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Bio-inspired adhesive hydrogel for wound healing

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ABSTRACT

Wound healing introduces a series of interesting challenges in healthcare, such as open wounds, burn wounds, and chronic infection wounds. Hence more accurate and effective biomedical materials are expected, for which emerging hydrogels with unique and excellent properties have demonstrated great potential. When they are applied as wound dressings, tissue patches, plugging agent, drug carriers, and biosensors, *etc*., there are urgent requirements for adhesive capabilities. The existence of interfacial water, soft tissue and moving surfaces, safety, and harmlessness, however, constitutes major constraints for medical adhesive hydrogels. In nature, organisms often adhere through unique structures or specific chemical components, which stimulates inspirations for medical hydrogels to overcome the above challenges. In this review, we will classify adhesive hydrogels into glue-like adhesives and tape-like patches according to their form. First, this review will introduce the principles and design ideas of biomedical adhesive hydrogels. Then, as biomedical materials, the applications and effects of their interactions with organisms through adhesion will be summarized. Finally, this review will summarize the achievements, challenges, and future development of bio-inspired adhesive hydrogel towards wound healing.

1. Introduction

Wound healing introduces a series of challenges in human healthcare, such as open wounds, burn wounds, chronic infection wounds, *etc*., resulting in physical and mental impairment to patients and a huge social burden. [1–4] To deal with these issues, the appeal of more appropriate and effective biomedical materials is increasingly urgent. Compared with traditional materials represented by metals, ceramics, and polymers, emerging hydrogels with various unique and excellent properties possess great potentials for wound healing. [5–9] Hydrogel is a kind of three-dimensional polymer network with high water content which holds great promise in the biomedical field. [6,10] Specifically, the macromolecular matrix of hydrogels confers preeminent biosecurity and adjustable biodegradability. [11–14] The highly hydrated, soft and elastic matrix conforms to the physicochemical properties of tissues. [15–17] The spatial network structure facilitates material exchange with organisms, providing a range of loading, release and adsorption capacities. [18–21] In addition, specific designs of hydrogels can further endow them with customized functions such as electrical conductivity, antibacterial, and adhesion. [22–25]

Different wound conditions and scenarios are suitable for different paradigms of hydrogels. More precisely, preformed medical patches always need to attach to the tissues for protection and repairing. [26–30] When dealing with irregular wounds, shape-adaptive adhesive hydrogels are often desired. [31–34] For bleeding wounds, sealing agents are supposed to bond tightly and resist certain stress. [35–38] Despite the variety, without exception, adhesion functions of hydrogels are urgently needed in wound healing. [39–41] Commercially available adhesives are, however, barely satisfactory. For example, cyanoacrylates tend to have uncontrolled adhesion times, rigid adhesion interfaces, and even potential toxicity [42,43], while fibrin glues provide relatively weak adhesion strength. [44,45] On the other hand, many researchers have obtained inspiration from the adhesion behavior of creatures such as tree frogs, [46–48] octopuses, [49–51] and mussels [52–55] in nature. Such bio-inspired hydrogels often have unique and advanced structural or molecular designs, such as reusable suction cup structures [56–59] or non-toxic catechol chemistry, [60–62] which enables safer, more robust, and more controllable adhesion behaviors.

According to the application forms, these bio-inspired hydrogels for wound healing can be divided into two main paradigms – adhesives and patches (Fig. 1). Figuratively, adhesives are like glue in the initial state of application. The advantages include injectability and in-situ adhesion.

