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WITHDRAWN (I) Fabrication of porous bioadhesives via liquid-liquid phase separation of multiprotein condensates

Wet adhesion is critical for many engineering, biomedical and dental applications; yet, current man-made glues and cements are ineffective on wet surfaces. Mussels, however, can glue themselves firmly on seashore surfaces using protein-based fibers known as byssal threads, each of which terminates in a small adhesive plaque. The byssal plaque is an important role model for understanding biological adhesion and has stimulated the invention of bio-inspired glues that mimic plaque protein chemistry. Yet, currently, little is understood about the plaque formation and curing process –crucial information for designing novel wet adhesives.

Here, I present recent work elucidating the plaque formation process. Mussels form byssal threads from a secretion of fluid protein precursors in a process lasting just minutes. Using a combination of advanced materials characterization techniques combined with traditional biochemical methods, we discovered that plaque precursor proteins are stored in secretory vesicles as condensed fluid phases (coacervates)[1]. During plaque formation, fluid proteins are secreted into an interconnected network of microchannels, resembling a microfluidic device. Within these channels, proteins undergo liquid-liquid phase separation spontaneously forming the hierarchically porous structure characteristic of the native plaque and important for its mechanics. Moreover, tiny vanadium- and iron-enriched particles are co-secreted with the plaque proteins, curing the glue via formation of protein-metal coordination cross-links. Purification of intact secretory vesicles enabled *in vitro* assembly studies, identifying pH, salt concentration and redox potential as key factors guiding phase-separation and cross-linking. Extracted concepts are directly applicable for generating novel bio-inspired adhesives.

1. Priemel et al. (2021) Science. 374, 206-211.

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