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Recommended Citation

Lee, C., Shi, H., Jung, J., Zheng, B., Wang, K., Tutika, R., Long, R., Lee, B., Gu, G., & Bartlett, M. (2023). Bioinspired materials for underwater adhesion with pathways to switchability. *Cell Reports Physical Science, 4*(10). http://doi.org/10.1016/j.xcrp.2023.101597
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Review

Bioinspired materials for underwater adhesion with pathways to switchability

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SUMMARY

Strong adherence to underwater or wet surfaces for applications like tissue adhesion and underwater robotics is a significant challenge. This is especially apparent when switchable adhesion is required that demands rapid attachment, high adhesive capacity, and easy release. Nature displays a spectrum of permanent to reversible attachment from organisms ranging from the mussel to the octopus, providing inspiration for underwater adhesion design that has yet to be fully leveraged in synthetic systems. Here, we review the challenges and opportunities for creating underwater adhesives with a pathway to switchability. We discuss key material, geometric, modeling, and design tools necessary to achieve underwater adhesion similar to the adhesion control demonstrated in nature. Through these interdisciplinary efforts, we envision that bioinspired adhesives can rise to or even surpass the extraordinary capabilities found in biological systems.

INTRODUCTION

Strong attachment to underwater or wet surfaces is a necessity for numerous organisms as well as a requirement in diverse technological applications.[1–3] However, strong adherence to underwater or wet surfaces is a significant challenge, especially when rapid attachment with high capacity and easy release is required. As such, most commercially available adhesives exhibit adequate adhesive ability only in dry environments.[4] However, nature is rich with examples of underwater adhesion, where organisms like the octopus and mussel excel at attaching to diverse surfaces. These two organisms represent two distinct regimes of underwater attachment (Figure 1). The octopus rapidly adheres to and releases from various interfaces through structurally tunable mm-cm scale attachment features (i.e., suckers) on their limbs, displaying switchable adhesion. The mussel on the other hand, utilizes specific surface chemistry (i.e., 3,4-dihydroxyphenylalanine [DOPA]) and creates a protein-based attachment plaque, displaying permanent adhesion.[5] Several other organisms also display wet or underwater attachment, providing strong inspiration for the design of synthetic adhesives that aim to achieve permanent or switchable adhesion in submerged environments (Figure 1).[6,7]

One of the grand challenges in adhesion science is the ability to create adhesives that strongly adhere but can still be rapidly released. This class of materials is typically called switchable adhesives, and display an ability to rapidly and reversibly switch between a high and low adhesive state.[8] Switchable adhesives are typically achieved through a variety of triggers including thermal activation,[9,10] fluidic/solvent swelling,[11,12] electromagnetic actuation,[13–15] and mechanically based triggers.[16–19] However, switchable

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https://doi.org/10.1016/j.xcrp.2023.101597

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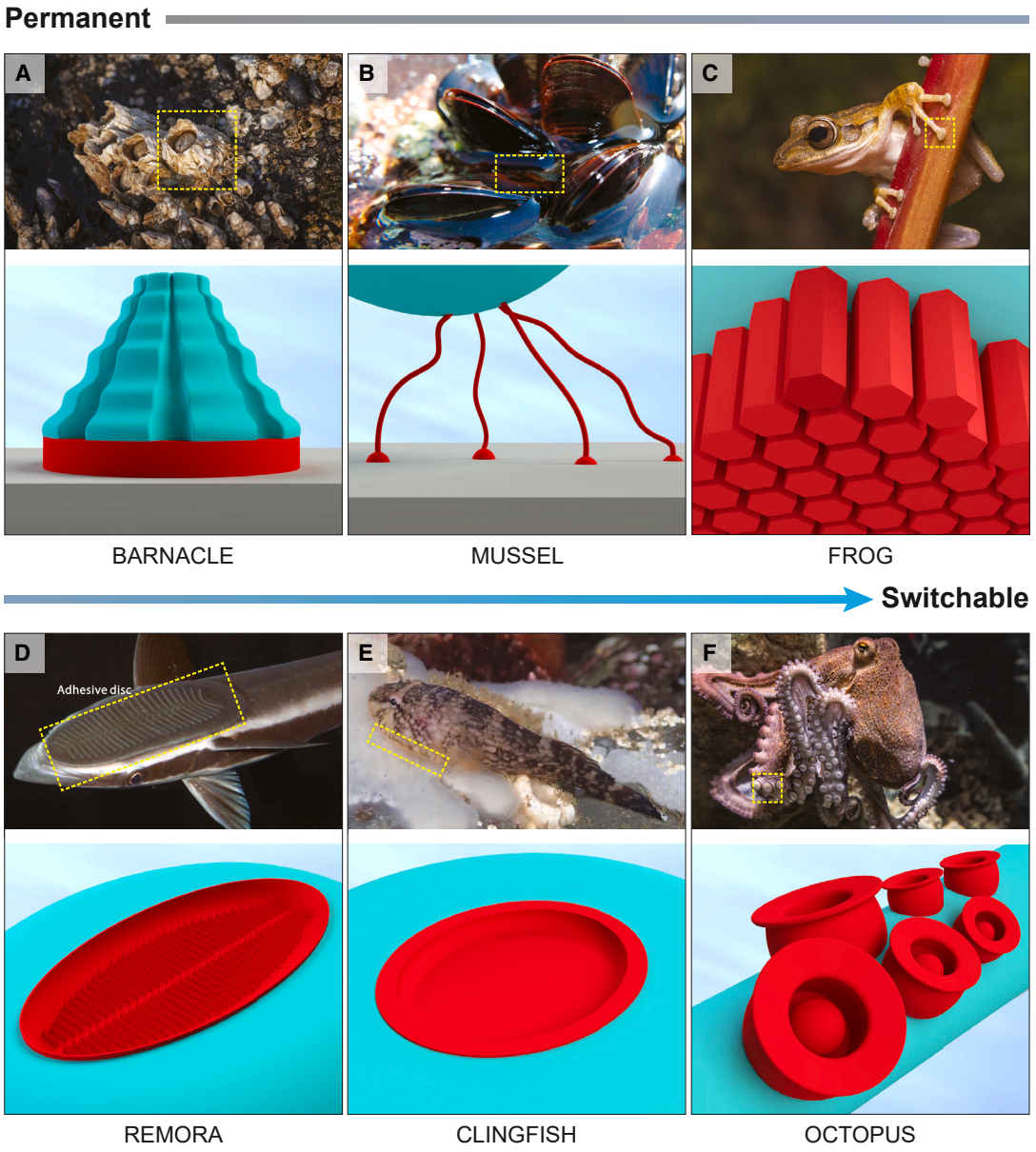

Figure 1. Wet adhesion in nature

(A) Permanent adhesion in barnacles through a protein cement on a base plate.

