

Marine Inspired Wet Adhesion

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Author approved release edition. Literature analysis and proposed research; no new VARUNÉ experimental results are reported.

Abstract

Marine adhesion offers a useful research strategy when its mechanisms are translated into specific material questions. It does not provide a transferable guarantee that a marine polymer, catechol group, or protein will form a reliable wet joint. This critical narrative report examines how phase separation, chemical state, molecular presentation, network formation, and testing conditions affect the interpretation of wet adhesion. The analysis connects two supplied marine-biomaterials chapters with primary studies of mussel proteins, peptide models, synthetic elastomers, and coacervates, including work published in 2025 and 2026. Its central argument is that successful delivery, intimate surface contact, interfacial binding, and resistance to subsequent loading require related but distinguishable evidence. A change that improves one stage can compromise another. Four equations clarify phase accounting, model-dependent force interpretation, separation work, and a proposed history-by-conditioning comparison. The resulting VARUNÉ Bio research proposition is a bounded material study testing whether changing the order of contact and consolidation improves performance after realistic conditioning. This is an analytical synthesis and proposed research design, not a report of experiments performed by VARUNÉ Bio. Its value lies in making an adhesive claim testable, identifying reasons for contradictory results, and specifying a contribution that an existing materials programme could evaluate.

Research question and contribution

The question is: under what conditions does a material that can reach and spread over a wet surface become a joint that remains useful during its intended service? This deliberately includes both the adhesive and the process used to place it. A formulation can be easy to dispense yet disperse before attachment. Another can resist dispersal but consolidate before achieving sufficient contact. A third can bind locally whilst failing through its own bulk. These are different problems, requiring different remedies.

The contribution proposed here is an interpretation and comparison structure. It links observations to the stage they can actually establish, then asks what additional comparison would discriminate between plausible explanations. This is not a claim to have discovered the distinction between adhesion and cohesion, a new physical law, or an unprecedented design framework. The underlying concepts are established. The author analysis consists in applying them together to the specific decisions facing a prospective VARUNÉ Bio material programme and in deriving a focused, falsifiable study from their connections.

The scope is deliberately material-level. It includes wet mineral and model organic surfaces and draws selectively on tissue-adhesive research for mechanistic comparison. It does not establish skin

efficacy, medical-device protection, clinical suitability, or environmental acceptability for a future formulation. Those claims require evidence matched to the actual product and use.

Evidence selection and access

The starting sources were chapters 14 and 39 of *Marine Biomaterials*, with targeted rereading of the extracted passages on mussel protein roles, coacervation, DOPA chemistry, production, polymer variability, and ion-dependent behaviour. The relevant locators are printed pages 289-306 and 766-780, respectively; these correspond to physical PDF pages 312-329 and 789-803 in the supplied copy. Selected network-defect passages in *Polymer Physics*, chapter 7, supplied a complementary distinction between material present and load-bearing connectivity. [1,2,15]

Targeted web searches on 28 October 2026 used combinations including “mussel adhesion redox mfp-6 2011”, “mussel wet adhesion hydrophobicity catechol synergy lysine”, “viscosity and interfacial properties mussel coacervates”, “coacervate wet adhesion fracture rate salt 2024 2025”, and the exact titles of relevant studies. Search results were followed to publisher, PubMed, PubMed Central, or institutional author records. Primary studies were selected for their ability to distinguish mechanisms, expose an adverse trade-off, or connect delivery conditions to subsequent performance. Numerical results were retained only where the relevant conditions could be checked.

This was a targeted narrative search, not a systematic review. No comprehensive database export, screening denominator, risk-of-bias score, meta-analysis, or literature-completeness claim is made. Studies using incompatible substrates or geometries were not pooled. Search-selected examples can overrepresent well-studied mechanisms, and inaccessible methods constrain assessment.

Full-text material was available for the Kaur elastomer study and selected passages from other author manuscripts. Several sources were checked through abstracts, indexed main-text passages, and figure captions rather than a continuous reading of every result and supplement. In particular, the 2026 Gimenez article was verified at abstract and bibliographic level; a direct full-text attempt was blocked. The 2025 hyaluronic acid-chitosan article was assessed through indexed results and the PubMed record. These limitations are retained in the reference list. No new whole-book or completed-shortlist reading claim follows from this report.

