REVIEWS

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Advances in flexible and wearable pH sensors for wound healing monitoring

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Abstract: Wound healing has been recognized as a complex and dynamic regeneration process and attracted increasing interests on its management. For effective wound healing management, a continuous monitoring on the wound healing based on sensors is essential. Since pH has been found to play an important role on wound healing process, a variety of pH sensors systems for wound healing monitoring have been greatly developed in recent years. Among these pH sensors, flexible and wearable pH sensors which can be incorporated with wound dressing have gained much attention. In this review, the recent advances in the development of flexible and wearable pH sensors for wound healing monitoring have been comprehensive summarized from the range of optical and electrochemical bases.

Key words: pH sensor; wound healing monitoring; flexible and wearable sensors; optical and electrochemical mechanism

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1. Introduction

As is known to us, skin plays an important role in protecting body from the external environment[1, 2]. Once the skin is wounded, human life will be in risk[1, 2]. Generally, it is a complex process for wound healing to repair the injured skin and even the underlying tissues[3, 4]. Typically, this process includes hemostasis, inflammation, proliferation, and remodeling phases[3–5]. Only all of these phases are completed in its proper sequence, the wound would be healed well. However, when these stages are influenced or prolonged due some unexpected factors for example diabetes mellitus, it may result in non-healing chronic wound[4–7]. Chronic wounds may greatly affect patients' quality of life cause rising healthcare costs, and then lead to a large social and medical burden.

It is found that using appropriate wound dressing to cover the wound would accelerate the wound healing process[8–10]. However, the wound dressing may be changed frequently according to the wounds situations and their healing cases. While, changing wound dressings frequently may interrupt the normal wound healing process, cause a second injury onto the wound and pain to the patients. Paradoxically, one cannot find the wound healing situations without removing the dressing. So, it is expected that there are some advanced technology in wound dressing which can provide some useful information to monitor the treatment efficacy earlier than the wound closure such as the proper time to change wound dressing or whether the wound needs redressing at all[4]. These information are crucial for the clinician to make treatment decisions and develop a targeted therapeutic approach for wound management and monitoring.

Ideally, the wound monitoring technologies and strategies should be simple, rapid, precise, could be completed during a standard medical consultation and cause minimal or no discomfort to the patient[11–17]. One option is to develop flexible and wearable pH sensors since the pH value is obviously changed during the wound healing process[18–23]. Herein, the recent advances in the flexible and wearable pH sensors based on different sensitive mechanism for wound healing monitoring were summarized and comprehensively discussed.

2. pH in wound healing process

Generally, wound healing process is a dynamic process including inflammation, proliferation, and remodeling stages[1–6], as can be seen from Figs. 1 (a)–1(c). Normally, the pH of healthy skin is in the range of 4–6, slightly acidic to impede bacterial proliferation[18–23]. While the pH value on the wound site is found to vary according to the wound healing stages as suggested in Figs. 1(d) and 1(e). Once an injury creates an acute wound, the underlying tissue was exposed and the pH regulated at 7.4, which may allow bacterial growth and in the risk of promoting infection. As a countering, a temporary acidosis will occur during the inflammation stages (Fig. 1(d))[18]. The pH of chronic wounds is more alkaline (oscillating between pH 7 and 9), as suggested in Fig. 1(e)[18], thus may be susceptible to infections due to bacterial incursion and colonization. Consequently, pH is believed to be able to suggest the present state of the wound. So, it is meaningful to develop pH sensors for wound monitoring[20–27].

3. pH sensors for wound monitoring

The pH notation is a measurement of H^+ concentration and can be measured in the range of 0–14 on the negative logarithmic scale ($pH = -\log[H^+]$)[28]. There are number of methods to determine the pH value of a milieu, i.e. the interstitial milieu of wounds. These methods contain a wide range from

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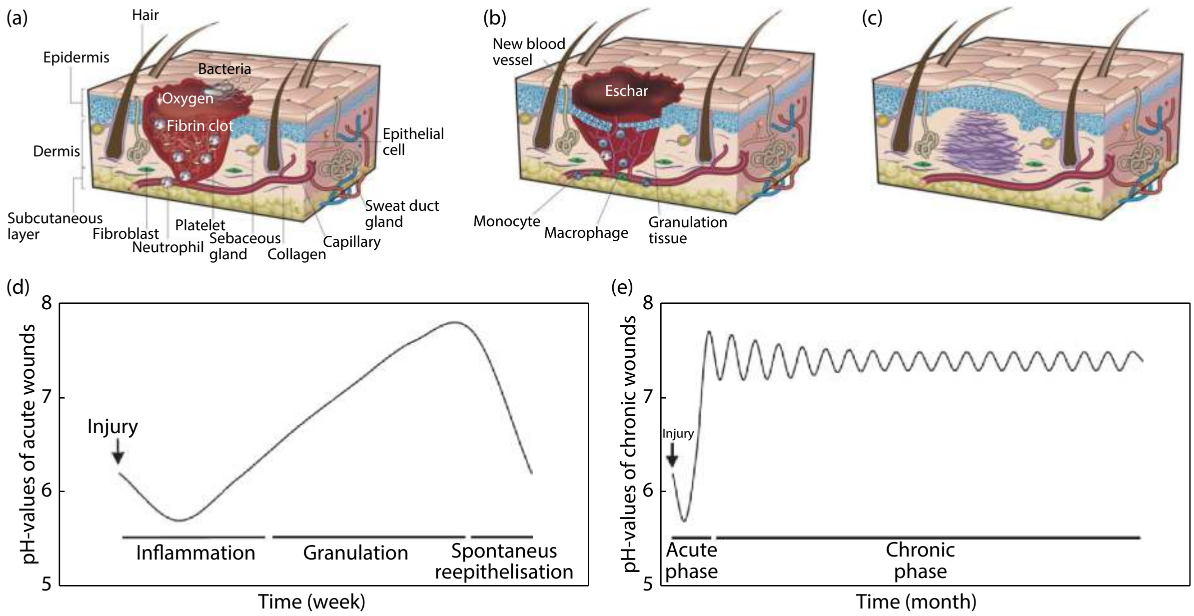