(B) Mussels create strong adhesion through adhesive proteins on an attachment plaque.

(C) Tree frogs show passive, reversible adhesion through microstructured toe pads.

(D) Remoras create switchable adhesion through an attachment disc with spinules. Adapted with permission from Wang et al.,[6] Copyright 2017, AAAS.

(E) Switchable adhesion in clingfish through an attachment disc.

(F) Cephalopods show switchable adhesion through controllable suckers.

adhesives reported to-date have demonstrated adhesion predominately to dry substrates.[8] Further, the performance of most synthetic adhesives is significantly compromised in the presence of moisture via two key mechanisms.[20,21] First, interfacial water reduces the available surface area between the substrate and adhesive, decreasing the intermolecular force. Second, water can diffuse into the adhesive material, interfering with physical crosslinks or acting as a chemically inert plasticizer, thereby reducing the overall cohesive strength. Therefore, there are significant opportunities to create strong

adhesives that can be rapidly released, but the way forward is challenging not only technically, but also because a wide array of possible approaches could be implemented.

Here, we address the challenges and opportunities for creating underwater adhesives with an aim to provide a pathway to switchability. We structure the discussion through some of the key aspects for underwater adhesion creation: material, geometric, modeling, and design tools. Although these topics are broad, to achieve underwater adhesion similar to the adhesion control demonstrated in nature, an interdisciplinary approach can be synergistic and can accelerate materials discovery. We conclude with outlooks for how we envision some of the future opportunities and prospects for switchable underwater adhesives.

Wet adhesion in nature

Nature has numerous examples of organisms that create strong bonds in moist or submerged environments.[22] The performance and specific properties of each adhesive system are dependent on the habitat and functional needs of the organism. For organisms that do not move or rarely move in their lifetime, a strong and permanent adhesive system is preferred. For example, mussels and barnacles secrete specialized adhesive proteins to attach to a wide range of moist surfaces.[23–30]

Temporary and reversible attachment is important for organisms that need to move. Tree frogs have a passive adhesive system in which attachment and release are essential for their locomotion in a range of dry, to moist, to nearly submerged environments. In these systems, a wetting liquid (mucus) is secreted into the gap between the organism's "adhesive structure" and the target substrate to create strong attachment and facilitate a degree of reversible controlled release.[31–35] Switchable adhesives are also used by organisms that include cephalopods such as the octopus and species of fish such as the clingfish and snailfish.[36–39] In these active, suction-based adhesion systems, organisms will increase the interior chamber volume of a sucker/attachment feature, thus creating a pressure differential between the chamber and the surrounding medium, which creates a force used for attaching.[38,40] The sucker is then actuated again for release. These suckers may display additional features, such as the lamellae on the remora disc or the fibrillar structures on the rim of the clingfish disc. This mechanism functions in dry or submerged environments, allowing for attachment and release enabling locomotion and environmental sensing in diverse underwater habitats.

Adhesive materials and geometry

Efforts to develop bioinspired or biomimetic wet adhesives are largely focused on two categories: (1) controlling the chemical composition or functional aspects of the interface, or (2) controlling geometry and structure of the attachment feature for passive or active adhesion control. Barnacle and mussel-inspired adhesives and pastes are based on the first category in which typically permanent adhesion is created from multi-protein complexes (Figures 2A and 2B). For instance, barnacle-inspired adhesives are created by studying proteins responsible for interfacial adhesion and cohesion in the natural barnacle cement.[30,41,42] However, there are some examples of completely engineered polymer-based systems instead of proteins such as tissue adhesives for rapid hemostatic sealing.[43] There also exist hybrid adhesives that are inspired from both the mussels and barnacle adhesion mechanisms.[44–46] Switchability is typically not present in such systems and only a few cases of reattachable adhesives are demonstrated; for example, tough hydrogels and adhesive hydrogel sensors for underwater electronic applications.[47,48] As such, the