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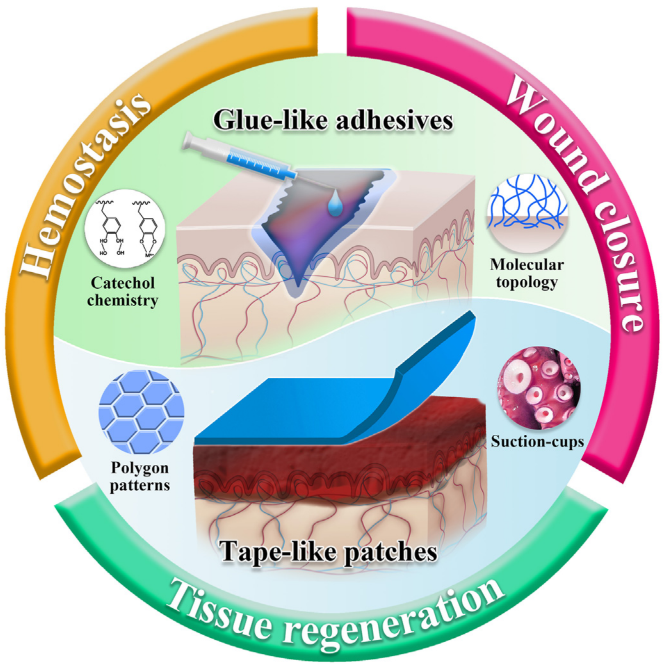

Fig. 1. Glue-like adhesives and tape-like patches are two common paradigms of bio-inspired adhesive hydrogels for wound healing. The adhesives described in this review reach the wound by injection, *etc.*, and are shaped in situ. While pre-formed patches are applied directly to the wound surface, a series of processes such as hemostasis, wound closure and tissue regeneration can be effectively achieved by applying the appropriate adhesive hydrogel to the desired wound surface.

That said, the amorphous character may also lead to difficulty in pre-designing the surface structure and manipulating the adhesion behavior. [32,63] For the other, hydrogel patches are like adhesive tapes. Their adhesive surfaces with desired structural and molecular properties are often pre-designed and pre-manufactured, and these properties can be highly controllable and reversible. However, the leakage of adaptability to shape may result in inappropriateness for tissues. [64–66] Therefore, in general, adhesive-like adhesives were very suitable for irregular wound defects, perforations, gaps, and wounds requiring in-situ sealing to stop bleeding. While tape-like patches, due to their better structural design and higher controllability, can be more accurately applied to various wound surfaces requiring programmed treatment, so as to achieve reversible, controllable and responsive adhesion and other functions.

Following such classification, we will conduct a systematic review of bio-inspired adhesive hydrogels from the perspective of wound healing. According to the two paradigms of glue-like adhesives and tape-like patches, the design strategies, principles and effects of their interactions with organisms through adhesion will be summarized respectively. Finally, this review will discuss the conclusion and perspective on current stages, challenges, and future development of bio-inspired adhesive hydrogel for biomedicine. Innovatively, this review aims to inspect bio-inspired adhesive hydrogels in the perspective of interactions between different paradigm materials and wounds in different situations, so as to provide a more refined, systematic, and organized approach to follow-up research in the interdisciplinary fields of biomedicine and material science.

2. Bio-inspired glue-like adhesives for wound healing

Human wounds often encounter irregular defects and bleeding. In this scenario, the application of adhesives is of great significance. [67–70] Their shape adaptability is conducive to fill gaps and cavities, [28,39,71,72] thereby blocking bleeding points and covering irregular wounds. [35,73–78] In order to eliminate ambiguity, the adhesives described in this chapter refer to hydrogels that are firstly applied in the form of glue, and then repair defects, adhesive cracks, or seal leakages through adhesion behavior. The often-mentioned tissue sealant is also covered in this broad concept of adhesives reservedly. It can often be injected into a specific site. [41,63,72] Then cohesion mediates its gelation, while force on the tissue interface mediates adhesion. Its in-situ adhesion and adaptive molding characteristics endow it unique advantage of adapting to irregular defects and leak shapes, as well as penetrating deep into ruptured tissue. [32,63,66] At the level of force, the main factor that determines whether glue-like adhesives can be successfully applied to wounds is the design and adjustment of the spatiotemporal properties of their cohesion and interfacial adhesion forces, which affect their gelation and adhesion behaviors macroscopically. [79,80]

