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Bio-inspired adhesion: local chemical environments impact adhesive stability MATTHEW A. GEBBIE, Materials Department, University of California, Santa Barbara, MICHAEL V. RAPP, University of California, Santa Barbara, JING YU, California Institute of Technology, WEI WEI, J. HERBERT WAITE, JACOB N. ISRAELACHVILI, Materials, Research Laboratory, University of California, Santa Barbara — 3,4-dihydroxyphenylalanine (Dopa) is an amino acid that is naturally synthesized by marine mussels and exhibits the unique ability to strongly bind to surfaces in aqueous environments. However, the Dopa functional group undergoes auto-oxidation to a non-adhesive quinone form in neutral to basic pH conditions, limiting the utilization of Dopa in biomedical applications. In this work, we performed direct surface force measurements with in situ electrochemical control across a Dopa-rich native mussel foot protein (mfp-5), as well as three simplified model peptide sequences. We find that the neighboring peptide residues can significantly impact the redox stability of Dopa functional groups, with lysine residues imparting a substantial degree of Dopa redox stabilization. Surprisingly, the local chemical environments only minimally impact the magnitude of the adhesion forces measured between molecularly-smooth mica and gold surfaces. Our results provide molecular level insight into approaches that can be used to mitigate the detrimental impact of Dopa auto-oxidation, thus suggesting new molecular design strategies for improving the performance of Dopa-based underwater adhesives.

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