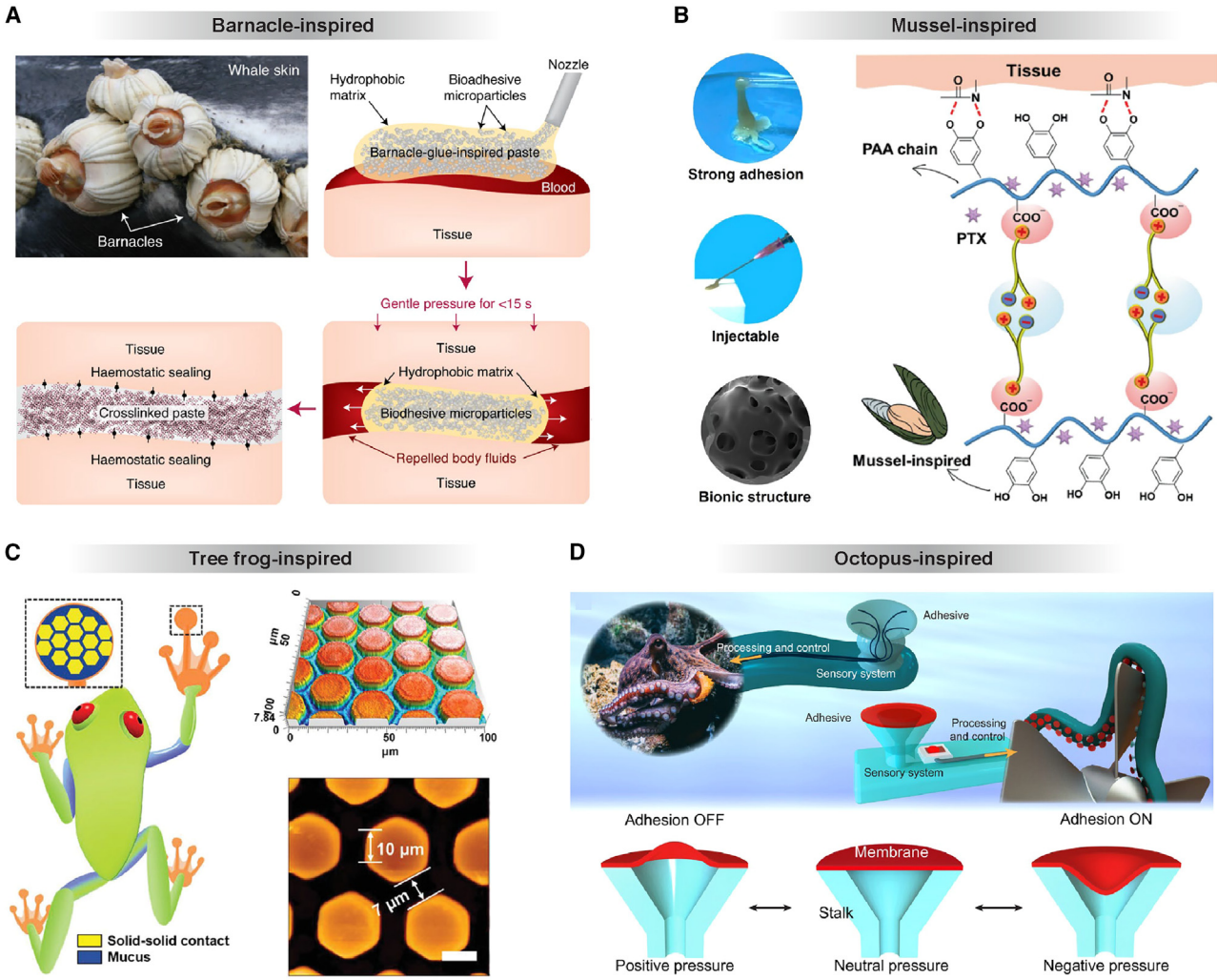

Figure 2. Adhesive materials and geometry

(A) Hemostatic sealing paste with barnacle glue. Adapted with permission from Yuk et al.,[43] Copyright 2021, Springer Nature.

(B) Biocompatible mussel-inspired adhesives for injured arteries. Adapted with permission from Xu et al.,[50] Copyright 2021, John Wiley & Sons.

(C) Hydrogel adhesive inspired from the toe pad of tree frogs (scale bar, 10 μm). Adapted with permission from Meng et al.,[51] Copyright 2021, John Wiley & Sons.

(D) Octopus-inspired rapid, switchable underwater adhesives. Adapted with creative commons attribution license from Frey et al.[52]

applications are primarily focused on bioadhesives for wound healing, bone adhesives, and hemostatic adhesives.[49]

In the second category are bioinspired adhesives that are based on structural hierarchy or geometrical features that include adhesives inspired by clingfish and frogs (Figure 2B). For example, tree frog toe-inspired adhesives are reversible adhesives that are typically fabricated with elastomeric materials. Synthetic frog-inspired adhesives often replicate the hierarchical hexagonal microstructures found on the frog's toe pad (Figure 2C).[51] Repeating hexagonal or polygonal structures on the toe pads create microchannels that expel the water layer from the contact surface and enable contact on wet surfaces.[32,53] Furthermore, the microchannels induce capillary force by forming a capillary bridge that creates attractive interaction with surface in wet environment.[31,33,54] This mechanism can be adopted to create surgical grippers, wearable devices, and adhesive patches.[55,56] However,

these systems do not typically use active-manipulation, which limits the switching time for delamination.

Octopus-inspired adhesives often mimic the sucker geometry to adhere to various surfaces in dry, moist, underwater and under oil conditions (Figure 2D).[52,57–59] This mechanism has inspired adhesives such as tentacle-like soft robotic grippers,[60] microstructured surfaces,[58,61] and active systems controlled by dielectric elastomer actuators and pneumatic pumps.[52,62–66] However, the wet adhesion strength is often limiting in octopus-inspired adhesives, which are typically below 100 kPa,[58,61] with active switchable adhesives often below 20 kPa.[64–66] However, some recent switchable octopus-inspired adhesives have reached over 60 kPa with switching times <50 ms.[52] Key fundamental challenges also remain around octopus-inspired adhesives. For example, experiments on cephalopods have shown that as sucker size decreases, the attachment strength increases,[40] which cannot be explained by suction force alone,[67] and requires further investigation.

Although numerous artificial adhesives are developed with inspiration from various organisms in nature, the challenges to create adhesives for underwater or wet substrates have also evolved with expanding application space and performance requirements. In the case of chemical composition of adhesives, limitations pertain to degradation of adhesion due to environment interaction and delay in forming contact with the substrate after application. For adhesives based on mimicking structure/geometry, the two steps of design and fabrication are the most complex aspects that further involve multiple sub-steps. Furthermore, due to the templating/molding approach typically used in creating these geometries, it is difficult to mass produce and often needs several steps of manual involvement. To overcome the challenges and create switchable adhesives that are durable, rapidly applied, and multi-responsive to various stimuli, interdisciplinary efforts are needed. Such multidisciplinary efforts also call for creating and/or maintaining common, comparable metrics for switchable adhesives for easy communication and dissemination of research results. For instance, a key performance metric for creating switchable adhesives is the adhesive switching ratio, $SR \equiv F_{high}/F_{low}$, where F_{high} is the adhesive force in the "on" state and F_{low} is the adhesive force in the "off" state. Furthermore, the time required to transition from a high to low adhesive state, i.e., switching time, *ST*, should be a careful consideration for rapid on-demand release. Expertise in areas of biology, chemistry, and materials science should be combined with emerging technologies such as machine learning and artificial intelligence in a convergent approach to create unique adhesive chemistries, geometries, or tailored combinations of both for next-generation switchable adhesives with high performance and predictable properties.

