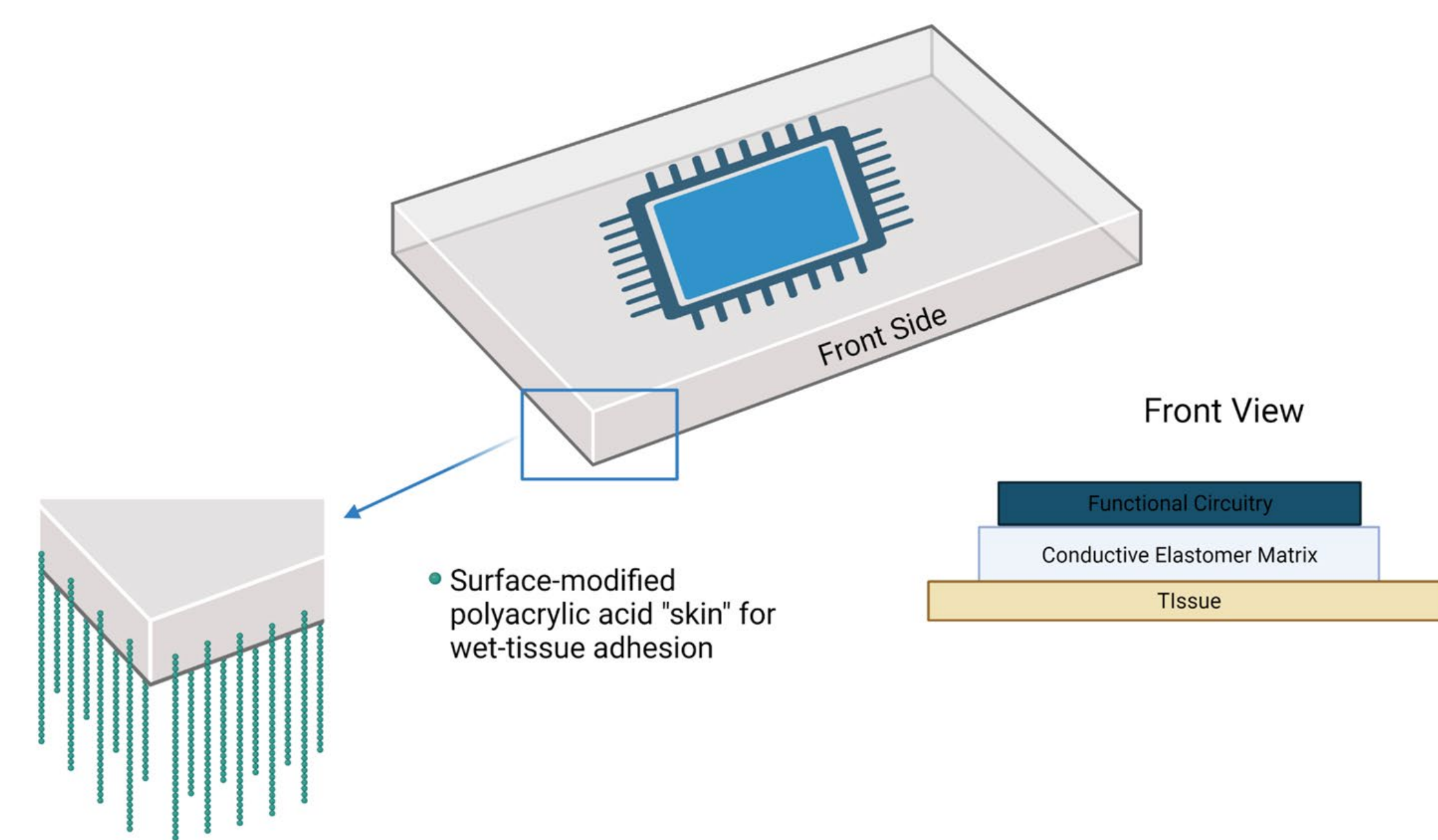


Figure 1. An PDMS elastomer matrix with surface-modified polyacrylic acid "skin" for adhesion. The thin layer of adhesive "skin" contains functional groups that can interact with various wet tissues and provide tough adhesion.. Figure created at biorender.com.