Fig. 1. (Color online) Wound healing phases: (a) inflammation, (b) proliferation, and (c) remodeling. Reprinted with permission from Ref. [5]. The time course of pH milieu in different wounds such as (d) acute and (e) chronic wounds[18].

optical to electrochemical ones[29]. In optical methods, pH sensors are relying on the pH sensitive dye characteristics which could possess absorption, excitation and emission signals especially in the visible light region[29–31]. Electrochemical pH sensors are mainly based on measurement of the pH sensitive potential, current, or impedance[29]. For wound healing monitoring, the pH sensors are usually not used alone, mainly incorporated with flexible substrates or wound dressing, which can conformably cover the wound and without cause excessive stimulation to the wound site[14–16, 22, 32].

3.1. Flexible optical pH sensors for wound monitoring

One of the effective and simple methods for monitoring pH changing is using pH indicator dyes, which can absorb selected visible light wavelengths depending on the pH and then show naked eye color changing[27]. The challenges of this type pH sensor is that ensuring the tight adhesion of the dyes incorporated into the substrates such as wound dressings and choosing the proper dyes responding to the rightly pH range encountered in wounds[27, 33, 34].

According to this approach, Trupp *et al.* have developed an optical pH sensor with synthesized hydroxyl-substituted azobenzene derivatives as the pH indicator dyes, and cellulose films as the substrate[35, 36]. The pH sensitivity of the sensors results from the pendent vinylsulfonyl groups in the synthesized dyes. The reported sensor can monitor pH from 6–10. For actual use, the pH sensitive cellulose films can be laminated onto flexible polyethylene terephthalate films to protect the pH sensors from mechanical and swelling forces. Moreover, these laminates could be patterned into arrays as shown in Fig. 2(a), and exhibited good reversibility and sensitivity on pH. As the pH changes, a significant color change of the arrays can be observed from yellow (acidic) to red (basic), as suggested in Fig. 2(b). This may give a pH map to monitor a wound healing process. They also developed a compact wound dressing materials and plasters which indicate pathological changes in the skin. For a healthy skin (slightly acidic), the dressing shows yellow color (Fig. 2(c)), while an infected skin area (slightly basic) displays a purple color (Fig. 2(d))[37].

Similarly, Liu *et al.* have designed a pH sensitive hydrogel wound patch incorporated with the Phenol red (PR) dye modified with methacrylate (MA)[38]. The modified pH sensitive dyes which consist of benzene rings with hydroxyl groups that render pH-sensitivity are copolymerized with the alginate/polyacrylamide (PAAm) hydrogel matrix, which can prevent the dye from leaching. The designed pH-sensitive hydrogel patch shows several advantages such as a porous structure, high swelling ratio and excellent mechanical property. Obviously pH-responding color changing from yellow (pH 5, 6 and 7) to orange (7.4 and 8), and finally to red (pH 9) can be found, as shown in Fig. 2(e). This color change range rightly matching the pH range of chronic or infected wounds. Consequently, this pH sensor has potential application on monitoring the wound healing process[38].

To achieve the higher sensitivity and more effective composite wound dressing, pH sensitive electrospun nanofiberous films were developed[39–45]. By using solution electrospinning method, the sensitive dyes can be easily added into the precursor and then electrospun into fibers with the dyes wrapped in the polymer fibers[46–52]. Thanks to the large surface-area-to-volume ratio and high porosity of nanofibrous webs, the electrospun fibrous films were believed to be an ideal wound dressing both for physically protecting wound sites from external bacterial, and providing an ideal repairing environment allowing gaseous exchange[53–56]. Moreover, the large surface area to volume ratio of the as-spun nanofibers can provide an increasing number of sites for analyte interaction or signal transduction, and then improve the sensor sensitivity[39, 46, 47].

More recently, an electrospun film is developed for monitoring the wound pH by loading pH-sensitive color changing curcumin into the polycaprolactone (PCL) matrix[49]. The curcumin contains keto–enol tautomerism, and the keto form is predominant in an acidic environment, while the enol form is stable in an alkaline medium[49, 57]. Due to the keto–enol tau-