Chemistry of adhesive materials

In addition to switchable structural or geometric designs, adhesives incorporated with dynamic chemistry have demonstrated switchable underwater adhesion. These switchable adhesives are either designed with tunable interfacial binding molecules or dynamic crosslinkers within the bulk of the adhesive.[68,69] To control interfacial adhesion, the binding strength of the interfacial molecule and the surface substrate are either increased or decreased. Conversely, breakage and reformation of dynamic crosslinkers is utilized to control the bulk mechanical property and thus the adhesive strength of the adhesive.

In nature, underwater adhesion control through chemistry is exemplified by mussels, which attach to various underwater surfaces by secreting unique mussel foot

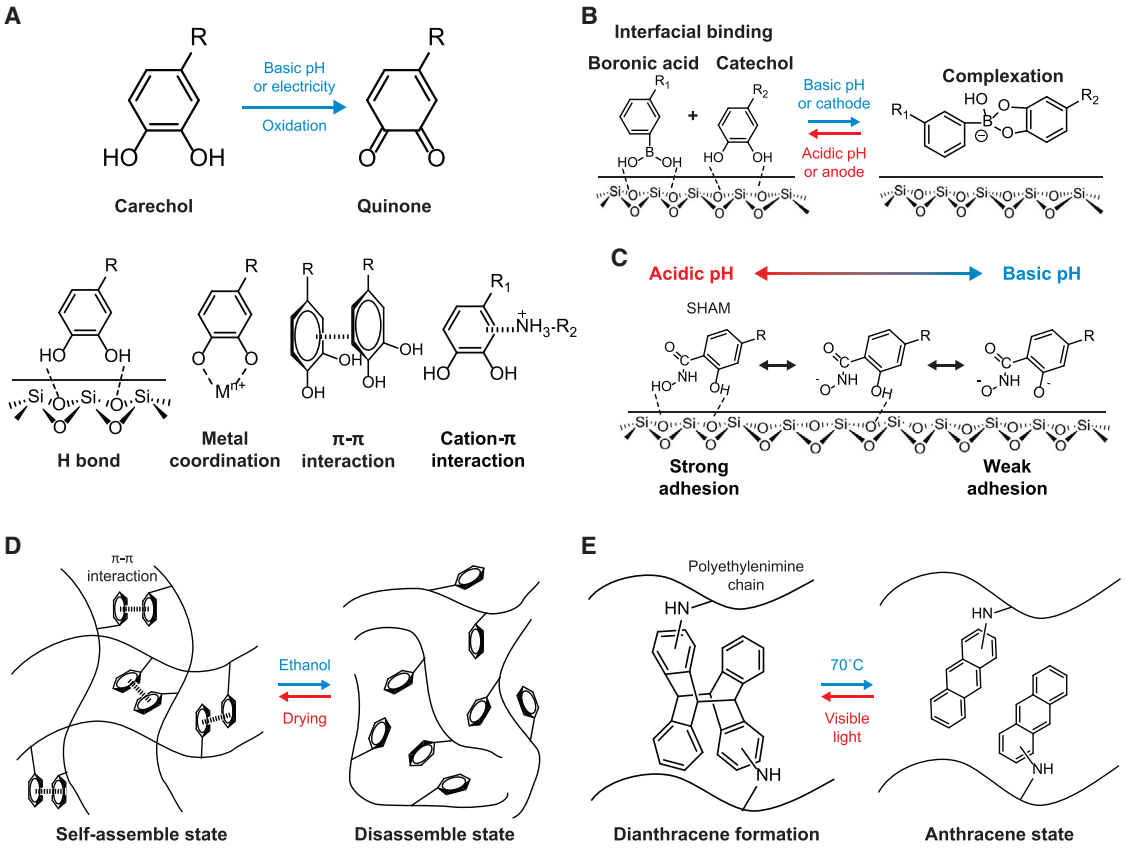

Figure 3. Chemistry of adhesive materials

(A) Chemistry of catechol.

(B) Reversible catechol-boronate complexation and corresponding reversible adhesion.[78]

(C) Switchable adhesion of SHAM.[79]

(D) Reversible transformation between the assembly state (strong adhesion) and the disassembly state (weak adhesion).[80]

(E) Anth-PEI as a reversible underwater glue between dianthracene formation (strong adhesion) and anthracene state (weak adhesion).[81]

proteins that contain a significantly high concentration of the catecholic amino acid, DOPA. Extensive studies have focused on the development of catechol-based adhesives for designing underwater adhesives since catechol can serve as the basis for remarkable adhesion abilities.[25,69,70] Catechol comprises a benzene ring carrying two hydroxy substituents ortho to each other, enabling a range of reversible interfacial interactions, including hydrogen bonding (H-bond), metal coordination, π-π stacking, and cation-π interactions (Figure 3A).[71] These interfacial interactions enable scientists to design synthetic adhesives that can bind to a wide range of surfaces.[72]

Although the mussel typically adheres for long periods of time, DOPA can also be used as a molecule for switchable adhesives. The adhesion strength of catechol is affected by its oxidation state, which can be controlled by changing pH.[73] Upon oxidation, catechol is transformed into the poorly adhesive quinone. The maximum adhesive force between catechol and borosilicate glass has been reported to decrease by 73% when oxidized to quinone in basic environments. Similarly, catechol can be oxidized to the quinone form through electrochemical oxidation.[74] By applying an electric potential to catechol-based hydrogel, the pH near the cathode increases, resulting in the accumulation of negatively charged OH^- ions during water electrolysis. The adhesion strength decreased by 96% when deactivated by applying

a 9 V voltage. Irreversible oxidative crosslinking also occurs and cannot be converted back to catechol, the more adhesive form. To preserve the reversible adhesion properties of catechol, a temporary protecting group in the form of phenylboronic acid can be incorporated to prevent irreversible oxidation.[75] Elevated pH results in the formation of a catechol-boronate complex, which can be reversibly controlled by pH (Figure 3B). This complex formation leads to a decrease in adhesive properties. When the pH is reduced, this complex breaks, allowing both the catechol and the phenylboronic acid moieties to become available for interfacial binding. The reversible nature of the complex enables the adhesive to transition between its adhesive and non-adhesive states in response to changes in pH as well as applied electricity.[76] This reversible chemistry has been used in aiding the locomotion of a wall-climbing robot.[77] In addition to catechol-based switchable adhesive, salicylhydroxamic acid (SHAM) is a newly reported adhesive molecule that consists of a benzene ring with pendant hydroxamic acid and hydroxyl group.[78] SHAM is able to form H-bonds, π-π interaction, and cation-π interaction like catechol, given their structural similarity. SHAM also exhibited equivalent adhesive property to a wide range of surfaces such as glass, metal, and amine-functionalized surface when compared with those of catechol. Remarkably, SHAM exhibits pH-responsive behavior through deprotonation and protonation (pKa1 = 7.4, pKa2 = 9.7) without undergoing oxidation. Unlike catechol, the reversibility of SHAM can be achieved without a temporary protecting group. This reversible adhesive molecule assumes deprotonated states, thereby reducing its interfacial bonding capabilities by 94% (Figure 3C).