What the marine chapters establish

The mussel chapter treats attachment as an organised material system. Different foot proteins occupy different regions and have different proposed functions: interfacial binding, bulk connections, coating, and redox support. This matters because total protein concentration does not establish which component reaches the substrate or remains chemically available there. The chapter also distinguishes adhesion across a boundary from cohesion within a material. Its examples therefore support a question about arrangement and state, not merely ingredient identity. [1, pp. 290-299]

The polysaccharide chapter makes a complementary point. Chitosan, alginate, carrageenan, and agar are discussed in several applications because their properties depend on composition, processing, surrounding ions, and physical form. Chitosan molecular weight and deacetylation, for example, cannot be collapsed into the single name “chitosan”. The chapter explicitly discusses batch and source variability. For an adhesive programme, polymer naming and nominal concentration would consequently be insufficient release specifications. [2, pp. 772-777]

Both chapters are historical foundations, published in 2013. Broad statements about biocompatibility, cost, commercial readiness, or environmental benefit are not adopted as present-day product evidence. A marine origin also says nothing by itself about sustainable sourcing. A synthetic or recombinant material may reproduce a useful interaction without reproducing the original biological system; whether that is advantageous depends on measured performance and production requirements.

One source-quality issue deserves preservation. The mussel chapter's printed page 297 states an inverse fourth-power dependence of tube flow rate on radius. That statement is not used here. More importantly, the coacervates under discussion can be non-Newtonian, so a simple capillary-flow relation would not automatically describe their delivery. The research design measures dispensing behaviour directly instead of importing an unvalidated equation from the chapter.

Primary evidence and what it changes

Chemical state can matter more than chemical inventory

Yu and colleagues studied the loss and restoration of mfp-3 adhesion using controlled pH changes and mfp-6 addition. Their surface-force results and thiol-modification comparison support a role for mfp-6 in maintaining reducing capacity. Returning the solution to an earlier pH did not simply reproduce the earlier adhesive state. This is evidence for history-dependent chemistry in a specified protein model, not proof that any antioxidant improves a finished adhesive. [3]

Zeng and colleagues provide an instructive contrary result to “more crosslinking reagent is better”. In their symmetric mfp-1 films, low ferric-ion concentrations enabled reversible bridging, whereas 100 µM abolished the bridging observed at 10 µM. Contact time also affected the response. The useful lesson is a conditional interaction, with an optimum that cannot be assumed to transfer to another protein, geometry, or formulation. [4]

Together these examples motivate measuring the state of the relevant groups at the time of contact and loading. A supplier's initial functional-group content is not a complete description of the use-state material. That inference is a research requirement, rather than a demonstrated explanation for any VARUNÉ candidate.

Molecular proximity can help one function whilst weakening another

Maier and colleagues used siderophore-derived models to investigate the joint contribution of catechol and lysine to adhesion on mineral surfaces. Rapp and colleagues then examined how functionality and molecular arrangement affected the interaction; simply increasing the amine-to-catechol ratio did not produce monotonically greater adhesion. These studies support testing arrangement and balance rather than counting functional groups alone. Their mineral-surface results are not universal rankings for polymers, proteins, or tissue. [6,7]

Shin and colleagues examined three model peptides and reported that flanking lysine enhanced surface adhesion whilst disrupting ferric-ion-mediated cohesion. The abstract supports an explicit trade-off between two useful functions. Detailed methods and effect sizes were not independently audited here, so this is treated as a mechanistic lead to verify rather than a quantitative specification. [10]

The author interpretation is that molecular optimisation needs an intended location. A design that is advantageous at an exposed boundary may be disadvantageous throughout the load-bearing volume. One possible response is spatial separation of roles, such as an interfacial layer and a mechanically different bulk. Another is timed transformation. Neither solution should be presumed to be superior without a comparison that includes the extra processing burden.

Water displacement and bonding are distinguishable

Kaur and colleagues designed protected and deprotected catechol elastomers with independent crosslinking chemistry and similar bulk responses. Their spectroscopy and adhesion measurements supported a contribution from catechol binding, whilst hydrophobic groups assisted partial removal of interfacial water. The interface remained patchy under wet conditions. The study also identified a confound in an oxidation treatment: the film was physically disrupted, so loss of adhesion could not be assigned only to oxidation of catechol. [9]

This is especially valuable because it challenges an apparently decisive control. A treatment intended to remove one chemical function can also change roughness, coverage, or cohesion. For the proposed programme, chemical controls therefore need physical characterisation. A lower force after treatment is an observation; its molecular explanation remains a separate inference.