Functional groups for tissue adhesion

- A commonly used polymer in hydrogel applications that provide tissue adhesion is crosslinked polyacrylic acid (PAAc) [1]. By introducing this polymer as a "skin" in the surface-modified system [10], we expect the polymer chains to provide accessible functional groups for interactions with tissue surface through hydrogen bonding [4].
- Additionally, another functional group, PAAc grafted with N-hydroxysuccinimide ester (PAAc-NHS) can lead to greater adhesion through the formation of covalent bonds with primary amines in the tissue [5].

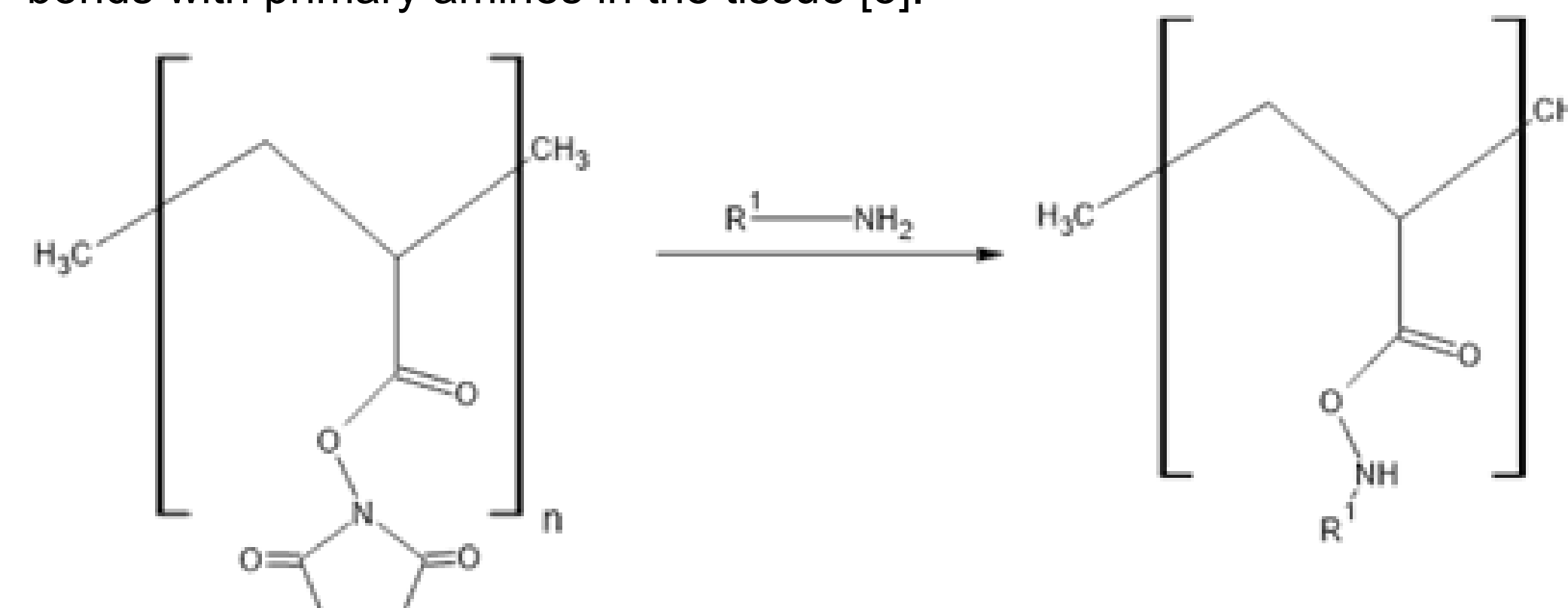


Figure 2. The reaction between PAAc-NHS group with primary amines in tissue, resulting in covalent bonding between tissue and the polymer. Figure created in ChemSketch.

PDMS Hydrosilylation

- The synthesis of the PDMS matrix is conducted through a chemical reaction termed "hydrosilylation".
- Tuning the ratio between the vinyl-terminated polymers and the hydride functional polymers allows for manipulation of the mechanical properties of the PDMS elastomer, such as softness, toughness, stretchability, and elasticity [9].

