

## Supplementary Information

### Synthesis of Dopamine Methacrylamide (DMA)

Sodium borate ( $\text{Na}_2\text{B}_4\text{O}_7 \cdot 10\text{H}_2\text{O}$ , 16.5 mmol, 6.29 g) and sodium bicarbonate ( $\text{NaHCO}_3$ , 30 mmol, 2.52 g) were dissolved in 60 mL of distilled water. Dopamine hydrochloride (20.6 mmol, 3.90 g) was added after the solution was bubbled for 90 minutes with nitrogen. Methacrylate anhydride (20 mmol, 3 mL) solution in 15 mL of degassed tetrahydrofuran (THF) was prepared and added dropwise into the mixture. The pH of the reaction mixture was monitored and maintained moderately basic (pH 8-9) by adding 1 M NaOH solution dropwise. The reaction mixture was stirred for 17 hours at room temperature under nitrogen atmosphere, then the solution was washed twice with 30 mL of ethyl acetate and the resulting aqueous layer was filtered under vacuum. The obtained solution was acidified to pH 2 with 6 M HCl solution. The mixture was extracted three times with 50 mL of ethyl acetate, and the organic layer was dried with  $\text{MgSO}_4$ . The solution was concentrated to about 15 mL under vacuum, and precipitated in 220 mL of 0°C hexane to yield the product. The final solid powder was dried overnight in a vacuum oven.

$^1\text{H}$  NMR (Bruker 400M, 400MHz,  $\text{DMSO-}d_6$ ):  $\delta$ (ppm) 7.93(t, 1H), 6.62-6.42(m, 3H), 5.61(t, 1H), 5.29(p, 1H), 3.22(m, 2H), 2.55(m, 2H), 1.83(s, 3H).

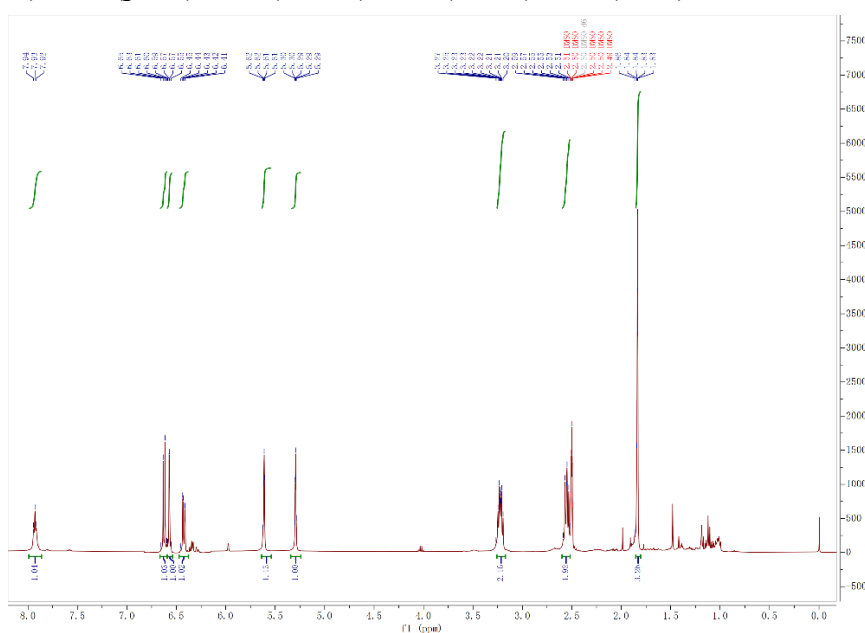


Figure S1.  $^1\text{H}$  NMR measurement of DMA

## **Synthesis of poly (dopamine methacrylate-co-2-methoxyethyl acrylate) (p(DMA-co-MEA))**

DMA (0.6 mmol, 132.8 mg), 2-methoxyethyl acrylate (MEA, 1.2 mmol, 154  $\mu$ L) and azobisisobutyronitrile (AIBN, 0.036 mmol, 5.91 mg) were added to 4 mL of *N,N*-dimethylformamide (DMF). The solution was bubbled with nitrogen for 30 minutes and then polymerization was carried out for 3 hours at 60°C. The resulting solution was added dropwise to pentane with stirring to precipitate the copolymer. The copolymer was dissolved in methylene chloride and precipitate in pentane for purification. The resulting product was dried overnight in a vacuum oven and stored in pentane.