Levine and colleagues found different peptide interactions with methyl- and hydroxyl-terminated model surfaces using surface-force measurements and molecular simulations. The substrate was an active part of the observed behaviour. The result supports including both a mechanistic reference surface and the intended application surface, whilst keeping their conclusions separate. [8]

Delivery and consolidation have different favourable states

Hwang and colleagues examined a recombinant mussel-protein-hyaluronic-acid coacervate. Mixing ratio was associated with differences in phase structure and shear response: the condition of greatest coacervation showed shear thinning, whereas marginal coacervation conditions showed shear thickening. The result connects composition to processing behaviour and cautions against treating “a coacervate” as a single material state. [5]

The 2026 Gimenez study extends this issue to a synthetic poly(acrylic acid) system. Its abstract reports droplet formation, sedimentation and coalescence, followed by a polymer-rich continuous phase containing water droplets; the transition accompanied increased viscoelasticity and tack-measured adhesion. This is timely evidence that phase topology deserves attention. Because full methods were not accessed, no quantitative performance or manufacturing claim is adopted. [13]

A distinct 2025 comparison by Yunus and colleagues found that hyaluronic acid-chitosan systems could differ in peak adhesion and salt resilience. The reported tack measurements used a fixed retraction speed of 100 $\mu\text{m/s}$, and the authors identified rate dependence as an outstanding question. This makes the study particularly useful for preventing a single-condition winner from becoming a general material ranking. [11]

A useful intervention may occur at the interface

Kwant and colleagues investigated bridging polymers in a salt-switchable complex-coacervate system. Polyallylamine pretreatment improved adhesion on their model hydrogels and tissue substrates, whereas the corresponding polyanion did not. The study supports an interfacial

contribution rather than the assumption that the bulk formulation alone determines performance. It also reported a slightly cytotoxic extract result despite a more favourable direct-exposure result; the material cannot be summarised simply as “safe”. [12]

For VARUNÉ Bio, the broader question is whether a targeted interfacial intervention solves the relevant failure more efficiently than reformulating the entire bulk. The cited tissue experiments do not authorise tissue use of a new material. A non-biological material comparison is sufficient to investigate this initial question.

Observation to interpret	Competing explanation	Evidence that would discriminate
Higher separation force	Stronger interface, stiffer bulk, larger contact, or changed failure path	Contact imaging, bulk response, and failure-surface inspection
Faster consolidation	Useful network formation or premature loss of spreading	Time-resolved coverage and joint formation under identical placement conditions
More polymer in the dense phase	Greater recovery or stronger enrichment of a small fraction	Both phase volumes, concentrations, and unaccounted material
Retention after soaking	Durable bonding or a large residual deposit with poor load transfer	Material balance together with service-relevant loading

Quantitative reasoning and its limits

Phase enrichment is not recovery

For a consistently defined polymer species in two accounted liquid phases, ordinary mass conservation gives:

$$f_d = \frac{c_d V_d}{c_d V_d + c_l V_l} = \frac{K \phi}{1 + (K-1)\phi}, \quad K = \frac{c_d}{c_l}, \quad \phi = \frac{V_d}{V_d + V_l}, \quad R_d = \frac{c_d V_d}{m_0}$$

Equation 1 Dense phase fraction and recovery from the starting material

Concentrations c_d and c_l may be measured in kg/m^3 ; volumes V_d and V_l in m^3 ; m_0 is the starting mass in kg. The fractions f_d and R_d , enrichment ratio K , and volume fraction ϕ are dimensionless. The ratio form requires c_l greater than zero. f_d describes distribution within the two measured phases; R_d describes recovery relative to the starting mass. They are equal only when all starting material is accounted for in those phases. Surface deposits, precipitates, degradation products, and sampling losses require separate accounting. This is an algebraic identity applied to the coacervation problem, not a fitted phase-equilibrium model. [1]

As an illustrative calculation, not experimental data, $K = 100$ and $\phi = 1/101$ give $f_d = 0.50$. A hundredfold concentration difference can therefore coexist with only half the accounted polymer in the dense phase. If material has additionally been lost elsewhere, recovery from the starting

preparation is lower still. This distinction directly changes how much usable material a production run would deliver.