Cohesion and interfacial adhesion are two concomitant characteristics, both from bottom-up action at the molecular level. Cohesion emphasizes the interaction within the hydrogel, while interfacial adhesion emphasizes the interaction between the hydrogel and the tissue interface. [41,80,81] The common chemical factors that form cohesion are Michael addition, [82–84] Schiff base, [85,86] free radical polymerization, [35,87] click chemistry [88], and so on. The physical level is more common in hydrogen bonding, charge interaction, molecular chain entanglement, and topology. [89,90] Specific discussions on the above-mentioned forces have been summarized from the perspective of injectable hydrogels in some excellent reviews. [32,79,91] Mediated by these molecular-level forces, general adhesives are injected into the wound in fluidic form, and then undergo shape-adaptive gelation to form a tight fit. The contribution of this cohesion-mediated gelation process to the adhesion behavior is an important issue that is easily overlooked by researchers but will be discussed in depth in this review.

On the other hand, the common adhesion-derived factors of adhesives can be classified into two categories according to the spatial scale: intermolecular interaction and molecule-network interaction. Intermolecular interactions mainly rely on non-specific adhesion groups or specific pairing of molecules. A typical example is the adhesion of mussels in nature through the catechol-rich mussel foot protein. [53,92] Because the catechol groups contained in these proteins can produce rich interactions with various substrates, such as hydrogen bonds, coordination bonds, hydrophobic interactions, π-π interactions, and so on. [52,54] Numerous adhesives have been designed by introducing catechol-related groups. The introduction methods include direct incorporation of small molecules, covalent grafting, incorporation of nanoparticles, and the like. [93–96] Similarly, some other adhesives with intermolecular interactions mediated adhesion were also designed, inspired by bacterial adhesions or by responses such as molecular pairing in organisms. [26,97,98] The molecule-network interaction represented by entanglement and topology is another form of adhesion, which can occur independently of specific groups. [41,99,100] In brief, when molecular chains diffuse between two network interfaces, entanglement leads to the occurrence of adhesion. And when the molecular chains are further cross-linked into a third network, stronger topological adhesion occurs. In addition to focusing on the origin of the adhesives, it is worth mentioning that many researchers have also focused the design of strategies on the phase behavior of adhesives, which were inspired by sandcastle worms. [101,102] The coacervation phenomenon of liquid-liquid phase separation occurs through the design of the forces such as the charge and hydrogen bond of the polymer chain, so as to maintain its stability of under underwater injection. [103–105]

As mentioned above, the cohesion-mediated gelation behavior and the interfacial adhesion-mediated adhesion behavior are not independent of each other. It can even be said that shape-adaptive gelation behavior is perfectly suited for adhesion, which is mainly manifested in macroscopic and molecular topologies. [41,63] The tissue surface is often not smooth, but has certain roughness, gaps, and holes. [106–108] At the macro-topological level, mechanical interlocking structures are formed when adhesives fill in for gelation. At this time, the elasticity of the hydrogel and the interaction force with the biological tissue can resist the force that separates the interface, providing adhesion. At the molecular topological level, the gelation behavior of adhesives is in line with the conditions for topohesion. Before gelation, the linear polymer penetrates into the tissue, and after gelation occurs, it crosslinks itself into another network. A firm topohesion is born from this. When combined with the intramolecular interactions strategy, such a close combination of macroscopic and molecular scales increases its effective area and further enhances the adhesion.

However, because of their amorphous, the adhesives are difficult to have an ordered adhesive micro-nano structure, so the common adhesion strategies are mainly molecular-related. Its adhesion is often designed to be irreversible, and can only be removed under specific conditions requires degradation or solation. [79,91] In addition, the dynamic process of internal gelation and tissue adhesion often takes a certain amount of time, and various physical and chemical influences can easily lead to adhesion failure. Of course, many studies have also attempted to actively control this adhesion process and prevent failure. [39,109]

For wound, the most unique and important applications of adhesives are hemostasis and closure. [67,68,112] Organisms heal spontaneously after injury. The first stage of healing is to stop the bleeding by closing the damaged blood vessel through blood coagulation. However, sometimes if there is a severe wound resulting in uncontrolled massive hemorrhage, the body's natural hemostasis process is not effective enough. [39,67] After hemostasis, adhesives need to cover the wound, hindering external adverse physicochemical factors and the risk of infection. During wound repair, adhesives are expected to generate tissue adhesion, acting like extracellular matrix.

