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INVITED REVIEW

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Advances in dermatological application of GelMA hydrogel microneedles

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Abstract

Background: Compared with systemic administration methods like injection and oral administration, traditional transdermal drug delivery has the advantages of rapid onset of activity and low side effects. However, hydrophilic drugs and bioactive substances are often unsuitable for traditional transdermal drug delivery.

Methods: The application of microneedles made from gelatin methylacryloyl (GelMA) has greatly expanded thepossibilities for skin transdermal drug delivery. We have reviewed the latest literatures about the dermatological application of GelMA hydrogel microneedles in recent years using Google Scholar, PubMed and Springer.

Results: GelMA hydrogel microneedles exhibit huge potency in the diagnosis and treatment of skin diseases, and this technology also brings broad application prospects for subcutaneous micro-invasive transdermal targeted drug delivery, which mainly used in skin tissue fluid collection, local substance delivery and wound healing.

Conclusion: With in-depth research on GelMA hydrogel, this technology will bring more breakthroughs and developments in the clinical diagnosis and treatment of skin diseases.

KEYWORDS

gelatin methylacryloyl, interstitial fluid collection, microneedle, substance delivery, wound healing

1 | BACKGROUND

Traditionally, transdermal drugs are absorbed locally through the skin or capillaries, and then enter the blood circulation to produce either local or systemic therapeutic effects. As a natural protective barrier, the stratum corneum of skin prevents water loss from the body and blocks the invasion of pathogenic microorganisms, which has a high density and low water content that severely limits the efficiency of

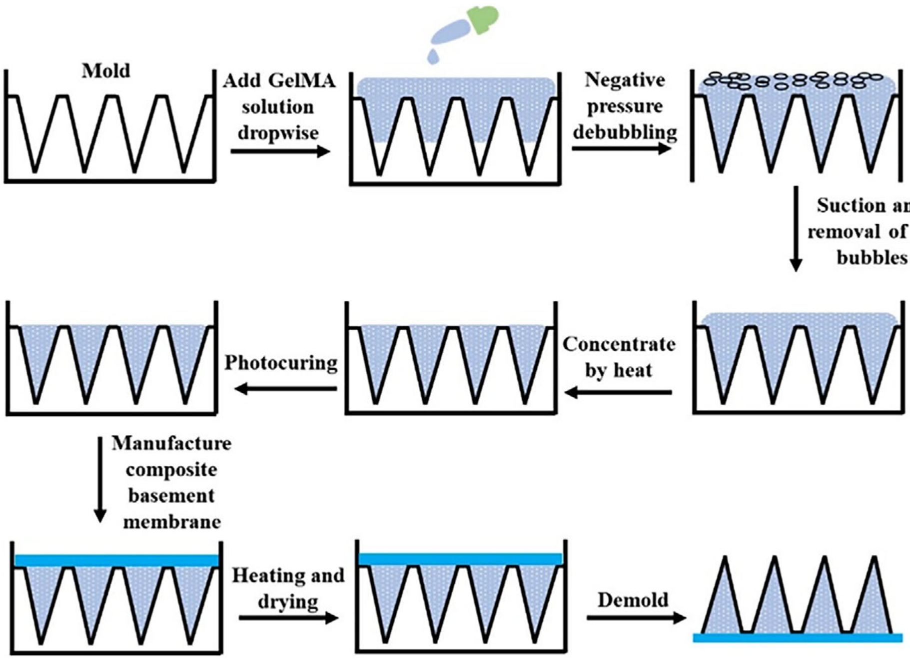

FIGURE 1 The basic fabrication process of GelMA microneedles.

transdermal absorption of many topical drugs, especially hydrophilic or macromolecular drugs.[1] With the development of transdermal drug delivery technology, the application of biomaterials has greatly enriched the scope of the application of drug delivery, and provided new ideas for precise diagnosis and targeted therapy of clinical diseases.

Microneedles, a new transdermal drug delivery technique, have received much attention in dermatological field. Microneedles generally consist of multiple matrix micro-needles less than 2 mm in length and are usually classified as hydrogel, solid, hollow and dissolving microneedle types.[2,3] Among them, the application of hydrogel microneedles in transdermal drug delivery has been a research hotspot in medical field. This type of microneedle has the advantages of strong permeability, convenient administration and few side effects.[1] It directly releases the loaded components into the deep layer of the skin through puncture, and the tiny holes formed can quickly heal while avoiding the liver first-pass effect and gastrointestinal degradation by systemic drug administration.[4] Furthermore, soluble microneedles made of biodegradable polymers loaded with drugs are current research hotspots, which can dissolve and release the drugs encapsulated inside the microneedles.[5]