Pull off force requires a contact model

The classical Johnson-Kendall-Roberts model gives the following pull-off magnitude:

$$F_{\text{pull}} = \frac{3}{2} \pi R^* W$$

Equation 2 Pull off force in the JKR elastic contact model

F_{pull} is force in N, R^* is the effective radius of curvature of the contacting bodies in m, and W is work of adhesion per area in J/m², equivalent to N/m. The model concerns smooth, elastic adhesive contacts in its quasi-static, short-range-adhesion regime; substantial viscoelasticity, liquid-bridge forces, or hydrodynamic forces require additional treatment. It is not a general conversion from the maximum force of a rough, viscoelastic, finite-thickness, or cohesively failing adhesive layer into an intrinsic molecular adhesion energy. The original model and its application in the elastomer study illustrate why geometry and assumptions accompany the reported number. [14]

Work of separation includes more than interfacial bonds

A measured work per nominal contact area can be defined as:

$$G_{\text{app}} = \frac{1}{A_0} \int_{\delta_i}^{\delta_f} F_{\text{sep}}(\delta) d\delta$$

Equation 3 Apparent separation work over a specified test path

F_{sep} is baseline-corrected tensile force in N, taken positive during separation; displacement delta is in m; A_0 is the stated initial nominal contact area in m². G_{app} is in J/m². The integration bounds, preload, contact time, retraction rate, and area definition must be reported. Elastic storage, viscous dissipation, fibrillation, fracture, and substrate deformation may all contribute. The measurement is useful even when it is not a thermodynamic work of adhesion, provided its operational definition is preserved. This is a standard work definition applied to the proposed test, not a source-specific constitutive model.

A history effect should be tested as an interaction

For a proposed comparison of consolidation after contact with consolidation before contact:

$$\Delta_H = (\bar{Y}_{\text{after, long}} - \bar{Y}_{\text{before, long}}) - (\bar{Y}_{\text{after, short}} - \bar{Y}_{\text{before, short}})$$

Equation 4 Proposed history by conditioning contrast

Y is one prespecified response, such as G_{app} in J/m²; bars denote appropriately estimated means across independent preparations. “Short” and “long” denote prespecified conditioning intervals. Δ_H has the same units as Y . This is a standard difference-of-differences contrast applied here, not a validated adhesive index. It tests whether the history difference changes with conditioning. It does not, alone, identify the mechanism, and it requires uncertainty estimates that account for preparation batches and repeated measurements.

Varun analysis of the connected findings

The material specification should include a path

The combined evidence suggests that a recipe is only part of the specification. An adhesive must enter a useful state at the right time and place. Two preparations with identical nominal ingredients could differ because one has already oxidised, phase-separated, lost solvent, or formed a network before it reaches the target. Conversely, two chemically different preparations might produce similar functional states under a tightly controlled process. The research objective is to determine which distinctions affect the intended decision, rather than insisting that every measurable difference is consequential.

The proposed interpretation is to describe the material at three decision points: immediately before placement, after contact formation, and after service conditioning. This does not require exhaustive characterisation at every moment. It requires enough information to distinguish the leading explanations for failure. For example, an improvement after delayed consolidation could arise from greater area of contact, a different network, or altered surface chemistry. Measuring only the final peak force would leave all three possibilities open.

A stronger bulk is neither necessary nor sufficient for a better joint

The polymer-network reading distinguishes elastically effective connectivity from total polymer or total chemical attachments. This makes it reasonable to question whether adding crosslinker actually produces useful load-bearing structure. It can also change mobility and the ability to conform to the substrate. These statements motivate tests; they do not provide a quantitative network model for a heterogeneous coacervate. [15, chapter 7]

A useful research outcome could therefore be a controlled compromise. A material may need enough mobility to establish contact and dissipate energy, but enough persistence to resist creep during service. The application determines which time scales matter. A high instantaneous separation force may be irrelevant if a modest sustained load causes displacement over the required use period. Conversely, an adhesive intended for temporary placement may be unsuitable if its strongest bond makes removal damaging or impractical.