Faced with this series of problems, many adhesives are designed with different entry points. Hong et al. designed a class of hemostatic hydrogels that gelate and adhere in seconds, and then withstand pressures up to 290 mmHg for powerful and rapid hemostasis (Fig. 2a). [37] Inspired by the extracellular matrix of connective tissue, this class of adhesives consists of grafted gelatin and hyaluronic acid photosensitive biomacromolecules and photoinitiators constituting prepolymers. The gelation was initiated by light, and the free radical polymerization of gelatin-based polymers and the photo-generated aldehyde groups of hyaluronic acid-based polymers occurred simultaneously. Then the aldehyde group further reacts with the amino group inside the hydrogel or on the surface of the tissue to undergo Schiff base reaction, which enhances the cohesion and adhesion. Such light-responsive adhesives are promising for massive wound bleeding.

Inspired by the heal-injury process, Chen et al. designed a type of adhesives for wound repair, which has the functions of hemostasis, antibacterial, and angiogenesis (Fig. 2b). [20] The hydrogel consists of a prepolymer of benzaldehyde-terminated four-arm polyethylene glycol and dodecyl-modified chitosan. Cohesion is maintained by a dynamic aromatic Schiff base reaction, while adhesion arises from the insertion and anchoring of dodecyl to the cell membrane. This adhesive exerts excellent hemostatic and adhesion functions in vivo; it can rapidly repair acute tissue damage such as vascular hemorrhage, and promote tissue repair.

Sundaram et al. used a hybrid strategy of hydrogels and inorganic ions to incorporate potassium aluminum sulfate and calcium chloride into chitosan hydrogels as wound hemostatic adhesives (Fig. 2c). [113] Among them, potassium aluminum sulfate plays the role of