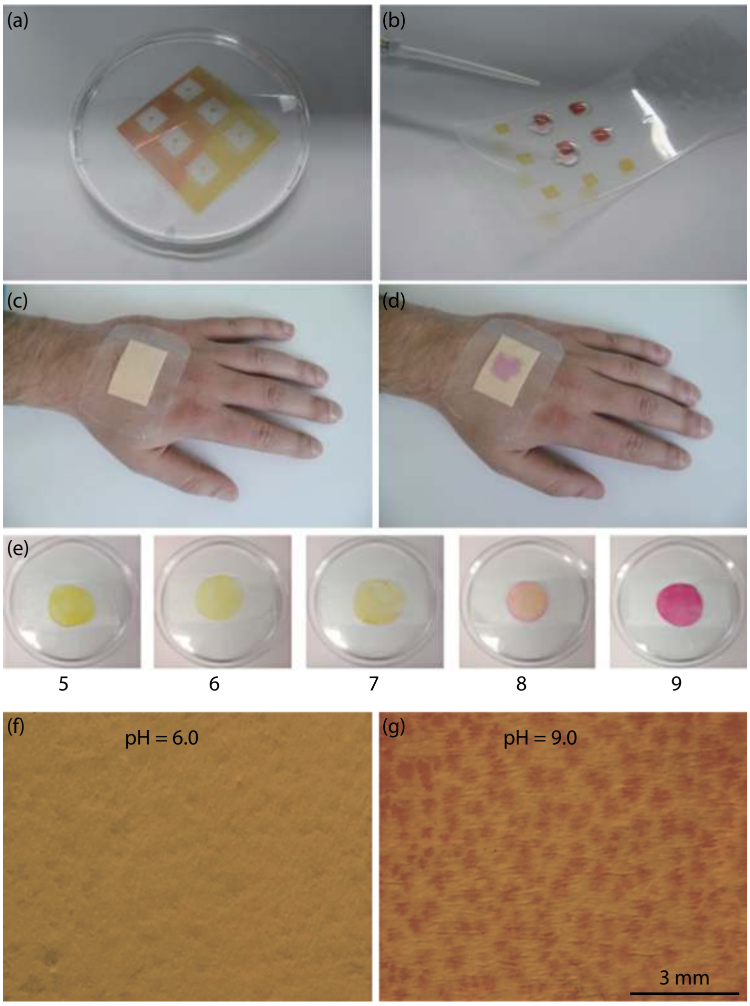

Fig. 2. (Color online) (a) Flexible array-type pH sensor. (b) Color change on the pH from acidic to basic[36]. (c) The flexible pH sensor can be fixed on the skin, and (d) change color when the pH varied[37]. (e) Photographic images of the hydrogel patch under pH values from 5 to 9. (f) and (g) Electrospun PCL-curcumin nanofibers under pH 6 and 9[49].

tomerism, curcumin showed an obviously chemical structure change due to pH. It is indicate that the curcumin-loaded fibrous mat exhibits an obvious pH-dependent color change from yellow to red brown with a change in pH range of 6.0 to 9.0, which can be simply detected by the human naked eye (Figs. 2(f) and 2(g)). There are several advantages with this color-changing electrospun PCL/curcumin fibrous materials to detect the wound pH. Firstly, the color can be easily in site detected without removing the wound dressing, giving a more intuitive method for pH monitoring. Secondly, one can easily judge the wound conditions according to the color change even without medical training, which can ensure the possible point-of-care diagnosis at home. Moreover, the as-spun fibrous mesh can also be used as a composite wound dressing, which can integrate wound healing and pH detection together. Furthermore, the as-spun fibrous materials can be applied to irregular wounds by processing these flexible meshes into kinds of shapes from 1D to 3D[49]. It is expected that this kind of smart optical pH-sensitive electrospun nanofibrous wound dressing may give a convenient and comfortable new approach for wound management. However, the visual pH sensors still showed a significant disadvantage that they cannot quantify the wound pH value precisely, which may require more precisely sensitive dyes, especially for naked eye visual[29].

3.2. Wearable electrochemical pH sensors for wound monitoring

To develop more precise pH sensors, electrochemical techniques were employed to measure potential, current and so on for responding pH[29]. Glass-electrodes based pH sensors were developed firstly to detect pH by measuring the electrical potential[58]. Soon after, inorganic electrochemical pH sensors have been significant developed containing metals (e.g., Pd, Bi, Sb), metal oxides (e.g., TiO_2, ZnO), and others (e.g., Si_3N_4, InAs)[29]. Specific for wound pH monitoring, kinds of flexible pH sensitive polymer materials were also developed including conductive (e.g., polyaniline (PANI)), semiconducting (e.g., polythiophenes), and insulating (e.g., parylene) ones[29]. Based on these pH sensitive materials, kinds of flexible and wearable pH sensors were designed.

Wang *et al.* have developed a wearable electrochemical pH sensor based on bandages for monitoring the pH of a wound[59]. The pH sensitivity comes from the electropolymerized PANi conducting polymer, due to the transition between emeraldine salt to emeraldine base in PANI[60–65]. As can be seen from Fig. 3(a), the sensor is fabricated through the screen-printing methodology, with Ag/AgCl as the reference electrodes, carbon as the working electrodes and PANI as the pH po-