To regulate bulk mechanical property of an adhesive, Wang et al.[79] successfully manipulated the π-π stacking interaction without deforming the aromatic structures. By utilizing the robust cohesive energy from water-insensitive siloxane assemblies, they achieved durable and strong adhesion on various surfaces, even under challenging conditions such as extreme pH (1–14) and temperatures ranging from −10°C to 90°C. This adhesive system exhibited the ability to swiftly lose adhesion upon infiltration by ethanol and regain adhesion after ethanol volatilization, accomplishing adhesion control in less than 1 min (Figure 3D). Furthermore, photo-responsive adhesion using anthracenyl-functionalized polyethylenimine (anth-PEI) emerged as another example that utilized switchable crosslinks.[80] By absorbing interfacial water molecules and spreading over diverse substrates, this hygroscopic glue achieved effective underwater adhesion. Notably, visible-light irradiation induced the formation of crosslinked polymer networks via photo-induced anthracene [4 + 4] dimerization reaction, dramatically augmenting the underwater adhesion strength (Figure 3E). Conversely, elevating the temperature caused the cycloreversion of dianthracene, resulting in reduced adhesion due to crosslink dissociation.

Switchable underwater adhesives have greatly developed for broad applications. Nonetheless, several challenges remain to be addressed. One challenge is achieving tunable adhesion strength in various applications, such as climbing robots, industrial assembly, and manipulation systems.[72,81] Apart from the on and off state of adhesion, the capability to dynamically and precisely control adhesion for object pickup and weak adhesion for object placement is still relatively rare. Another challenge is achieving fast switching speed, high switching ratio, and large adhesion strength simultaneously.[68] For instance, topologically enhanced cation–π interactions have greatly enhanced the multifunctional adhesive performance on various substrates, including polytetrafluoroethylene; however, with a longer switching time.[82] Therefore, integrating multiple stimuli, including temperature, light, and electricity, to modulate chemistry is necessary to simultaneously achieve these goals.

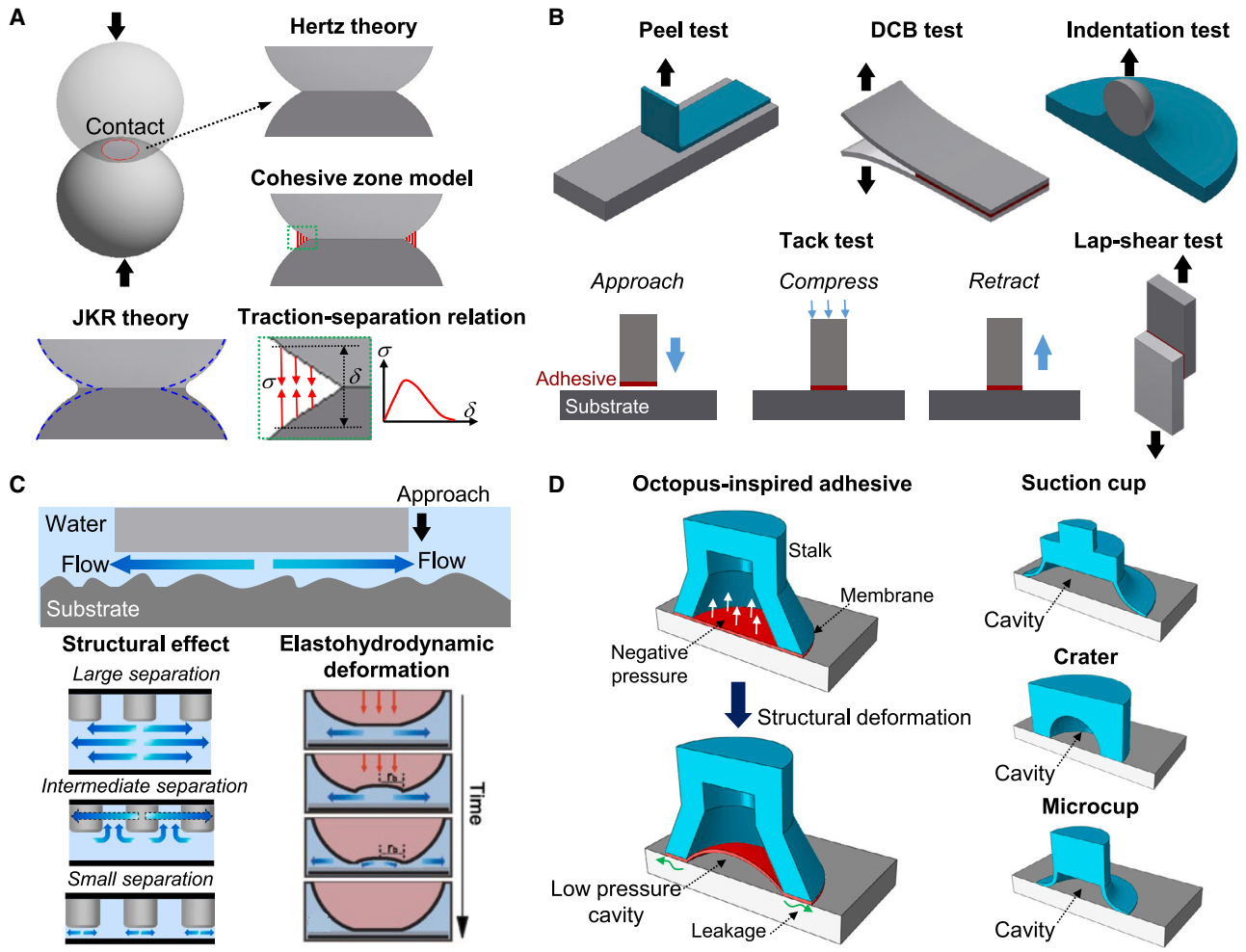

Figure 4. Modeling and testing underwater adhesion

(A) Mechanics models for contact and adhesion between two elastic spheres: Hertz theory for adhesion-less contact, JKR theory where adhesion increases the contact area (the dashed line illustrates the contact profile of Hertz theory for comparison), and cohesive zone model where adhesion is described by the relation between the adhesive traction σ and the interface separation δ.