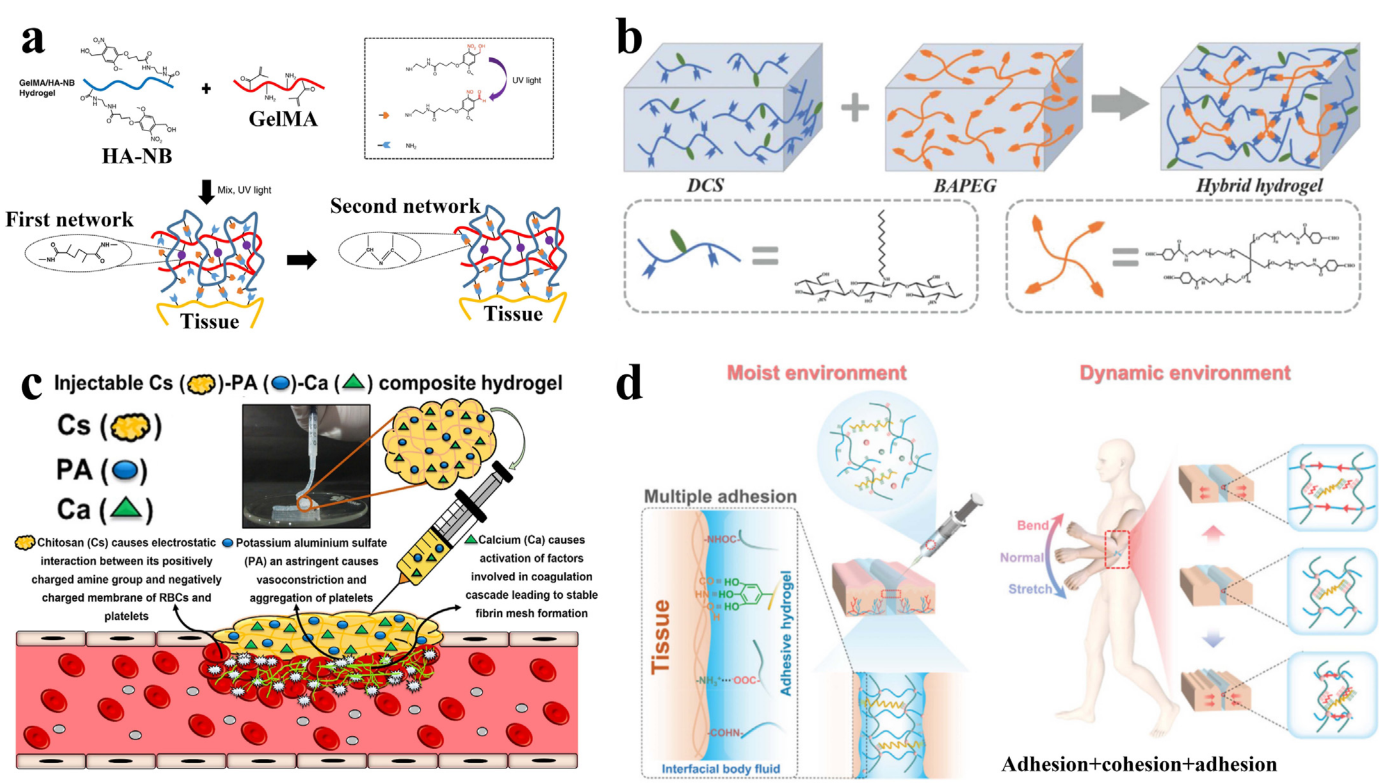

Fig. 2. The applications of glue-like hydrogel adhesives. a) Chemical structures of photo-triggered adhesives. The fast-formed strong chemical bonds endow it with excellent cohesion and interfacial adhesion, allowing it to withstand enormous pressures after a few seconds of gelation. Reproduced with permission. [37] Copyright 2019, Springer Nature. b) Adhesives formed by chitosan derivatives and modified four-arm PEG. When the two components are mixed, the action of aromatic Schiff base maintains the cohesion, and dodecyl has the ability to adhere to cells. Reproduced with permission. [20] Copyright 2018, Wiley-VCH. c) Inorganic ion hybrid hydrogel adhesives. The shear-thinning properties of polymers regulate cohesion, while mucoadhesion mediates interactions with tissue interfaces. Inorganic components have vasoconstriction and coagulation abilities. Reproduced with permission. [110] Copyright 2022, Elsevier. d) Adhesives with excellent adhesion and mechanical properties, which come from abundant and strong cohesion and adhesion. This adhesive is resistant to adverse moisture and dynamic environments. Reproduced with permission. [111] Copyright 2022, Wiley-VCH.

vasoconstrictor-potassium, and calcium chloride plays the role of coagulation activator. The cohesive force regulating its injection and molding is believed to come from the shear thinning of the polymer, and the adhesion behavior comes from the mucoadhesive of chitosan.

Chen et al. focused on the mechanical properties and energy dissipation of adhesives after molding, achieving tough and firm adhesion on the joint wound surface (Fig. 2d). [111] The hydrogel forms cohesion and interfacial adhesion through strong amide reaction, and maintains strong mechanical elasticity. Due to the grafting of gallic acid, more abundant forces resulted in more diverse forces at the adhesion interface and excellent dissipative ability, which consolidated the adhesion.

Looking at the above studies, "one-size-fits-all" and "perfect" adhesives do not exist because the problems faced by wound healing are too complex. For example, a fast gelation time may be detrimental to its conformation to shape, but a slow one may lead to dilution of adhesives by tissue fluid; strong cohesion, adhesion is often non-degradable and irreversible, and an overly dynamic structure may lead to poor mechanical performance. However, it is believed that in the future, adhesives that are more suitable for wound healing will be developed.

3. Bio-inspired tape-like patches for wound healing

Likewise, to remove ambiguity, patches described herein refer specifically to preformed tape-like hydrogels. [21,56,64,114] It can directly adhere to the wound surface under the action of pressure and play a repairing role without additional gelation process. The pre-forming of patches comes with some inherent disadvantages compared to the adhesives above. For example, it does not fit completely tightly to the wound shape, resulting in a smaller effective contact area. [63] The non-injectable nature also limits its application in living organisms at the spatial scale.