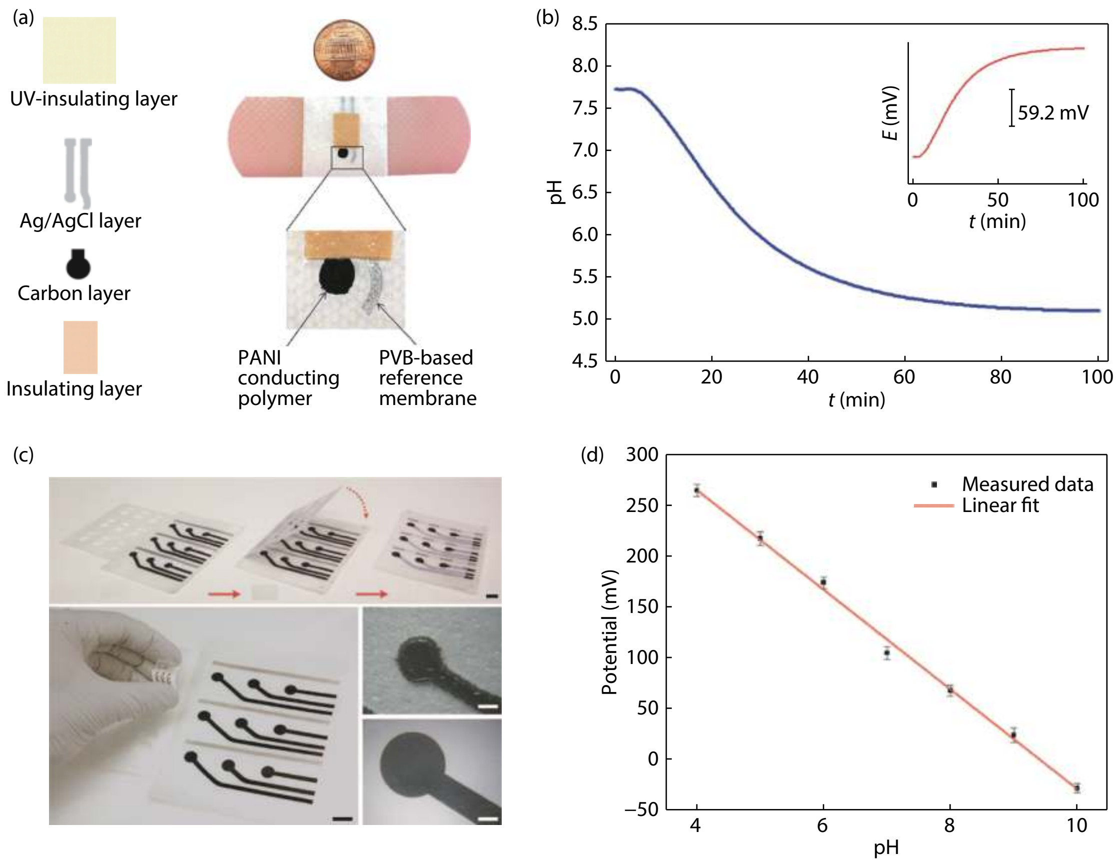

Fig. 3. (Color online) (a) Fabrication process to create the pH-sensitive bandage. (b) A real-time pH changes in a simulated wound[59]. (c) Fabrication process of paper based pH sensor array. (d) Relationship between the potential and pH values for the paper based pH sensor array[66].

tentiometric sensor. All these electrodes and the PANI sensors are screen-printed onto a commercial bandage. It is shown that the wearable pH bandage sensor shows a 59.2 mV/pH Nernstian response over a pH range 5.5–8, which is physiologically relevant to a wound (Fig. 3(b)). The sensor also shows several advantages including good mechanical stress elasticity, superior repeatability and reproducibility[59].

Similarly, Rahimi *et al.* designed an economical flexible pH array sensors based on a commercial paper (palettepaper) as illustrated in Fig. 3(c)[66]. The pH sensor also contains a screen-printed Ag/AgCl reference electrode and a carbon working screen-printed electrode coated, along with a pH sensitive PANI film. Moreover, a self-aligning passivation layer with maintenance holes is created by lamination technology. The pH sensor shows an average sensitivity of 50 mV/pH in the pH range 4–10, as suggested in Fig. 3(d). Moreover, the designed pH sensor showed good biocompatibility confirmed with human kertinocyte cells, which making it appropriate for effective functional low-cost wound dressings[66].

3.3. Wireless communication for flexible and wearable pH sensors

Generally, for the optical flexible pH sensors, the change of the pH in wound site could be examined by naked eye through the color change, which is one of the effective and simple methods to show wireless communication for monitoring wound pH changing.

However, for the electrochemical pH sensors mentioned above, the pH signal must be measured and readout by specialized instruments, which limited these sensors for personalized application in home. To make the pH sensors more convenient for use, wireless technologies were employed on the fabrication of pH sensors[32, 67].

Sridhar and Takahata have designed a hydrogel-based wireless pH sensor as shown in Fig. 4(a). It can be seen that this sensor has a sandwich structure with two aligned coils by folding a flexible conductive polyimide film forming a gap and a pH sensitive poly(vinyl alcohol)-poly(acrylic acid) (PVA-PAA) hydrogel in the middle to adjust the gap. Since the PVA-PAA hydrogel shows a pH relative volume/dimension change in different pH range from 2–7, the inductance of the coil could be changed, as well as the frequency changed along with the inductance. Additional with a designed wireless measurement device, an magnetically coupled external coil, the change of frequency depending on pH can be detected by a 4396B network-spectrum analyzer[24, 27, 68]. This opens a newly application methods for the pH sensors.

Following this way, Pal *et al.* have developed a low-cost, single-use, omniphobic paper-based smart bandages (OPSBs) to monitor the pH of open chronic wounds, as shown in Fig. 4(b)[69]. A repeatable wearable potentiostat was fabricated simply by attaching it onto the interface of the OPSB. To detect the wound pH, a Ag/AgCl electrodes is printed on omniphobic paper and separated by a layer of Ag/PANi composite. Due to the pH sensitivity of PANi, the accurate measurement of the wound exudate pH could be achieved by applying a low voltages (100 mV). It is found that there is no frequency dependence of the impedance up to 10 kHz across the electrodes of the pH sensor (Fig. 4(b)). The relationship between pH and impedance for this omniphobic, paper-