Gelatin methylacryloyl (GelMA) is a photocurable gel material that has been studied more popularly in recent years (see Figure 1 for the preparation process), which is mainly composed of methacrylamide groups and supplemented by methacrylate groups with characteristics such as high-water content, low immunogenicity and strong adjustability.[6] GelMA possesses both arginine-glycine-aspartate (RGD) sequences and matrix metalloproteinase sequences, thereby retaining the key properties of gelatin[7] and playing a key roles in skin wound healing, morphogenesis and tissue repair.[8] With the development of microneedle preparation and crosslinking technology, GelMA hydrogel has gained wide application attention in tumors, anaesthesia, dentistry, etc., which is also used for model construction,[9] drug screening[10] and 3D printing.[11] Based on the adjustable mechanical properties and swelling capacity, GelMA microneedles with biocompatibility and bioactivity have become a hot spot for current applications in the field of skin diseases, ranging from simple cell culture scaffolds to more complex tissue engineering platforms and delivery of substances such as drugs, genes or growth factors.[6,12]

GelMA microneedles exhibit huge potency in skin diagnosis and treatment of skin diseases, and this technology also brings broad application prospects for subcutaneous micro-invasive transdermal targeted drug delivery. This article mainly reviews the application of GelMA hydrogel microneedles in skin tissue fluid collection, local substance delivery and wound healing (Figure 2).

2 | COLLECTION OF SKIN INTERSTITIAL FLUID

Blood samples are a common standard for examining patients' biochemical indicators and judging the disease's clinical outcome. Although blood collection is minimally invasive and easy to operate for skin diseases, the systemic blood sample information cannot fully reflect the microenvironmental state of local skin lesions. Furthermore, interstitial fluid, as the most common liquid, surrounds tissues or cells to build the bridge between cells and blood.[13] Hence, the liquid in intradermal space has been found as a meaningful way for the diagnosis of skin disease.[14] As a local body fluid rich in biomarkers, skin interstitial fluid can be used as a supplement for the clinical tests of blood and