Patches have many design strategies similar to adhesives. Intermolecular interactions are also applicable in patches. The most common strategy is still the introduction of catechol functional groups. [21,59,115] Adhesion groups with certain specificity such as nucleobases are also used in examples. [116,117] Such adhesive groups present on the patch surface can interact with molecules at the tissue interface to mediate adhesion. The topohesion mentioned above can exist in a bridging manner between the patch and the organization interface, providing firmer adhesion and better dissipation. [90,118]

The special surface structure that can be designed also brings rich ideas different from adhesives to its adhesive. The surface structure can produce strong adhesion together with reversible and controllable adhesion behavior without chemical contamination. Common structural paradigms include octopus-inspired suckers, [59,119] bee sting-inspired mechanical interlocking arrays, [120,121] and tree frog-inspired polygonal patterns. [122] All have the potential to be designed as wound patches.

The sucker structure of the aquatic animal octopus is worth learning from the adhesion strategy. [123–125] The source of its adsorption force is mainly the negative pressure generated by the suction cup and the surface tension of the wet adhesion interface. Specifically, when the suction cup made of elastic material is pre-stressed to discharge the fluid in the chamber, the restoring force will make the hydraulic pressure in the chamber lower than the external hydraulic pressure, and the negative pressure is generated thereby. At the same time, the liquid layer at the interface makes the effective contact area of the wet interface much larger than that of the solid-solid interface. It always generates capillary force which is greater than van der Waals force of solid-solid interface, while maintaining the negative pressure in the chamber. [126–129] In some more detailed studies, the protuberance structure in the suction cup makes two chambers formed after pre-pressure applied. [127] After water enters the upper chamber due to capillary force, the negative pressure below is further enhanced.

Inspired by it, various hydrogel patches with corresponding micro arrays for wound healing were developed. Huang et al. designed a versatile wound patch using tailorable micro-suction cup patterns (Fig. 3a). [18] The elastomeric material, Eco-flex, was cast using a mold method to produce tailorable suction cup arrays. Then, the gelatin-based photosensitive hydrogel tailored to the wound shape was polymerized on demand. Such materials can reversibly adhere to wounds, induce cell adhesion, deliver drugs, and ultimately promote wound healing. Zhang et al. developed a hydrogel patch with combined adhesion, antibacterial and drug delivery capabilities (Fig. 3b). [59] The elastic base material of the sucker array is mainly a blend of polydopamine and gelatin, and each sucker structure in the array has a diameter of several hundred microns with a protuberance inside. In the synergistic effect of suction cups and catecholamines, the array produced a peeling adhesion of more than 1 N/cm^2 at both dry and wet interfaces and was repeated in ten adhesion-peel cycles. This patch shows great potential for wound repair.

The related structure of mechanical interlocking is another type of adhesion strategy for patches which is mainly inspired by some kinds of parasite. [130,131] During the adhesion process, the structures are embedded in the matrix surface of the wound, and in turn to be bonded and then mechanically interlocks. [132] The microarray corresponding to this strategy is often an intumescent microneedle. Seung et al. reported an swellable double-layer microneedle adhesive. [120] Before contacting with water, the smooth tapered needle can be easily inserted into the tissue in a dry and hard state. By absorbing the water in the tissue, a rapid increase in the cross-sectional area is occurred, achieving local tissue deformation and subsequent interlocking, which eventually provides adhesion. As the hydrogel swells, the soft microneedle tip can be removed without significantly damaging the tissue or facing the microneedle breakage during rigid removal. The researchers believe this strategy has potential in wound repair such as skin grafting. It ensures continuous contact between the grafted skin and underlying tissue, avoiding separation between tissue layers caused by movement or shearing. A similar strategy is also used in the design of Eun et al. (Fig. 3c). [115] It uses silk fibroin as a raw material to prepare microneedle patches that are mechanically interlocked by swelling. It has excellent adhesion on the wound surface, which can resist the adverse physical and chemical factors of moisture and dynamics. The adhesion of the patch achieves rapid hemostatic sealing and wound closure, thereby healing the wound.