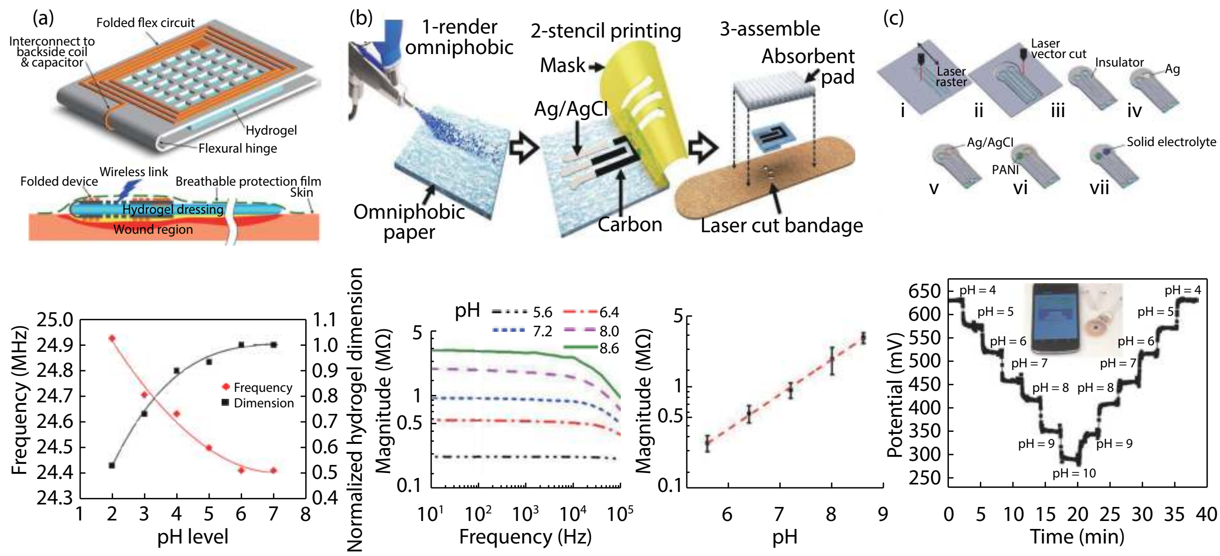

Fig. 4. (Color online) (a) Design of a pH sensitive gel placed between two inductive coils. Relationship between the resonant frequency, the gap and the pH[68]. (b) Schematic diagram of the fabrication of OPSBs and the magnitude relation to pH[69]. (c) Fabrication process of pH sensor on ITO film and potential related to pH[70].

based sensors can be found in Fig. 4(b). According to this relationship, the sensor can be used to precisely quantify pH levels in the wound. Based on these results, the the OPSB integrated with wearable potentiostat can be applied to simultaneously quantify pH at the wound site in the range of 5.5–8.5, and wirelessly report wound status to the user or medical personnel[69].

Furthermore, Rahimi *et al.* have presented a low-cost smartphone based pH sensing platform that consists of a flexible, disposable, and transparent pH sensor interfaced with a custom-designed flexible, battery-less, and reusable near-field communication (NFC) tag[70]. As shown in Fig. 4(c), there are two major parts in the composite sensors: a flexible disposable and transparent pH sensor and a flexible wireless NFC interface circuit. The pH sensor is mainly based on an open circuit with an Ag/AgCl reference electrode and a pH-sensitive PANI working electrode onto an ITO film. The wireless low-battery operated NFC interface allows wireless communication with either a smartphone (with NFC communication function) or PC-based NFC reader (AS3911) by bringing the reader in the range of < 4 cm. It is found that the sensors have a high sensitivity of 55 mV/pH within the physiologically relevant pH range of 4–10 and stable performance even under bending (Fig. 4(c)). Moreover, the wireless pH sensors can be easily integrated into any standard wound dressings and found to accurately detect pH changes in a gram-positive Staphylococcus epidermidis infected simulated in vitro wound conditions. For convenience, a designed Android app can provide a simple user interface to read the pH in the wound bed[70], which make the designed pH sensor system showing potential applications in the wound monitoring in home.

To take both the advantages of the optical and electrochemical pH sensors, kinds of sensors integrated with custom CMOS readout electronics for wireless monitoring wound pH and data transmission has been developed[71–74].

As shown in Fig. 5, Punjiya *et al.* have designed a wireless pH sensing bandage with numerous pH sensing smart threads for chronic wound monitoring[72]. The pH sensing threads are prepared using commercial cotton thread as a substrate. Firstly, the threads are immersed in isopropanol, and then insoaked, pulled through a carbon ink to make the threads conductive. Subsequently, the conductive threads are coated several dips in a PANI ink to make the threads pH sensitive and finally dry in a desiccator for later use. Similarly, a reference Ag/AgCl thread is also prepared by coating thread in Ag/AgCl ink. To detect the pH chronic wounds conveniently, all of the functionalized threads are seamlessly sewn into a commercial bandage, as shown in Fig. 5(a). The threads exhibit a high pH sensitivity of 54 mV/pH as suggested in Fig. 5(b). Moreover, a custom CMOS potentiostat readout IC incorporated with an Arduino Nano and commercial Bluetooth module are used for the wireless data acquisition, as displayed in Fig. 5(c). The open circuit voltage can be acquired from all the threads sensors and then mapped to responding the different pH values in the corresponding location area on the bandage (Fig. 5(d)). The same group also reported a new flexible pH sensor with a creation of high surface area (HSA) sensing electrodes via a three-dimensional sewn polyaniline (PANI) structure to achieve a more accurate result for wound pH monitoring[73].

4. Conclusion and future challenges

In conclusion, pH sensors for monitoring the wound healing have gained much attention in recent years. These sensors based on several pH sensitive mechanisms such as optical and electrochemical ones by providing different signals. Moreover, the reported pH sensors mostly based on flexible substrate such as polymer films, textiles and bandages to ensure their flexible and wearable properties. Furthermore, wireless transmission is also a popular trend for the development of these pH sensors, even can use a smart phone to collect the wound pH values and then determine the wound healing situation. While, there still some challenges for the pH sensors at the present stage.