### **The operation method of coating dopamine polymers on micro-wedge-shaped adhesive pads:**

Because the wedge-shaped structure is not flat, the traditional dip-coating method cannot be adopted, and the shape is irregular, the solution tends to accumulate at the bottom and the surface of the wedge-shaped tip, resulting in an irregular structure. Key points: pre-cleaning with ultrasonics, controlling the amount of dip-coating solution, and the sequence of dip-coating.

**Cleaning:** Before coating, put the bristle piece into a dropper and add anhydrous ethanol for ultrasonic oscillation.

**Standstill:** After removing it, let it stand for a while, waiting for the anhydrous ethanol to evaporate completely.

#### **Specific coating techniques:**

- ① Use a pipette to draw the prepared suspension, tilting the pipette.
- ② Push out the liquid as evenly as possible, generally the liquid droplet at the top, naturally wetting the microstructure surface, encapsulating the wedge-shaped structure surface according to the tension of the liquid droplet itself.
- ③ Follow the direction of the grooves to wet the suspension, not irregularly dipping the liquid from the pipette onto the bristle piece.

**Standstill:** Put it in a clean, ventilated environment and let it air dry naturally (try not to do this in dusty weather, don't wait for it to dry by the window, surface dust will still have some impact).

After the anhydrous ethanol in the suspension has air-dried, DOPA polymers remain on the surface of the micro wedge-shaped structure.

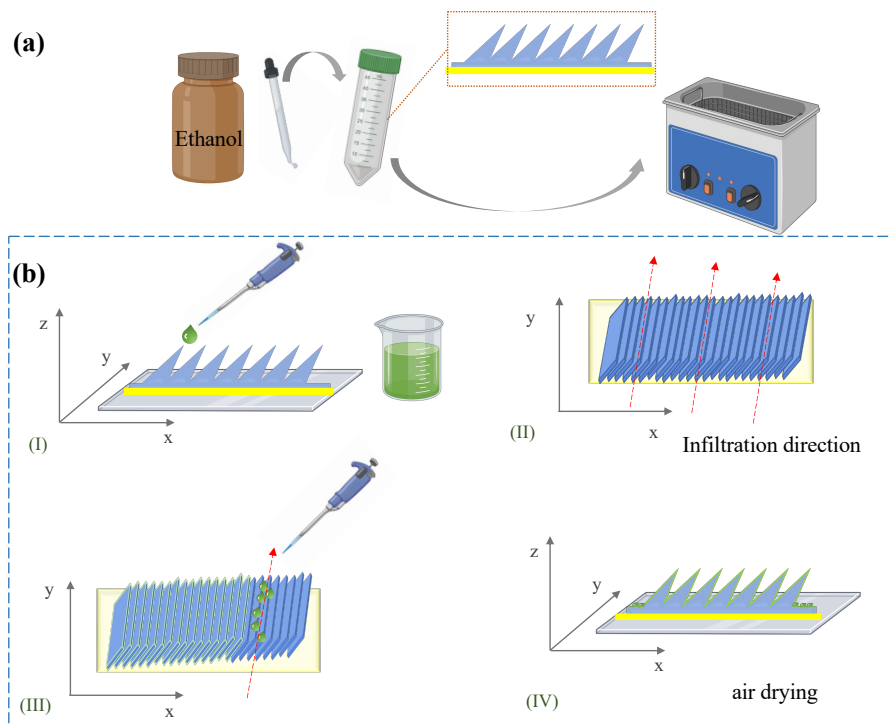


Figure S2. The method of applying dopa polymer on the adhesion pad.

### Specific conjectural analysis of the mechanism part

Mussel adhesive proteins contain an unusual amino acid, 3,4-dihydroxy-L-phenylalanine (DOPA), which is largely the reason for their cohesion and adhesion.<sup>1</sup> The strong bond between dopa and the surfaces of organic and inorganic substances is formed in the presence of water.<sup>2</sup>

#### In terms of detailed analysis:

During the adhesion phase, the wedge structure of the Microscopic Adhesion Polymer (MAPMCs) uses the direction of water flow to promote an environment conducive to forming chemical bonds. The flow pushes the cations in the NaCl solution towards the MAPMC surface, encouraging the formation of ionic bonds, or other types of chemical bonds, between the cations and the MAPMC surface coating.<sup>3,4</sup> Essentially, the water flow acts as a delivery mechanism for these cations to the MAPMC surface, thus promoting adhesion.