(B) Testing configurations for adhesion: peel test, Double-Cantilever Beam (DCB) test, indentation test, tack test, and lap-shear test.

(C) Top: schematic illustrating the flow of interface dehydration between a smooth surface and a rough substrate. Bottom left: surface structures can alter the flow pattern of interface dehydration. Adapted with permission from Gupta and Fréchette,[35] Copyright 2012, American Chemical Society. Bottom right: Elastohydrodynamic deformation in a compliant probe. Adapted with permission from Wang et al.,[87] Copyright 2015, American Physical Society.

(D) Mechanism and structures of suction. Left: illustration of the suction mechanism using the octopus-inspired adhesive.[52] Applying a negative pressure to the membrane causes it to deform and generates a cavity of low pressure. Leakage may occur on the contact interface surrounding the cavity. Right: other suction structures including the thin-walled suction cup,[88] crater,[89] and microcup.[90]

Modeling and testing underwater adhesion

Like dry adhesives, high-fidelity modeling and reliable experimental testing are essential for the design, fabrication, and optimization of underwater adhesives. Modeling and testing capabilities are especially necessary toward switchable underwater adhesion, either through guiding the active control of adhesion switching or by enabling efficient exploration of the parametric space for switchability. While the modeling and testing approaches developed for dry adhesives can be extended to underwater adhesives, the wet nature of underwater interfaces can lead to unique challenges. First, establishment of contact between an underwater adhesive and a submerged surface may be coupled to hydrodynamic effects as the two surfaces

approach each other.[83] Second, the hydration layer on wet surfaces can prevent the formation of molecular bridging (e.g., van der Waals forces) that underlie dry adhesion.[84–86] Consequently, alternative mechanisms such as chemical bonding and suction as reviewed above are needed to enhance adhesion on wet interfaces, making the modeling and testing of these mechanisms important for underwater adhesion.

Before elaborating on these challenges for underwater adhesion, we first briefly review the modeling and testing methodology for dry adhesion. A mechanistic adhesion model needs to address at least two questions: (1) how an adhesive interface is formed through the contact of two adherends, and (2) what mechanical load is required to separate the adhesive interface. The former question is the subject of contact mechanics. For example, the Hertz contact theory provides analytical solutions on how the contact area between two spherical, elastic bodies increases with the compressive contact force (Figure 4A).[91] Even for nominally flat surfaces, surface roughness can result in a force-dependent true contact area that is smaller than the apparent contact area.[92] For the latter question, the separation of adhesive interface often involves a coupling between the attractive forces on the interface and the stress and deformation in the adherends. As a result, the adhesive interface rarely separates in a uniform manner, but often proceeds incrementally, which can be modeled as the propagation of an interface crack and theoretically treated using concepts from fracture mechanics.[93] For example, the onset and stability of interface crack propagation can be evaluated by comparing an energetic driving force, known as the energy release rate G, with a parameter that describes the resistance to interface separation, known as the critical energy release rate G_c. For ideally smooth interfaces between elastic solids, G_c is equivalent to the thermodynamic work of adhesion W_{ad} if adhesion only comes from the change of surface energy upon contact.[93] This approach has led to adhesion theories such as the Johnson-Kendall-Roberts (JKR) theory for the adhesion between two elastic spheres (Figure 4A),[94] and the Kendall's peel force formula for peeling an elastic tape from a rigid substrate.[95,96] Alternatively, one can introduce a cohesive zone model at the tip of the interface crack, which prescribes a relationship between the adhesive traction across the interface and the relative separation of the interface (Figure 4A).[97,98] In both modeling approaches, adhesion is quantified as a mechanical parameter, either in terms of the critical energy release rate G_c or the traction-separation relation. Because of the multi-scale and multi-physics nature of adhesion mechanisms, it is often difficult to predict adhesion from first-principle calculations or molecular simulations. Instead, the parameters for adhesion are usually calibrated through experimental testing as illustrated in Figure 4B. The testing configurations such as the peel test,[99] the Double-Cantilever Beam (DCB) test[100] and the indentation test[93] allow measurements of G_c, while other configurations such as the tack test and lap-shear test provide measurements of adhesion strength (i.e., maximum force that can be sustained by an adhesive).[93]

The formation and separation of underwater adhesive interfaces entails additional complications. First, establishing contact between an adhesive and a submerged surface requires dehydration of the contact interface. The hydrodynamics of interface dehydration adds a new dimension to the contact mechanics. For example, the dry contact between an elastic slab and a rigid rough surface is governed by the applied contact pressure, the roughness profile, and deformation of the slab.[92] However, when fluid is present on the interface, the hydrodynamics of fluid drainage under contact pressure causes the process of contact formation to be time-dependent (Figure 4C).[83,101] Moreover, the hydrodynamics of interface

dehydration can be influenced by surface structures (Figure 4C),[35,102,103] or be coupled to bulk deformation of the contacting objects (Figure 4C).[87,104] Second, during the separation stage, strategies such as chemical bonding and suction have been developed to enhance adhesive bonding in the underwater environment. On one hand, effects of the chemical bonding can be readily modeled by adjusting the adhesion parameter (i.e., G_c or the traction-separation relation of cohesive zone). In case of switchable bonding,[73,74] the adhesion parameter can be altered according to external stimuli. On the other hand, modeling for the suction cup effect requires a new framework. Unlike conventional adhesive interface, which fails through crack propagation, suction relies on a pressure differential between an enclosed cavity and the environment.[52] Generating and maintaining the pressure differential depend on multiple factors, including large deformation of the enclosed cavity formed by the suction cup and the interface, pressure reduction within the cavity due to volume change, and leakage between the cavity and the environment (Figure 4D). Although suction has been widely exploited as an adhesion mechanism in dry or wet environments, its theoretical modeling has been lagging. This contrasts with the promising perspective of using suction to achieve switchable underwater adhesion and warrants additional discussion below.