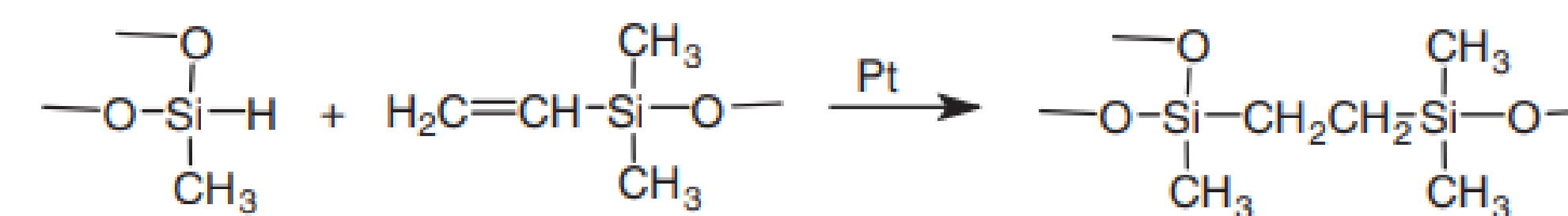


Figure 3. The reaction of the hydrosilylation of PDMS.

Elastic Moduli of Elastomer Matrix

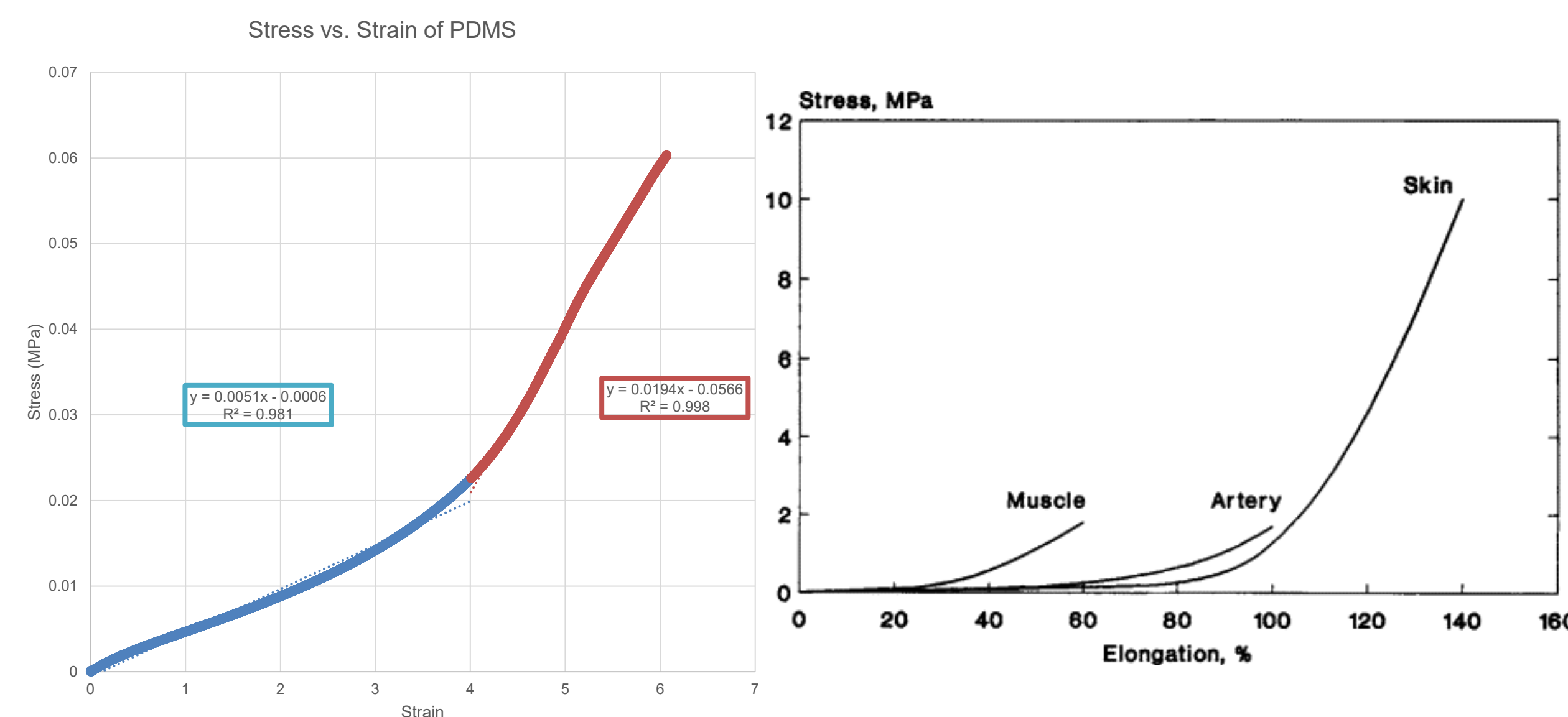


Figure 4. The graph shows the stress versus strain plot for the elastomer matrix that is used. This PDMS substrate matches the low elastic modulus of biological tissue and the moduli stiffening of biological tissues.

Figure 5. The graph shows the stress versus elongation plot for different biological tissues. The