To achieve reversible, firm, and rapid underwater adhesion, drainage on two adjacent surfaces is an important issue. The existence of the water layer has a certain negative influence on the time of the interface formation, the properties of the hydrogel, and the force at the interface. [133–135] In nature, the interconnected groove structure on the foot pad of the tree frog and the sticky disc of the clingfish is believed to promote the drainage of water between the interface. [136–138] Rao et al. reported a hydrogel inspired by it. The hydrogel is designed as a hexagonal facet pattern separated by interconnecting grooves (Fig. 3d). [122] When the hydrogel is in contact with the adhered interface, this interconnected surface groove can serve as a channel for rapid drainage when in contact under water. The dynamic bonds of the hydrogel based on chemical forces will form a bridge with the adhered surface, and finally form a good contact. Such a pattern also has the effect of improving the flexibility of the gel and preventing continuous cracks from spreading to the entire interface. These two effects significantly enhance energy dissipation and delay the interfacial peeling time, resulting in strong but also reversible adhesion. The authors believe that it has the potential to become a reusable wound dressing. In addition to the design of surface structures, the anisotropy design of adhesive patches is also unique. Wang et al. designed a class of double-sided "Janus" patch. [21] One side has a mussel-inspired adhesive structure, while the other side is inspired by the superhydrophilic nature of the eyeball surface to prevent adhesion. This kind of design is of great significance in the programmed precision repair of wound surface. In view of the above studies, adhesive patches, with their unique structural design, are expected to bring more precise and tunable new methods to the field of wound repair.