Existing models for the suction effect are often specific to certain structures of the suction cup. For example, the suction force of thin-walled suction cups (Figure 4D) has been studied,[88,105] where analytical and experimental methods were used to explore the optimal preload for maximizing the suction force. However, deformation of the suction cup shell was not explicitly solved, which limits its utility for structural optimization. Another suction cup geometry is the crater consisting of a concave dimple on an elastomeric surface (Figure 4D).[106] Qiao et al.[89] modeled the suction force generated by a crater and explored its dependence on geometric and material parameters, while Wang et al.[67] further studied the effect of surface tension on the suction force. Both works considered large deformation of the crater and assumed that the pressure changes for the air trapped in the crater followed the idea gas law. Potential leakage between the trapped air in the crater and the ambient air was neglected. Parametric studies for the crater shape, material stiffness, and preload were carried out to determine their optimal combination for maximizing the suction force. The crater suction model was extended to underwater adhesion by Qiao et al.[107] where they adopted the assumption of water-vapor coexistence so that the ideal gas law could be applied to determine the suction pressure. In addition, they compared the suction performance of the crater in air and underwater and revealed the dependence of suction force on the submerged depth in water. Recently, Wang et al.[90] studied the underwater suction of microcups with thin-walled contact lips (Figure 4D). Specifically, they simplified structural components of the microcup to linear springs with different spring constants and developed a phenomenological model to capture the effects of microcup deformation and leakage on the evolution of suction pressure. To summarize, the models mentioned above were developed for specific suction cup structures with assumptions to simplify analysis, which limits their range of applicability. A predictive modeling framework incorporating the mechanics of suction cup deformation and the physics of contact, gas expansion, and leakage[108] is needed for the structural optimization of suction cups to achieve strong and yet switchable underwater adhesion.

Testing for underwater adhesion can follow similar experimental configurations as those for dry adhesion (Figure 4B).[84] Additional considerations may arise due to the underwater environment. For example, sufficient preload during the stage of making contact may be required to dehydrate the interface. Also, high-velocity adhesion testing may become

difficult when submerged. For structure-based underwater adhesives (e.g., octopus-inspired adhesive[52]), the tack test (Figure 4B) is commonly used since it can provide direct assessment on the adhesion strength and switchability. Here, an adhesive is pressed against a substrate with a prescribed speed until a certain preload is reached. The adhesive is then retracted until full detachment, during which the maximum tensile force is defined as the adhesion strength. This testing procedure focuses on the normal force perpendicular to the substrate. In practice, the adhesive may need to sustain shear forces parallel to the substrate, or more generally forces along an arbitrary angle to the substrate.[109] The orientation of force can influence the adhesive strength, especially for adhesion mechanisms that are sensitive to structural deformation (e.g., suction), but this effect has not been studied in detail. More general loading conditions including different combinations of normal and shear forces need to be explored in future experimental studies.

Data-driven adhesive design

In the recent decade, the prevalence of data-driven techniques, such as artificial intelligence, machine learning (ML), and optimization, has contributed to understanding and designing engineering systems at an accelerated pace compared with traditional approaches.[110–114] Additionally, the availability of powerful open-source ML libraries, such as scikit-learn[115] and PyTorch,[116] allows for physical insights to be gleaned from large datasets generated from both experiments and advanced numerical simulations. Data-driven inverse designs are preferred in design tasks, which use the target functionality as input and the ideal material/structure parameters as output, often powered by ML techniques such as neural networks. This design strategy differs from the traditional design philosophy where designers conceive a fixed configuration as a starting point and rely heavily on labor-intensive trial-and-errors. Established data-driven inverse design approaches are numerous. Here, we briefly explore two categories–global optimization and generative models—and how they can be applied to underwater adhesive design.

Global optimization uses an algorithm to find an optimal solution of the target function given a design space. Examples such as Bayesian optimization (BO) and genetic algorithm (GA) are often seen in inverse design tasks. BO systematically finds the optimum of a black-box function without assumption of its form, allowing for solving difficult optimization problems.[117–120] Specifically, BO uses the information obtained by all prior measurements to inform the "next-best" measurement to conduct, allowing for identifying optimal configurations with high data efficiency. Son et al. designed adhesive fibrils using a combination of BO and fibrils to outperform previously proposed designs by over 70%.[121] Rooney et al. developed an autonomous platform for adhesive material optimization using a Bayesian optimizer. By intelligently informing users which adhesive formulations to test next, the data-driven workflow shortens the design cycles of new adhesive materials.[122] GA is an optimization algorithm that mimics the natural selection process.[123] A random population of candidates is first initialized, and each candidate has a fitness value related to the optimization objective. Genetic operations such as crossover and mutations are then applied to obtain the next generation of candidates. GA iterations are repeated until some acceptance criteria or a time constraint is reached. Kim et al. optimized the adhesive pillar shape using deep learning and genetic algorithms in a large design space.[124] Using a trained neural network surrogate model that replaces more expensive finite element simulation, genetic optimization was conducted to maximize a fitness function favoring the uniform interfacial stress distribution.