notable characteristics are low elastic moduli and elastic stiffening with increasing loads [7].

Stable Adhesion to Tissue

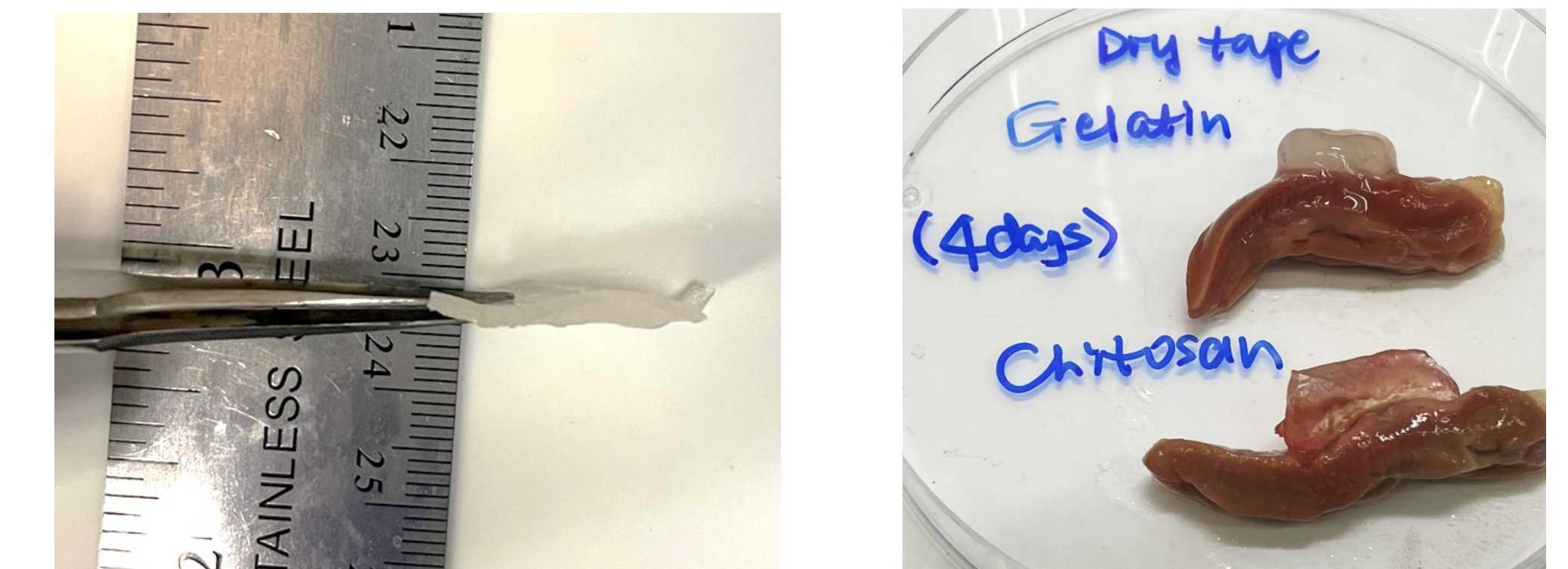


Figure 6. These photos show the swelling of a typical hydrogel system by Yuk et al. [8] after 4 days (the photo on the left before attachment and the photo on the right after attachment with 4 days in PBS). Additionally, the hydrogel does not stay adhered to tissue after a minimal load due to a lack of mechanical properties.