(1) Development of large scale pH sensors for monitoring the large-area chronic wounds such as burn injury and diabetic foot wound. To produce large scale pH sensors, electro-

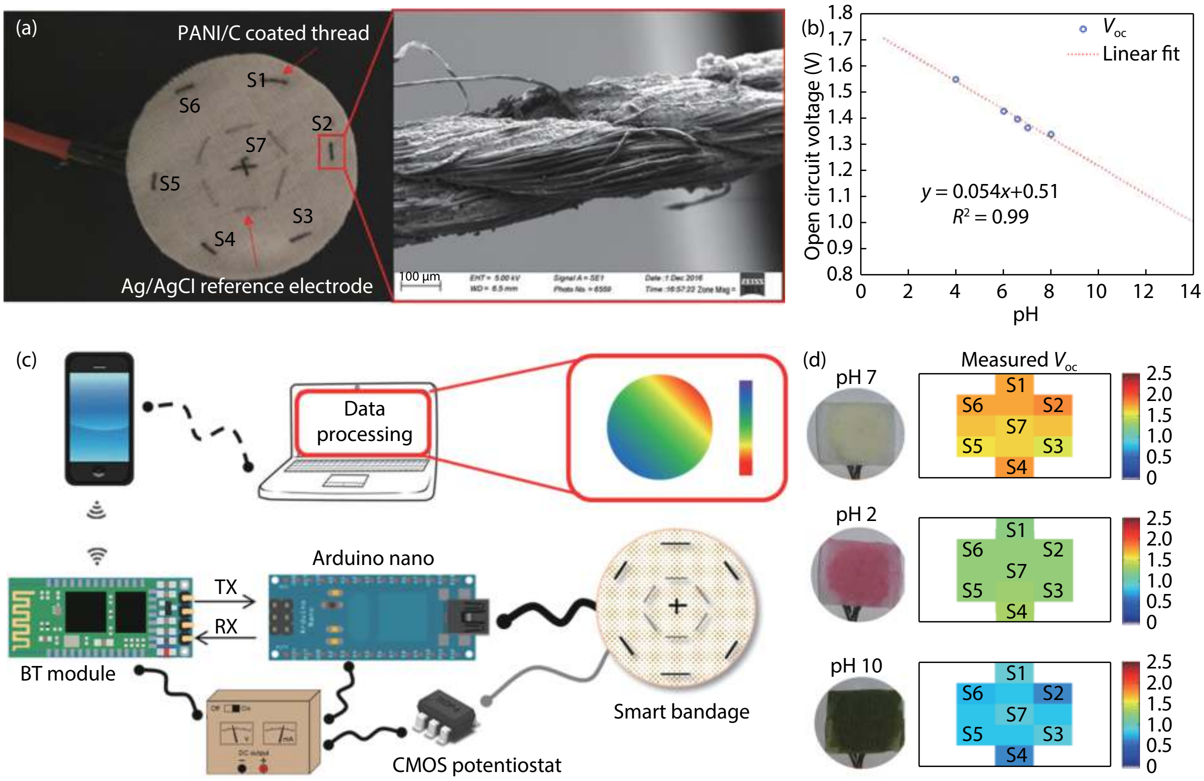

Fig. 5. (Color online) (a) Optical image of pH sensing bandage (left) and SEM of the pH sensitive threads. (b) Open circuit voltage versus pH for the threads. (c) Data acquisition setup for pH sensing smart bandage. (d) Smart bandage pH map[72].

spinning may be a possible method to achieve it.

(2) Development of *in situ* fabrication of pH sensors incorporated with composite wound dressing directly onto the wound site. As the development of the *in situ* electrospinning technology through portable electrospinning devices and elctrospun pH nanofiber sensors, the composite nanofibrous wound dressing will be a new approach for wound healing.

(3) Development more precise optical pH sensors with naked eye color change. The color change should be more clear to be distinguished by the naked eye.

(4) pH sensors is a critical concern for wound monitoring, especially for directly contacting the wound. Researcher should pay attention on this point and develop the pH sensors using biocompatible materials.

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References

[1] Yildirimer L, Thanh N T K, Seifalian A M. Skin regeneration scaffolds a multimodal bottom-up approach. Trends Biotechnol, 2012, 30, 638

[2] Dias J R, Granja P L, Bártolo P J. Advances in electrospun skin substitutes. Prog Mater Sci, 2016, 84, 314

[3] Davidson J M, DiPietro L. The wound-healing process. 2nd ed. Totowa: Humana Press, 2006

[4] Mehmood N, Hariz A, Fitridge R, et al. Applications of modern sensors and wireless technology in effective wound management. J Biomed Mater Res Part B, 2014, 102B, 885

[5] Gurtner G C, Werner S, Barrandon Y, et al. Wound repair and regeneration. Nature, 2008, 453, 314

[6] Tonnesen M G, Feng X, Clark R A. Angiogenesis in wound healing. J Invest Dermatol Symp Proc, 2000, 5, 40

[7] Menke N B, Ward K R, Witten T M, et al. Impaired wound healing. Clin Dermatol, 2007, 25, 19

[8] Abdelrahman T, Newton H. Wound dressings: Principles and practice. Surgery (Oxford), 2011, 29, 491

[9] Mao N, Russell S J. Nonwoven wound dressings. Textil Progr, 2004, 36, 4, 1

[10] Thomas S. Wound and wound healing, wound management and dressings. London: Pharmaceutical Press, 2004

[11] Bandodkar A J, Wang J. Non-invasive wearable electrochemical sensors: a review. Trends Biotechnol, 2014, 32, 363

[12] Wang S, Chinnasamy T, Lifson M A, et al. Flexible substrate-based devices for point-of-care diagnostics. Trends Biotechnol, 2016, 34, 909