This leads to a decision rule: optimise the required service outcome, then use mechanistic measurements to explain and stabilise it. Do not optimise a convenient modulus or ingredient percentage and assume the outcome follows.

Failure location contains information that averages discard

Suppose two preparations have similar apparent separation work. One separates cleanly at the interface; the other leaves material on both substrates. Those are not equivalent outcomes. The second may have a stronger interface but inadequate bulk integrity, or may contaminate a surface that must remain clean. A single mean force erases the distinction. The proposed report of every mechanical comparison should therefore include a classified failure mode and uncertainty in that classification.

Mixed failure should remain mixed when the evidence is ambiguous. Staining alone can mislead if a dye partitions differently from the polymer; imaging must be checked for representation. Where feasible, independent compositional measurements of residue provide corroboration. This connects

the adhesion problem to the broader series' concern with attributing function: apparent presence, chemical identity, and useful load transfer are different observations.

Robustness can be more valuable than the highest favourable result

A candidate that performs exceptionally only on pristine glass after a precise waiting time may be scientifically interesting and commercially narrow. Another with lower peak adhesion but stable behaviour across the intended contamination, salinity, and placement window could be more useful. This is an application judgement, not a universal preference for low variability over high performance. The threshold must come from the actual task.

The counterargument is that broad testing can exhaust a small programme before a mechanism is understood. The response is sequential selection. Begin with a clear mechanistic comparison, then challenge the promising result with the smallest set of conditions likely to reverse the decision. Do not search every substrate or add marine origin as a second objective. A useful stop decision can save more resources than another favourable demonstration.

Proposed VARUNÉ Bio material study

A bounded question

The proposed study asks whether establishing wet contact before consolidation improves a selected adhesive's retained performance after conditioning, compared with consolidation before contact. It would use one candidate system, one intended non-biological substrate, and a mechanistic reference surface. A specific material, application owner, scientific lead, budget, and rights position have not been established by this report. The study becomes an active programme only when those items and a meaningful acceptance criterion are agreed.

The first stage is method feasibility. It must establish that the adhesive can be dispensed reproducibly, that the contact area can be measured or bounded, and that the test distinguishes a weak reference from a useful commercial or laboratory benchmark. A benchmark should be selected for the same placement conditions and use case. A dry-bonded incumbent is not automatically a fair comparison for a formulation applied underwater.

Comparisons and controls

The principal comparison varies the order of contact and consolidation whilst accounting for composition, solids, deposited mass, substrate preparation, temperature, and final conditioning. If the intervention inevitably changes chemistry or water content, those changes should be measured and described; the experiment cannot then be interpreted as isolating chronology alone.

A limited first study could use at least three independently prepared batches to estimate reproducibility, with technical repeats nested within batches. That is a proposed feasibility minimum, not a power calculation or sufficient replication for a definitive performance claim. Final sample size would follow the decision-relevant effect and variance. Randomised test order and coded specimens reduce avoidable bias. Testing personnel should record whether physical appearance defeats blinding.

Comparison	Main purpose	Result that would change the interpretation
Contact before versus after consolidation	Test the practical effect of preparation order	Difference disappears after deposited mass and contact area are accounted for
Short versus long conditioning	Test persistence rather than initial tack alone	Initial improvement reverses or disappears during conditioning
Reference versus intended substrate	Test whether mechanism transfers	Advantage is confined to the clean reference surface
Candidate versus relevant benchmark	Test usefulness	Candidate provides no valued benefit after delivery burden is included

Measurements should include dispensing consistency, retained deposited mass, contact coverage, a suitable bulk mechanical measurement, the selected joint response, and failure mode. The priority is complementarity, not instrument count. If a measurement is used to infer oxidation, phase state, or coating presence, its specificity for the candidate must be established. The most elaborate measurement is not automatically the most informative.

Analysis and stopping

The primary response and conditioning interval should be chosen before comparing outcomes. Analysis should estimate history, conditioning, and their interaction, with batch represented appropriately. Confidence intervals and the distribution of individual batch outcomes should accompany the mean. Failed dispensing, excluded specimens, and catastrophic failures remain part of the record; they are not inconvenient outliers to remove retrospectively.