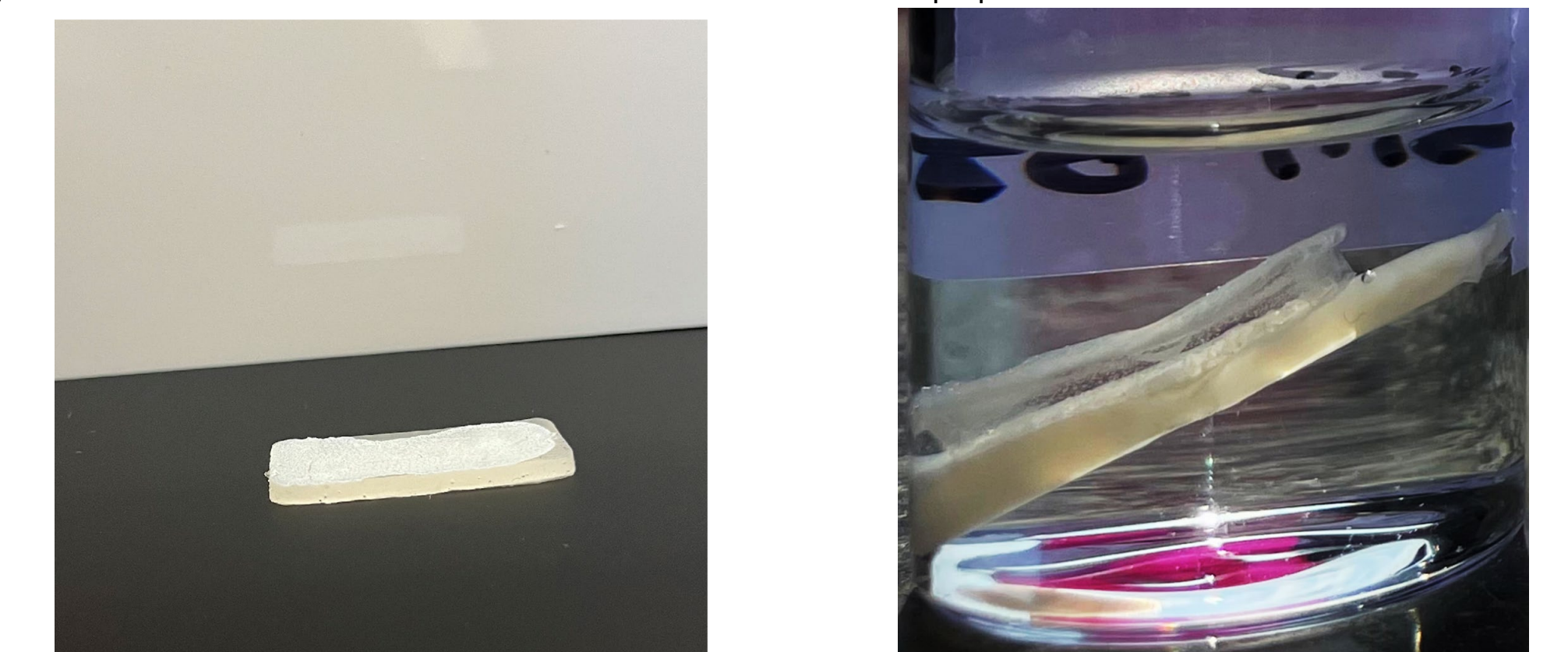


Figure 7. These photos show the continued adherence of the surface-modified PDMS system to biological tissue after 6 weeks. The photo on the left shows the surface-modified PDMS before adherence, and the photo on the right shows the robust adherence to tissue and lack of swelling of the system after 6 weeks in PBS.