[13] Gianino E, Miller C, Gilmore J. Smart wound dressings for diabetic chronic wounds. Bioengineering, 2018, 5(3), 51

[14] Brown M S, Ashley B, Koh A. Wearable technology for chronic wound monitoring: current dressings, advancements, and future prospects. Front Bioeng Biotechnol, 2018, 6, 47

[15] Ochoa M, Rahimi R, Ziaie B. Flexible sensors for chronic wound management. IEEE Rev Biomed Eng, 2014, 7, 73

[16] Wang X, Liu Z, Zhang T. Flexible sensing electronics for wearable/attachable health monitoring. Small, 2017, 13, 1602790

[17] Sun Q, Qian B, Uto K, et al. Functional biomaterials towards flexible electronics and sensors. Biosens Bioelectron, 2018, 119, 237

[18] Schneider L A, Grabbe A K S, Dissemond J. Influence of pH on wound-healing: a new perspective for wound-therapy. Arch Dermatol Res, 2007, 298, 413

[19] Gethin G. The significance of surface pH in chronic wounds. Wounds UK, 2007, 3, 3

[20] Shukla V K, Shukla D, Tiwary S K, et al. Evaluation of pH measure-

ment as a method of wound assessment. J Wound Care, 2007, 16, 7
[21] Schreml S, Szeimies R M, Karrer S, et al. The impact of the pH value on skin integrity and cutaneous wound healing. J Eur Acad Dermatol, 2010, 24, 373
[22] Percival S, McCarty S, Hunt J, et al. The effects of pH on wound healing, biofilms, and antimicrobial efficacy. Wound Repair Regen, 2014, 22(2), 174
[23] Lei S, Sarah R, Ryan E, et al. pH in the bacteria-contaminated wound and its impact on clostridium histolyticum collagenase activity: implications for the use of collagenase wound debridement agents. J Wound Ostomy Cont, 2011, 38, 514
[24] Khan M A, U Ansari U, Ali M N. Real-time wound management through integrated pH sensors: a review. Sens Rev, 2015, 35, 183
[25] Schaude C, Fröhlich E, Meindl C, et al. The development of indicator cotton swabs for the detection of pH in wounds. Sensors, 2017, 17, 1365
[26] Al-Hillia S M, Willander M, Öst A, et al. ZnO nanorods as an intracellular sensor for pH measurements. J Appl Phys, 2007, 102, 084304
[27] Dargaville T R, Farrugia B L, Broadbent J A, et al. Sensors and imaging for wound healing: a review. Biosens Bioelectron, 2013, 41, 30
[28] Solomon E, Schmidt R, Adragna P. Human anatomy and physiology. 2nd ed. USA: Saunders, 1990
[29] Alam A U, Qin Y, Nambiar S, et al. Polymers and organic materials-based pH sensors for healthcare applications. Prog Mater Sci, 2018, 96, 174
[30] Wencel D, Abel T, McDonagh C. Optical chemical pH sensors. Anal Chem, 2014, 86, 15
[31] Miao Y, Chen J, Fang K. New technology for the detection of Ph. J Biochem Biophy. Methods, 2005, 63, 1
[32] Finlay D. Connected health approaches to wound monitoring. In: Smart Bandage Technologies. Academic Press, 2016, 229
[33] Der Schueren L V, Clerck K D. Coloration and application of pH-sensitive dyes on textile materials. Color Technol, 2012, 128, 82
[34] Der Schueren L V, Clerck K D. The use of pH-indicator dyes for pH-sensitive textile. Text Res J, 2010, 80, 590
[35] Trupp S, Alberti M, Carofiglio T, et al. Development of pH-sensitive indicator dyes for the preparation of micro-patterned optical sensor layers. Sens Actuators B, 2010, 150, 206
[36] Mohr G J, Müller H, Bussemer B, et al. Design of acidochromic dyes for facile preparation of pH sensor layers. Anal Bioanal Chem, 2008, 392, 1411
[37] https://www.fraunhofer.de/en/press/research-news/2010/11/dressing-indicates-infections.html
[38] Liu L, Li X, Nagao M, et al. A pH-Indicating colorimetric tough hydrogel patch towards applications in a substrate for smart wound dressings. Polymers, 2017, 9, 558
[39] der Schueren L V, T Mollet T, Ceylan Ö, et al. The development of polyamide 6.6 nanofibres with a pH-sensitive function by electrospinning. Eur Polym J, 2010, 46, 2229
[40] Zhang C, Li Y, Wang W, et al. A novel two-nozzle electrospinning process for preparing microfiber reinforced pH-sensitive nanomembrane with enhanced mechanical property. Eur Polym J, 2011, 47, 2228
[41] Diculescu V C, Beregoi M, Evanghelidis A, et al. Palladium/palladium oxide coated electrospun fibers for wearable sweat pH-sensors. Sci Rep, 2019, 9, 8902
[42] da Silva C K, da S Mastrantonio D J, Costa J A V, et al. Innovative pH sensors developed from ultrafine fibers containing açaí (Euterpe oleracea) extract. Food Chem, 2019, 294, 397
[43] Gorji M, Sadeghianmaryan A, Rajabinejad H, et al. Development of highly pH-sensitive hybrid membranes by simultaneous electrospinning of amphiphilic nanofibers reinforced with graphene oxide. J Funct Biomater, 2019, 10(2), 23
[44] Agarwal A, Raheja A, Natarajan T S, et al. Development of universal pH sensing electrospun nanofibers. Sens Actuators B, 2012, 161, 1097
[45] Devarayan K, Kim B S. Reversible and universal pH sensing cellulose nanofibers for health monitor. Sens Actuators B, 2015, 209, 281
[46] Schoolaert E, Hoogenboom R, Clerck K D. Colorimetric nanofibers as optical sensors. Adv Funct Mater, 2017, 27, 1702646
[47] Steyaert I, Rahier H, Clerck K D. Nanofibre-based sensors for visual and optical monitoring. In: Electrospinning for High Performance Sensors. NanoScience and Technology. Springer, 2015
[48] Schoolaert E, Steyaert I, Vancoillie G, et al. Blend electrospinning of dye-functionalized chitosan and poly(ε-caprolactone): towards biocompatible pH-sensors. J Mater Chem B, 2016, 4, 4507
[49] Pan N, Qin J, Feng P, et al. Color-changing smart fibrous materials for naked eye real-time monitoring of wound pH. J Mater Chem B, 2019, 7, 2626
[50] Yan X, You M H, Lou T, et al. Colorful hydrophobic poly(vinyl butyral)/cationic dye fibrous membranes via a colored solution electrospinning process. Nanoscale Res Lett, 2016, 11, 540
[51] You M, Yan X, Zhang J, et al. Colorimetric humidity sensors based on electrospun polyamide/$CoCl_2$ nanofibrous membranes. Nanoscale Res Lett, 2017, 12, 360
[52] Liu M N, Yan X, You M H, et al. Reversible photochromic nanofibrous membranes with excellent water/windproof and breathable performance. J Appl Polym Sci, 2018, 135(23), 46342
[53] Li C, Lou T, Yan X, et al. Fabrication of pure chitosan nanofibrous membranes as effective absorbent for dye removal. Int J Biol Macromol, 2018, 106, 768
[54] Liu G S, Yan X, Yan F F, et al. In situ electrospinning iodine-based fibrous meshes for antibacterial wound dressing. Nanoscale Res Lett, 2018, 13, 309
[55] Chen S, Liu G S, He H W, et al. Physical structure induced hydrophobicity analyzed from electrospinning and coating polyvinyl butyral films. Adv Cond Matter Phys, 2019, 2019, 6179456
[56] Yan X, Yu M, Ramakrishna S, et al. Advances in portable electrospinning devices for in-situ delivery of personalized wound care. Nanoscale, 2019
[57] Priyadarsini K I. Photophysics, photochemistry and photobiology of curcumin: Studies from organic solutions, bio-mimetics and living cells. J Photochem Photobiol C, 2009, 10, 81
[58] Graham D J, Jaselskis B, Moore C E. Development of the glass electrode and the pH response. J Chem Educ, 2013, 90(3), 345
[59] Guinovart T, Ramírez G V, Windmiller J R, et al. Bandage-based wearable potentiometric sensor for monitoring wound pH. Electroanalysis, 2014, 26, 1345
[60] Huang W S, Humphrey B D, MacDiarmid A G. Polyaniline, a novel conducting polymer. Morphology and chemistry of its oxidation and reduction in aqueous electrolytes. J Chem Soc Faraday Trans, 1986, 82, 2385
[61] Quillard S, Berrada K, Louarn G, et al. In situ Raman spectroscopic studies of the electrochemical behavior of polyaniline. New J Chem, 1995, 19, 365
[62] Lindfors T, Ivaska A. pH sensitivity of polyaniline and its substituted derivatives. J Electroanal Chem, 2002, 531, 43
[63] Kaempgen M, Roth S. Transparent and flexible carbon nanotube/polyaniline pH sensors. J Electroanal Chem, 2006, 586, 72
[64] Lindfors T, Ervela S, Ivaska A. Polyaniline as pH-sensitive component in plasticized PVC membranes. J Electroanal Chem, 2003, 560, 69
[65] Zhang X, Ogorevc B, Wang J. Solid-state pH nanoelectrode based on polyaniline thin film electrodeposited onto ion-beam etched carbon fiber. Anal Chim Acta, 2002, 452, 1
[66] Rahimi R, Ochoa M, Parupudi T, et al. A low-cost flexible pH sensor array for wound assessment. Sens Actuators B, 2016, 229,