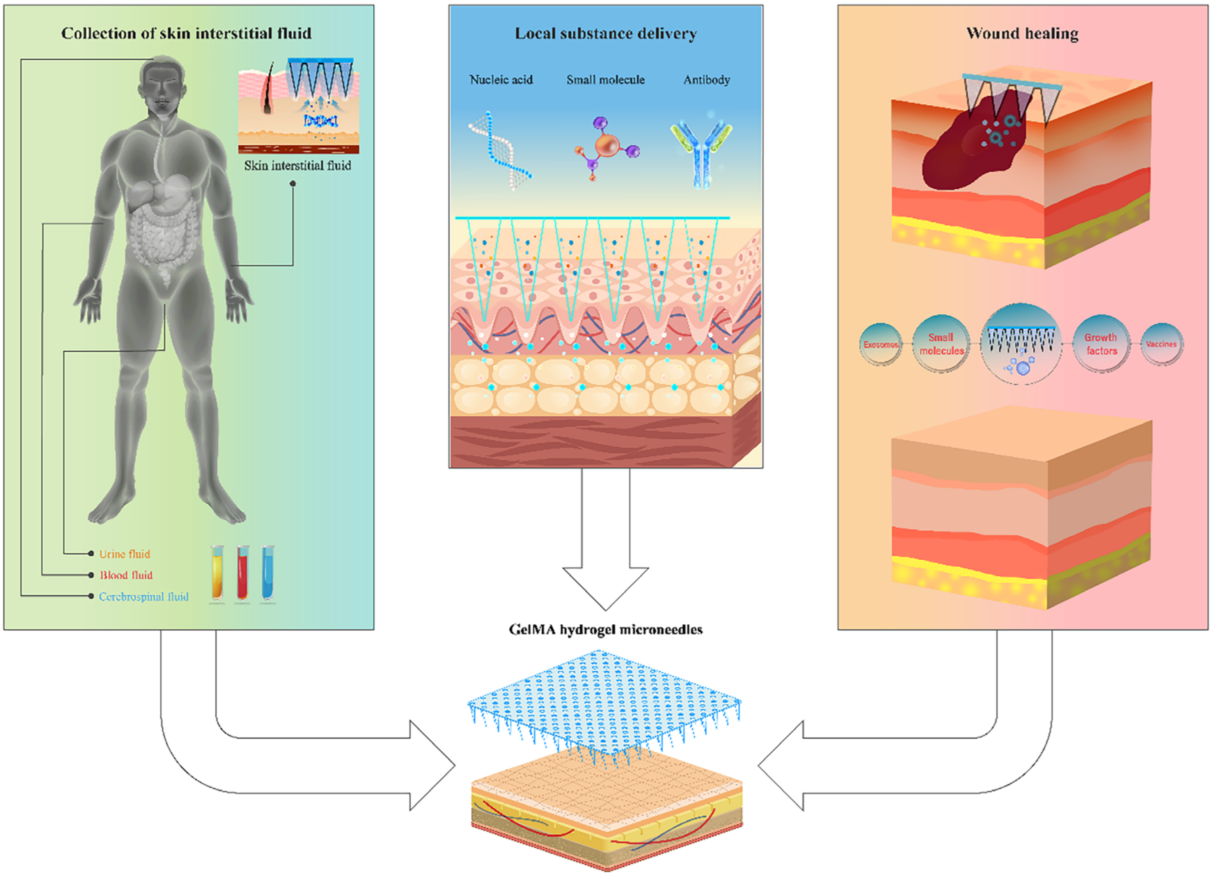

FIGURE 2 The multiple application directions of GelMA microneedles targeting human skin. Through the transdermal route, GelMA hydrogel microneedles can target human skin and be used in a variety of directions such as skin tissue fluid collection, local substance delivery and wound healing.

urine, and is of great significance for the discovery and clinical diagnosis of skin disease biomarkers.[15,16]

The classical method for extracting skin tissue fluid has great application limitations, such as infection risk by biosensors placed in the body,[17] skin irritation and frequent calibration by reverse iontophoresis,[18] tissue damage and time consuming by open-flow microperfusion and microdialysis.[19,20] Due to the good swelling properties, GelMA microneedles can directly absorb skin interstitial fluid when applied locally on the skin, which have broad application prospects for the detection of local drug or metabolite concentration and biomarker monitoring.[6]

In terms of normal skin tissue fluid extraction, considering the swelling characteristics and mechanical properties of GelMA microneedles with different concentrations, the researchers found that the GelMA microneedles with a concentration high than 20% had a stronger skin penetration ability, and the concentration of the drug extracted by microneedles in the skin model of agarose gel was equal to the actual concentration of drug in the model, and the glucose extracted in the local tissue fluid of the microtargeted skin in the rat model was comparable to the concentration in serum.[21] In the application of the agarose skin model, the recovery rate of GelMA microtargeted urea can reach 98%, and it also exhibits no cytotoxicity in keratinocytes.[22] Microneedle patches made of GelMA and hyaluronic acid methacryloyl (HAMA) possess good mechanical properties with a high swelling rate of 700%, which can be used to efficiently extract interstitial skin fluid in agarose and fresh porcine skin models for efficient quantification of miRNA and copper ions.[23] For lesion tissue fluid extraction, researchers constructed GelMA/graphene oxide (GO) microneedles using both GelMA and GO, which have strong mechanical properties and swelling capacity to collect 21.34-μL interstitial skin fluid within 30 min, for analyzing miRNA in the local microenvironment of psoriasis.[24] Furthermore, the multifunctional skin patch prepared by CNTs/graphene/GelMA has good air permeability and swelling due to its porous nanofibrous structure, and it can dynamically monitor wound tension by absorbing interstitial fluid in a rat abdominal skin wound model conditions and humidity changes.[25]

3 | LOCAL SUBSTANCE DELIVERY

The percutaneous absorption efficiency of drugs is generally affected by the physical and chemical properties of the drug (such as molecular weight and lipophilicity), the drug dosage (such as ointment, gel, patch and spray) and the physiological state of the skin (such as, the thickness of the stratum corneum, inflammatory state and skin temperature).[26] Therefore, most drugs do not work well by transdermal infiltration, while the microneedles prepared by GelMA hydrogel have a porous network structure, which can realize the local material delivery of active ingredients, such as nucleic acid, antibody and small molecule drugs, thereby greatly improving the application range of transdermal drugs.

In terms of active substance delivery, researchers prepared GelMA hydrogels microneedles wrapped in poly-β-amino ester (PBAE) nanoparticles for the local release of plasmid DNA after local transdermal delivery through the skin, which in turn has great potency for the applications in tissue regeneration and cancer therapy.[27] In addition, novel P-glycoprotein antibody-modified GelMA porous hydrogel particles were applied to capture multidrug-resistant tumor cells.[28] Regarding small-molecule drug delivery, researchers used carboxymethyl β-cyclodextrin and GelMA for the amidation reaction to make microneedles, and the hydrophobic inner cavity of the former was loaded with hydrophobic curcumin, the resulting microneedles could deliver drugs deep into the gel and thus inhibit the growth of tumor clusters in an agarose gel-grown B16F10 cell mass model.[29]

For hydrophilic drug delivery, microneedle preparation can be performed by directly mixing hydrophilic drugs with GelMA hydrogels. Researchers prepared microneedles by mixing lidocaine hydrochloride with GelMA. Compared with lidocaine patches, the local absorption of the drug was more efficient after GelMA microneedle administration, and the skin action site of rats could return to normal appearance within a short period without significant skin adverse reactions.[30] Degradable microneedles prepared from the water-soluble antitumor drug adriamycin and GelMA can penetrate the mouse stratum corneum effectively while exerting potent antitumor effects on melanoma A375 cells.[31] In addition, by increasing the crosslinking density of GelMA microneedles, the sustained release time of the coated hypoglycemic drug metformin can be extended accordingly, and the hypoglycemic effect on diabetic mice is better than that of subcutaneous injection.[32]