Methods

- The elastic moduli of the elastomer matrix were determined by a tensile test with a 10 N load cell.
 - The elastomer was loaded onto the machine with clamps and measured for determination of area and length.
 - The tensile test was performed under a constant elongation of 50 mm/min until the point of failure.
- PDMS matrix is made by mixing together vinyl-terminated polymers with hydride functional polymers at a range of ratio. A platinum catalyst is then added to accelerate the hydrosilylation reaction.
 - After mixing, a degassing procedure occurs to remove bubbles in the elastomer.
 - The mixture is then placed in a 90 °C oven for 10 minutes to cure.

References and Acknowledgements

- [1] S. Nam and D. Mooney, "Polymeric Tissue Adhesives," *Chem. Rev.*, vol. 121, no. 18, pp. 11336–11384, Sep. 2021, doi: [10.1021/acs.chemrev.0c00798](https://doi.org/10.1021/acs.chemrev.0c00798).
- [2] J. Yang, R. Bai, B. Chen, and Z. Suo, "Hydrogel Adhesion: A Supramolecular Synergy of Chemistry, Topology, and Mechanics," *Advanced Functional Materials*, vol. 30, no. 2, p. 1901693, 2020, doi: [10.1002/adfm.201901693](https://doi.org/10.1002/adfm.201901693).
- [3] C. Cai, Z. Chen, Y. Chen, H. Li, Z. Yang, and H. Liu, "Mechanisms and applications of bioinspired underwater/wet adhesives," *Journal of Polymer Science*, vol. 59, no. 23, pp. 2911–2945, 2021, doi: [10.1002/pol.20210521](https://doi.org/10.1002/pol.20210521).
- [4] J. O. Zoppe, N. C. Ataman, P. Moczny, J. Wang, J. Moraes, and H.-A. Klok, "Surface-Initiated Controlled Radical Polymerization: State-of-the-Art, Opportunities, and Challenges in Surface and Interface Engineering with Polymer Brushes," *Chem. Rev.*, vol. 117, no. 3, pp. 1105–1318, Feb. 2017, doi: [10.1021/acs.chemrev.6b00314](https://doi.org/10.1021/acs.chemrev.6b00314).
- [5] N. Eliaz, "Corrosion of Metallic Biomaterials: A Review," *Materials (Basel)*, vol. 12, no. 3, p. 407, Jan. 2019, doi: [10.3390/ma12030407](https://doi.org/10.3390/ma12030407).
- [6] C. Zhang, N. Luo, and D. E. Hirt, "Surface Grafting Polyethylene Glycol (PEG) onto Poly(ethylene-co-acrylic acid) Films," *Langmuir*, vol. 22, no. 16, pp. 6851–6857, Aug. 2006, doi: [10.1021/la053272z](https://doi.org/10.1021/la053272z).
- [7] F. A. Duck, "Chapter 5 - Mechanical Properties of Tissue," in *Physical Properties of Tissues*, F. A. Duck, Ed., London: Academic Press, 1990, pp. 137–165. doi: [10.1016/B978-0-12-222890-1.50009-7](https://doi.org/10.1016/B978-0-12-222890-1.50009-7).
- [8] H. Yuk et al., "Dry double-sided tape for adhesion of wet tissues and devices," *Nature*, vol. 575, no. 7781, Art. no. 7781, Nov. 2019, doi: [10.1038/s41586-019-1710-5](https://doi.org/10.1038/s41586-019-1710-5).
- [9] P. Hu et al., "One reaction to make highly stretchable or extremely soft silicone elastomers from easily available materials," *Nature Communications*, vol. 13, Jan. 2022, doi: [10.1038/s41467-022-28015-2](https://doi.org/10.1038/s41467-022-28015-2).
- [10] Y. Yu et al., "Multifunctional 'Hydrogel Skins' on Diverse Polymers with Arbitrary Shapes," *Advanced Materials*, vol. 31, no. 7, Feb. 2019, doi: [10.1002/adma.201807101](https://doi.org/10.1002/adma.201807101).

Special Acknowledgements to the National Science Foundation SUNFEST program with NSF REU grant no. 1950720.