On the other hand, during the detachment process, the water flow helps to disrupt this environment and facilitates the breaking of these bonds. When the water flow is reversed or increased, it could dilute the concentration of cations at the surface of the MAPMC, reducing the strength of the ionic bonds and facilitating release. Alternatively, it could physically interfere with the bonds and make it more energetically favorable for them to break.

So, water flow doesn't necessarily have opposite effects on chemical bonding itself. Instead, it plays a role in changing the environment or conditions in which these bonds are formed or broken, thus affecting the adhesion and detachment processes.

#### During the adhesion process, with the applied preload,

① the micro-wedge structure of MAPMC gradually collapses. (In this process, the finite element local magnification image in Figure 1(b) can be seen, the fluid velocity is consistent with the direction of the structural collapse, and the fluid will promote the collapse of the microstructure).

② the mussel-inspired polymer coating on the surface of MAPMC adheres to the surface of the object.

③ After contacting the object, a chemical adhesion effect occurs (especially for NaCl solution as the liquid, in the finite element analysis local magnification image in Figure 1(b), the fluid velocity in the area near the microstructure is significantly increased, to some extent enhancing the contact between the cations in the NaCl solution and the mussel-inspired polymer coating on the MAPMC surface, making it easier to form chemical bond adhesion.<sup>3,4</sup>

④ the object is adhered to by MAPMC.

[1] Guvendiren M, Brass D A, Messersmith P B, et al. Adhesion of DOPA-functionalized model membranes to hard and soft surfaces[J]. The Journal of adhesion, 2009, 85(9): 631-645.

[2] Lee H, Scherer N F, Messersmith P B. Single-molecule mechanics of mussel adhesion[J]. Proceedings of the National Academy of Sciences, 2006, 103(35): 12999-13003.

[3] Xiang L, Zhang J, Wang W, et al. Nanomechanics of  $\pi$ -cation- $\pi$  interaction with implications for bio-inspired wet adhesion[J]. Acta Biomaterialia, 2020, 117: 294-301.

[4] Gebbie M A, Wei W, Schrader A M, et al. Tuning underwater adhesion with cation- $\pi$  interactions[J]. Nature chemistry, 2017, 9(5): 473-479.

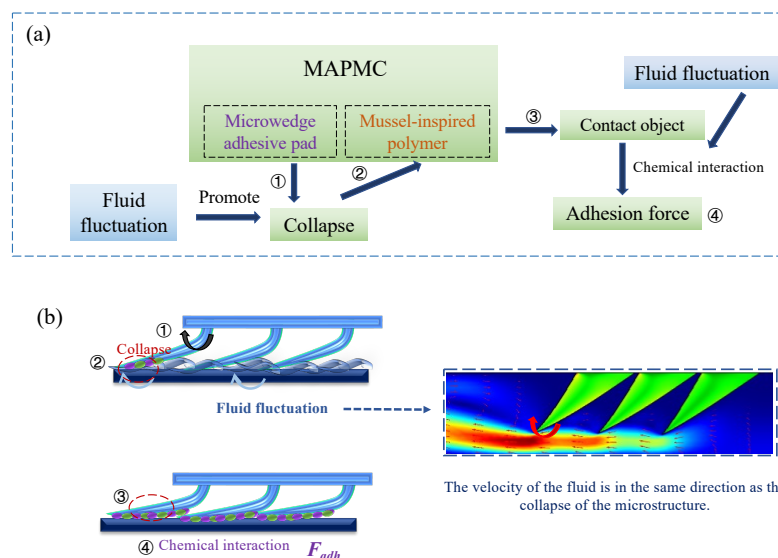


Figure S3. adhesion process

**In the detachment process, with the removal of prepressure,**

① the micro-wedge structure of MAPMC gradually returns to its original state

② in this process, the micro-wedge structure releases the elastic strain energy stored from previous deformation

③ at the same time, as can be seen from the local magnified image in Figure 2(b), the speed of the fluid around the microstructure is consistent with the direction of the structure's restoration, and the fluid will promote the recovery of the microstructure

④ MAPMC restores its original structure, and the object detaches.