4 | WOUND HEALING

Wound management is an important challenge in the field of clinical care. Traditional skin wounds often burden patients with a huge psychological burden because of complicated dressing and lengthy healing process. Due to the degradable effect, the microneedles can load exosomes,[33] small molecules,[34] vaccines,[35] growth factors and other biologically active substances to the wound site in the form of microneedles or dressings to promote angiogenesis and collagen deposition, thereby promoting tissue repair and wound healing.[36]

Regarding wound healing, drugs and bioactive substances that promote wound repair can be mixed into GelMA hydrogel to make microneedles for the wound healing of damaged skin. In the animal model of diabetic skin wounds, researchers mixed tazarotene, HUVEC-derived exosomes and GelMA hydrogel to make microneedle patches, which could accelerate the healing of diabetic mice skin wounds by promoting cell proliferation, migration and angiogenesis while maintaining the activity of exosomes and tazarotene.[33] Besides, microneedles supported by insulin in a specific ratio to GelMA hydrogel can accelerate the healing of diabetic wounds, reduce inflammatory responses, and enhance collagen deposition at regenerated tissue sites.[37] Furthermore, GelMA hydrogel can also be loaded with oxygen-carrying haemoglobin to promote the internal oxygen supply of diabetic rat skin, thereby promoting the healing of full-thickness skin wounds.[38]

In addition, GelMA hydrogel can also be loaded with antibacterial components or growth factors to promote the wound healing process. The researchers immobilized Lactobacillus reuteri in GelMA hydrogel through covalent crosslinking of HAMA. In vivo and in vitro experiments confirmed that the prepared antibacterial dressing has good antibacterial and anti-inflammatory capabilities, which promoted the healing of infected wounds and tissue regeneration.[39] The antimicrobial peptide Tet213 was added to the hydrogel prepared by mixing methacrylate recombinant human proprotein (MeTro) with GelMA. The resulting dressing could be used for sutureless wound closure, thereby inhibiting bacterial infection and promoting wound healing.[40] The composite hydrogel formed by photocrosslinking of human amniotic membrane and GelMA has also been verified to be used for skin wound reconstruction and regeneration.[41] Similarly, growth factors such as vascular endothelial growth factor,[42] vasoconstrictor peptide endothelin-1 (ET-1),[43] fibroblast growth factor[44] have all been reported to be effective in biological dressings made of GelMA hydrogels to facilitate the wound healing process.

5 | CONCLUSION AND PERSPECTIVE

As reported, a variety of biological materials are used in the building of skin microneedles, and different materials can be mixed together to furtherly improve the physicochemical and biological properties of microneedles. GelMA commonly combined with other materials to construct a more ideal microneedle system, such as HAMA,[23] GO,[24] carboxymethyl β-cyclodextrin,[29] PBAE,[27] PEGDA.[33] Furthermore, silk fibroin meth acryloyl (SilMA) was also used to extract tissue fluids and transport the extracted fluids for color displaying in a pH-responsive colorimetric microneedle.[45]

The application of hydrogel biomaterials greatly promoted the development of clinical diagnosis and treatment of skin diseases, especially boosted the conversion application in the field of transdermal drug delivery. From traditional molecular drugs to biologically active substances (including exosomes, growth factors, Etc), the application research of GelMA hydrogel as a substance carrier has proved its potential feasibility in dermatological application. Compared with traditional transdermal drug delivery, firstly, GelMA microneedles were more efficient for drug delivery by penetrating the skin directly and bypassing the skin's natural barrier. Secondly, GelMA microneedles exhibited more controllable administration and rapider onset of action for special skin medication area. Thirdly, a wider range of drugs can be loaded in GelMA microneedles, such as small molecule, growth factor, exosomes. However, GelMA hydrogel microneedles may collect skin interstitial fluid with limited volume and the contamination of blood,[46,47] and the drug-coated with microneedles also challenged by small drug loading volume, uncontrolled drug activity and limited drug action.[48,49] Furthermore, the practical application of such biomaterials still faces multiple problems, such as how to determine the physical parameters of GelMA microneedles under different physiological states of the skin, and the safety, pharmacokinetics and pharmacodynamics still need to be furtherly investigated through clinical trials. With in-depth research on GelMA hydrogel, this technology will bring more breakthroughs and developments in the clinical diagnosis and treatment of skin diseases.

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CONFLICT OF INTEREST STATEMENT

The authors declare no conflict of interest.

DATA AVAILABILITY STATEMENT

Data are openly available in a public repository.

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