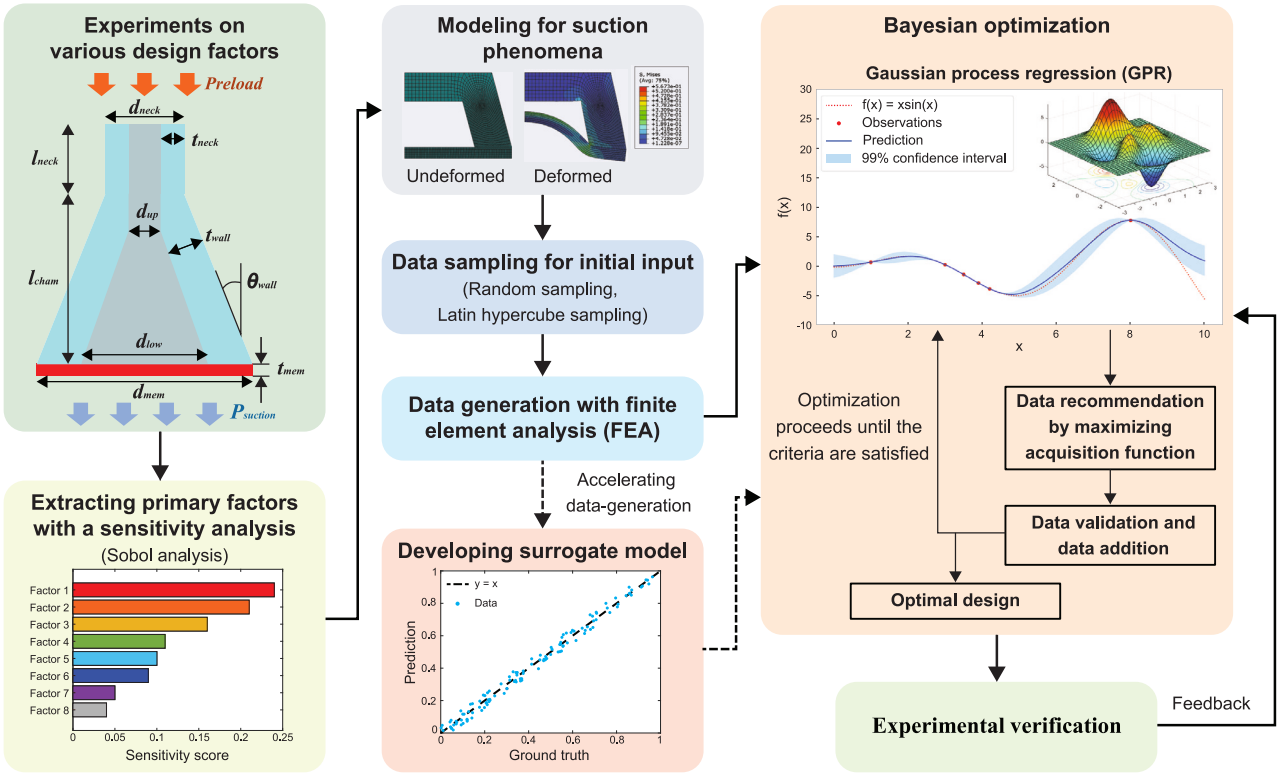

Figure 5. Workflow chart for data-driven adhesive design

Experiments are conducted on various design factors, and primary factors are selected via sensitivity analysis. Modeling of underwater suction phenomena is performed, and data are generated through FEA. Bayesian optimization is carried out using the generated data, and experimental verification on the optimized design is conducted.

Generative models are deep learning models that encode the high-dimensional design space into a lower-dimensional latent space and generate new data using knowledge embedded in the latent space. Two common generative models are generative adversarial networks (GANs) and variational autoencoders (VAEs). A GAN consists of two competing deep neural networks: a generator that generates candidates, and a discriminator that distinguishes these candidates from the true data distribution.[125] The generator is trained to increase the error rate of the discriminator, and the discriminator is trained to minimize that. The end goal is for the generator to generate high-quality data, in our context, high-performance adhesive materials or devices. A VAE consists of an encoder and a decoder.[126] The encoder encodes the design parameters together with the corresponding performance into a latent space. By forcing a given prior distribution (for example, those corresponding to high-performance designs) in the latent space, new high-performance designs can be reconstructed by the decoder. Nevertheless, to the best of our knowledge, the use of generative models in designing adhesives has not been abundant.

Although data-driven design in the context of underwater adhesives has not been widely explored, there are promising research directions in this area such as multi-objective optimization for adhesive strength and switchability. Additionally, because the chemistry of the adhesives plays an important role in the aqueous environment, a high-dimensional design space is created by both geometric and chemical variables, motivating the use of more advanced generative ML models. The power of generative models enables us to explore a much larger design space, such as

device shapes characterized by Bézier curves with many control points,[121,127] potentially with the addition of chemical species. Another beneficial practice is to train neural network surrogate models to accelerate expensive simulations such as finite element analysis. Finally, as an illustrating example, Figure 5 proposes a flow diagram of how data-driven design approaches such as BO can be potentially applied to design underwater adhesives, where various design parameters are inputs, and potentially multiple objectives such as the adhesive strength and the switching ratio are outputs. Therefore, assisted by fast-developing ML techniques, data-driven design can potentially allow researchers to explore a larger design space of underwater adhesives in a fast, autonomous fashion.

Conclusions and outlook

In summary, the strong adherence to wet or underwater surfaces poses a significant obstacle in applications such as tissue adhesion and underwater robotics. This challenge becomes more evident when there is a need for switchable adhesion, which requires quick attachment, high adhesive capacity, and easy release. Nevertheless, nature has provided ample inspiration in designing switchable, underwater adhesives. The natural examples surveyed here have inspired a diverse range of bioinspired strategies to control adhesion. Many of the reported examples rely solely on one biomimetic strategy. We envision that future designs would benefit from combining multiple biomimetic design strategies to create switchable adhesives with improved performance. While such a combinatorial strategy is enticing, it would raise the number of design parameters needed to optimize the performance of the adhesive. This further reinforces the need for a multidisciplinary approach to accelerate discovery and establish foundational understanding to build more complex systems. Through this type of approach, we expect to not only better understand the natural organisms that display switchable adhesion but allow for the creation of bioinspired materials that far exceed what is currently available.

ACKNOWLEDGMENTS

C.L., R.T., and M.D.B. acknowledge support from the National Science Foundation (NSF) under the DMREF program (award no.: 2119105). K.W. and B.P.L. acknowledge support from NSF under the DMREF program (award no.: 2119019). H.S. and R.L. acknowledge support from NSF under the DMREF program (award no.: 2118878). J.J., B.Z., and G.X.G. acknowledge support from NSF under the DMREF program (award no.: 2119276). We also acknowledge Daniel Albacarys for help with schematics in Figure 1.

AUTHOR CONTRIBUTIONS

All authors contributed to the writing of the manuscript.

DECLARATION OF INTERESTS

The authors declare no competing interests.

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