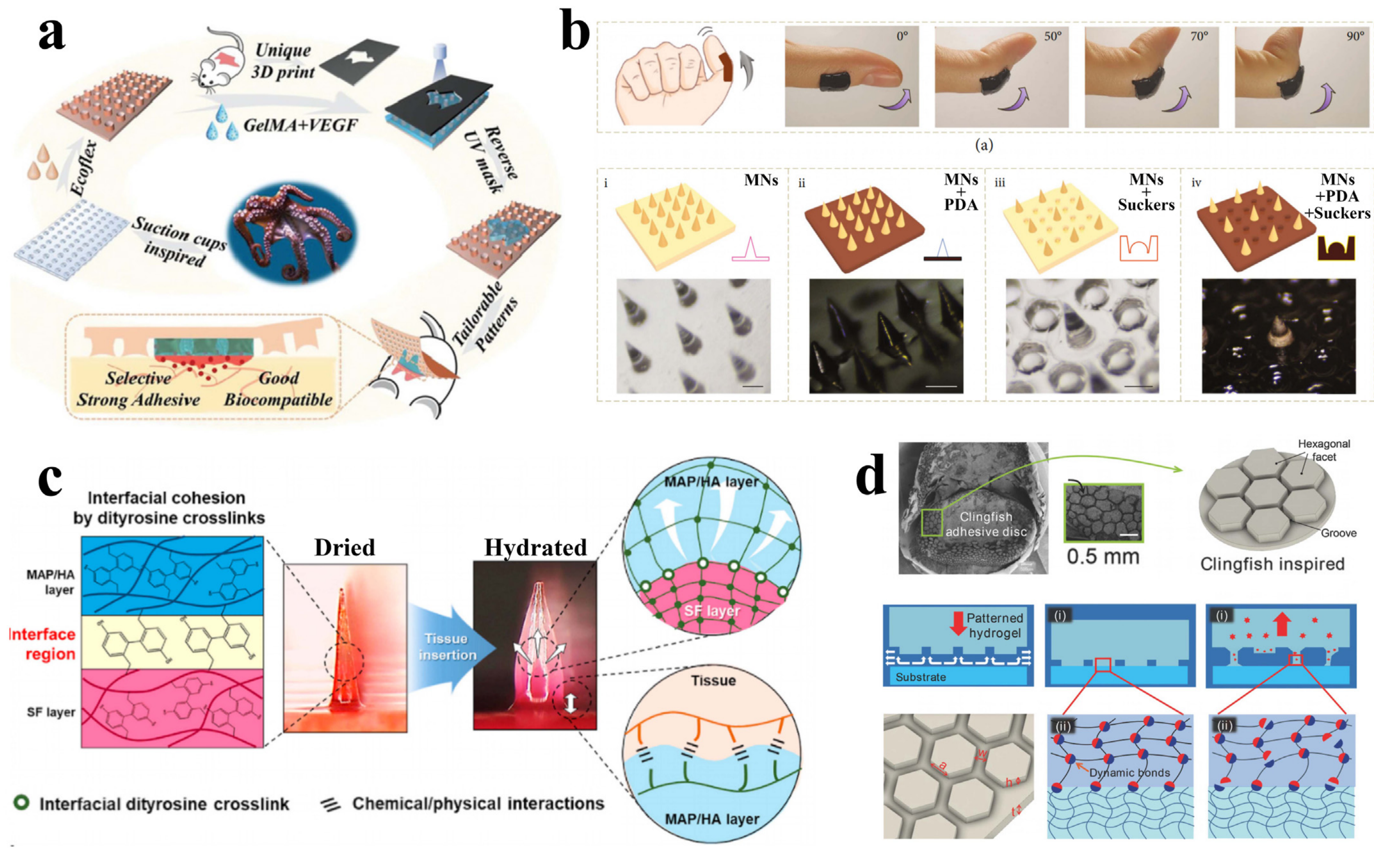

Fig. 3. The applications of glue-like hydrogel adhesives. a) Octopus-inspired hydrogel patch for wound repair. First, a patch with a micro suction cup array was prepared, and then a drug-loaded hydrogel was custom-cured on it according to the wound shape. Reproduced with permission. [18] Copyright 2021, Wiley-VCH. b) Patches with octopus-inspired arrays, made from chemical reactions between dopamine and gelatin, enable multiple adhesion. It can be used for wound closure and drug delivery, *etc*. Reproduced with permission. [59] Copyright 2020, Science and Technology Review Publishing House. c) The hydrogel microneedles are designed to have a bilayer structure. After insertion into the wound tissue, the outer layers are mechanically interlocked by swelling, resulting in a strong anchorage. Reproduced with permission. [115] Copyright 2019, Elsevier. d) Clingfish-inspired hydrogel patch. The hexagonal facet pattern on the surface is beneficial to expel interfacial water and increase the effective adhesion area. Furthermore, the dissipative ability of the hydrogel dynamic bonds enhances adhesion. Reproduced with permission. [111] Copyright 2018, Wiley-VCH.

4. Conclusions and perspectives

Wound repair is still a thorny problem in medicine, and the emerging diversiform bio-inspired adhesive hydrogel undoubtedly provides a promising solution. Such hydrogels can be roughly divided into two categories, glue-like adhesives and tape-like patches, and their design strategies and application scenarios have their own merits. Glue-like adhesives are more suitable for irregular shaped wounds and can stop bleeding well. Their adhesion design strategies tend to be bottom-up at the molecular level. Patches, on the other hand, achieve more precise and procedural behavior through structural design. Its adhesion strategy can be at the molecular level as well as at the top-down structural level. In summary, it has different design ideas and application scenarios.

Although many "patches" and "adhesives" were designed, at present, there is still no "universal" or perfect adhesive hydrogel wound dressing. As mentioned before, common hydrogels can only address the staged problems in the entire repair process. This is due to the complexity of wound repair. Various types of adhesive hydrogels themselves also face many challenges, such as the firmness and reversibility that are difficult to tune during adhesion. This still requires more interdisciplinary research in materials science, medicine, *etc*. Of course, with the development of various multifunctional, stimuli-responsive, and practical hydrogels, we are also pleased to see the development of bio-inspired adhesive hydrogel towards a more versatile, controllable, precise and high-performance direction. These studies will bring great advancement to wound repair and have impactful potentials.

Ethics statement

Authors declare human or animal ethics approval was not needed for this study.

Author contribution

W. L. and P. L. conceived the idea; W. L. wrote the manuscript; all authors were involved with the scientific discussion and revision of the article.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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