A continuation result would be a reproducible improvement that survives the intended substrate and conditioning, exceeds a pre-agreed practical threshold, and has an interpretable cost in processing and residue. A mechanistic explanation is stronger if independent chemical or spatial observations change in the predicted direction. A positive mechanical result without that support can still justify an application-specific finding, but not the claimed mechanism.

Stop or redirect if the advantage requires unrealistic surface preparation, disappears during useful service, depends on uncontrolled batch state, or creates unacceptable residue. Redirect towards process control if the material is useful only within a reproducible placement window. Redirect towards interfacial preparation if the bulk is adequate but contact remains limiting. This preserves useful negative findings and prevents a narrow failure from being misdescribed as proof that the whole material class is ineffective.

Contributions to existing programmes

The most credible contribution is a comparison package that resolves a programme's uncertain interface, preparation, or conditioning step. Published research groups associated with the Groningen coacervate studies, the Bordeaux simple-coacervation work, and established mussel-

protein research are scientifically relevant counterparties. Their publications establish expertise, not willingness to collaborate, an unmet requirement, available funding, or accessible rights. [3,11-13]

For an environmental sampling mission, an adhesive component could be useful only if attaching a membrane, coating, or sensor solves an identified integration problem. The proposed contribution would compare retention, leaching, and measurement interference under the mission's conditions. It should not begin with a claim that a mission needs a new adhesive. Sample-contamination risk could eliminate an otherwise strong material.

For a materials or formulation programme, VARUNÉ Bio could potentially contribute a reproducible placement method, batch-to-function specification, or independent comparison on the customer's substrate. A paid feasibility study is a possible transaction structure: a defined result, acceptance conditions, ownership of methods and data, and an agreed follow-on decision. No sponsor commitment or live funding call is asserted here. The potentially ownable contribution would need to be defined before confidential development, with background technology and jointly generated improvements treated explicitly.

Potential contribution	Useful output	Gate before committing resources
Process comparison for a coacervate programme	Evidence identifying an effective placement and conditioning window	Team confirms the unresolved question and supplies a relevant comparator
Wet component attachment for a sampling system	Retention and contamination evidence in the actual fluid environment	Integrator confirms a component problem and acceptable leachate limits
Batch specification for a biological adhesive	Relationship between material state and reproducible performance	Source rights, qualified measurement, and a buyer-relevant endpoint are available

Limitations and publication boundary

This report does not rank the cited adhesives. Their geometries, substrates, loads, chemical states, and endpoints differ too much for a defensible single performance table. The accessible literature also favours successful formulations and may underreport failed preparations or manufacturing variation. Abstract-level and indexed-text checks cannot replace inspection of complete methods, supplementary information, and raw data for a decisive development choice.

The proposed comparison structure has not been validated as a predictor of commercial success. Its purpose is to improve experimental interpretation and selection. It would fail in practice if it adds measurements without changing decisions, if the nominated stages cannot be measured sufficiently independently, or if an application is already well served by a simpler reference material.

The research reports in this series share concepts concerning identity, state, and observation. This volume's distinct contribution is their application to adhesive delivery, consolidation, and joint failure. It should be cross-referenced with the protein-interface report rather than represented as an independent discovery of the same general principle.

Conclusion

The useful lesson from marine adhesion is to ask what the material must do at each stage of making and using a wet joint. Delivery, contact, binding, and resistance to loading are related, but evidence for one does not establish the others. Published results show why simple optimisation rules can fail: more metal, more catechol-associated functionality, faster consolidation, or a larger favourable force need not improve the intended outcome. VARUNÉ Bio's strongest next contribution would be a bounded comparison that identifies whether preparation order and conditioning explain a practical failure, and whether controlling those variables produces a reproducible benefit. That would turn biological inspiration into evidence a materials team can use.

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Authorship and research provenance

Varun Sharma is the sole author and approved this edition for release on 28 October 2026. No new experimental data are reported; published findings remain attributable to the cited investigators.

Competing interests. Varun Sharma founded VARUNÉ Bio, which may benefit commercially from applications discussed here. Funding and contributor declarations remain subject to author confirmation.

This report shares concepts and sources with the five companion reports dated 28 October 2026; it presents a distinct literature analysis.