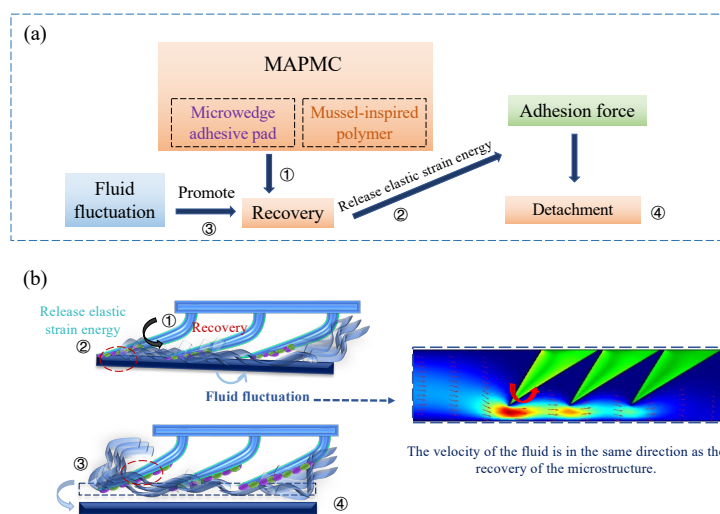


Figure S4. detachment process

Table S1 The comparison of the adhesion performance of MAPMC with other types of materials and structures.

	operation mode	operational object	mechanism	structural characteristic	main materials
Octopus-inspired adhesive pad <sup>5</sup>	pressure differential	objects with flat and hard surfaces	suction, capillary force	isotropic structure	PDMS
Dytiscus lapponicus-inspired adhesive pad <sup>6</sup>	pressure differential	objects with flat and hard surfaces	suction, capillary force	isotropic structure	PDMS
Tree-Frog adhesive pad <sup>7</sup>	temperature variation	objects with flat and hard surfaces	capillary force	isotropic structure	biomimetic hydrogel
Gecko-inspired mushroom-structured adhesive pad <sup>8</sup>	temperature variation	objects with flat and hard surfaces	chemical adhesion	isotropic structure	PDMS, (acrylamide-isopropyl acrylamide-acrylic acid)/Fe (PAAmPNIPAAm-PAA/Fe), p(DMA-co-MEA)
<b>MAPMC</b>	collapse	<b>curved</b> , hard objects and easily <b>deformable soft</b> objects	chemical adhesion	<b>Anisotropic structures</b>	silicone rubber materials, p(DMA-co-MEA)

[5] Baik S, Lee H J, Kim D W, et al. Capillarity-enhanced organ-attachable adhesive with highly drainable wrinkled octopus-inspired architectures[J]. ACS applied materials & interfaces, 2019, 11(29): 25674-25681.

- [6] Li S, Liu H, Tian H, et al. Dytiscus lapponicus-Inspired Structure with High Adhesion in Dry and Underwater Environments[J]. ACS Applied Materials & Interfaces, 2021, 13(35): 42287-42296.
- [7] Zhang B, Jia L, Jiang J, et al. Biomimetic microstructured hydrogels with thermal-triggered switchable underwater adhesion and stable antiswelling property[J]. ACS Applied Materials & Interfaces, 2021, 13(30): 36574-36586.
- [8] Zhang Y, Ma S, Li B, et al. Gecko's feet-inspired self-peeling switchable dry/wet adhesive[J]. Chemistry of Materials, 2021, 33(8): 2785-2795.

Indeed, it is noteworthy to mention that the performance of MAPMC somewhat pales in comparison to other analogous underwater adhesion apparatus when assessed based on the mass of the objects it can adhere to in an aquatic environment. Its distinctive attributes encompass a micro-wedge-shaped external morphology and structurally anisotropic features. Employing the principle of collapse and subsequent restoration of its structure, MAPMC demonstrates the ability to engage and disengage objects submerged underwater. On this basis, MAPMC can be integrated into a mechanical claw like an array, realizing the adhesion and detachment of soft objects in the underwater environment. In particular, a detailed investigation employing finite element analysis reveals the existence of a coupling effect between the micro-wedge architecture and the directionality of the water current. This dynamic interaction serves to enhance the adhesive and detaching operations of the MAPMC, lending credence to its functionality and potential applications.