609

[67] Loh K J, Lynch J P, Kotov N A. Inductively coupled nanocomposite wireless strain and pH sensors. Smart Struct Syst, 2008, 4, 531

[68] Sridhar V, Takahata K. A hydrogel-based passive wireless sensor using a flex-circuit inductive transducer. Sens Actuators A, 2009, 155, 58

[69] Pal A, Goswami D, Cuellar H E, et al. Early detection and monitoring of chronic wounds using low-cost, omniphobic paper-based smart bandages. Biosens Bioelectron, 2018, 117, 696

[70] Rahimi R, Brener U, Chittiboyina S, et al. Laser-enabled fabrication of flexible and transparent pH sensor with near-field communication for in-situ monitoring of wound infection. Sens Actuators B, 2018, 267, 198

[71] Hizawa T, Sawada K, Takao H, et al. Fabrication of a two-dimensional pH image sensor using a charge transfer technique. Sens Actuators B, 2006, 117, 509

[72] Punjiya M, Rezaei H, Zeeshan M A, et al. A flexible pH sensing smart bandage with wireless cmos readout for chronic wound monitoring. 19th International Conference on Solid-State Sensors Actuators and Microsystems (TRANSDUCERS), 2017, 1700

[73] Lyu B, Punjiya M, Matharu Z, et al. An improved pH mapping bandage with thread-based sensors for chronic wound monitoring. IEEE International Symposium on Circuits and Systems (ISCAS), 2018, 1

[74] Christodouleas D C, Kaur B, Chorti P. From point-of-care testing to ehealth diagnostic devices (eDiagnostics). ACS Cent Sci, 2018, 4, 1600