Fan *et al*. *Soft Sci* 2024;4:11
DOI: 10.20517/ss.2023.47

Soft Science

Perspective

Open Access

From natural leather to intelligent wearable nanocomposite: design and application

Ziyang Fan[1], **Min Sang**[1], **Xinglong Gong**[1,2], **Ken Cham-Fai Leung**[3,*], **Shouhu Xuan**[1,2,*]

[1]CAS Key Laboratory of Mechanical Behavior and Design of Materials, Department of Modern Mechanics, University of Science and Technology of China (USTC), Hefei 230027, Anhui, China.
[2]State Key Laboratory of Fire Science, University of Science and Technology of China, Hefei 230026, Anhui, China.
[3]State Key Laboratory of Environmental and Biological Analysis, Department of Chemistry, The Hong Kong Baptist University, Kowloon 999077, Hong Kong, China.

[*]**Correspondence to:** Prof. Shouhu Xuan, CAS Key Laboratory of Mechanical Behavior and Design of Materials, Department of Modern Mechanics, University of Science and Technology of China (USTC), 443 Huangshan Road, Shushan District, Hefei 230027, China. E-mail: xuansh@ustc.edu.cn; Prof. Ken Cham-Fai Leung, State Key Laboratory of Environmental and Biological Analysis, Department of Chemistry, The Hong Kong Baptist University, 224 Waterloo Road, Kowloon 999077, Hong Kong, China. E-mail: cfleung@hkbu.edu.hk

How to cite this article: Fan Z, Sang M, Gong X, Leung KCF, Xuan S. From natural leather to intelligent wearable nanocomposite: design and application. *Soft Sci* 2024;4:11. https://dx.doi.org/10.20517/ss.2023.47

Received: 22 Oct 2023 **First Decision:** 21 Nov 2023 **Revised:** 4 Dec 2023 **Accepted:** 27 Dec 2023 **Published:** 20 Feb 2024

Academic Editors: Zhifeng Ren, Martinez Ramses **Copy Editor:** Pei-Yun Wang **Production Editor:** Pei-Yun Wang

Abstract

As a natural material, leather has been widely used in daily life due to its high biocompatibility, wearing comfort, and excellent mechanical strength. However, with the increasing demand for a better life among people, the single function of leather has difficulty in meeting the requirements, which limits its application prospects. It is particularly important to develop multifunctional leather composites with diverse characteristics. Therefore, leather can be modified and functionally designed through physical and chemical methods towards intelligent wearable devices. From this perspective, we review the research progress of intelligent leather-based wearable composites, mainly focusing on the preparation methods and application directions in recent years. Finally, we emphasize the challenges that leather composites will face in practical applications and propose future research directions.

Keywords: Leather, flexible electronics, soft sensors, safeguarding, thermal management

INTRODUCTION

Leather, as an ancient natural polymer, has a simple source and preparation, abundant reserves, high biocompatibility, good breathability, strong stability, wear resistance, biodegradability, *etc.*[1-3]. The preparation process for the leather preserves the flexibility and strength of animal skin; thus, it has been used as a popular clothing and armor material throughout history[4,5]. In the early days, people's demand for leather products was practical, beautiful, sturdy, and stab-resistant. Compared to traditional weaving fabrics, the natural 3D weaving structure and intertwined fiber bundles of leather can better reduce acupuncture damage. However, with the rapid development of soft functional composites and flexible electronic devices, the single function and application of leather composites cannot satisfy the requirement[6,7]. In comparison to flexible substrates with a single structure, such as polydimethylsiloxane (PDMS), polyethylene terephthalate (PET), *etc*., the leather is more easily modified and highly breathable due to the natural porous structures[8]. Furthermore, the mechanical strength of leather is reliable compared to other fiber-based substrates, such as cotton-based textiles and polyimide (PI)[9]. Therefore, leather will be expected to have more potential applications in wearable electronic devices, electromagnetic interference shielding, flame retardant protection, and intelligent thermal management[10].

When processed into leather, the original skin of animals requires many steps, such as removing impurities, tanning, fat-liquoring, and drying[11,12]. After a series of treatments, the nanoscale collagen fibers, unique 3D porous network structure, and multilevel hierarchical structure were ultimately retained[13]. Based on this special microstructure and biocompatible collagen fibers, a large amount of research has emerged on multifunctional leather composites in recent years[14]. On the one hand, various functional materials were selected to combine with collagen fibers to form physical adhesion or chemical crosslinking bonds[3,15], which were composed of conductive materials including graphene oxides (GO)[16,17], carbon nanotubes (CNTs)[18,19], silver nanowires (AgNWs)[20], MXene[21], poly(3,4-ethylenedioxythiophene)-poly(styrene sulfonate) (PEDOT:PSS)[22], *etc*., and insulating materials include silicon dioxide particles[23], boron nitride[24], montmorillonite[25], *etc*. Then, the structural design was carried out on the leather at the macro- or micro-scale by utilizing the above functional materials to obtain leather composites with editable properties[26]. These methods are similar to dyeing in the leather-making process. On the other hand, the natural microstructure of leather fiber networks could be used for *in-situ* growing hydrogels or other composites through the induction of catalytic materials. At last, the obtained hydrogels or other composites could be further designed by the above functional materials, which enabled leather composites to get other functions[27,28].

By combining various functional materials, the smart leather with tunable functionalities, such as conductive leather[29,30], electromagnetic shielding leather[31], flame retardant leather[32,33], antibacterial leather[34-36], thermal camouflage leather[37], waterproof leather[11,38], and so on, has been developed. From this perspective, we mainly introduce several common and efficient preparation methods for constructing intelligent leather composites and then focus on their applications in flexible sensors, electromagnetic interference shielding, flame retardant, body safeguarding, thermoregulatory clothing, and other aspects. Finally, the potential difficulties and future development trends of leather composites in practical applications are discussed. This discussion will help intelligent leather composites enter people's daily lives and promote the industrialization development stage of functional leather[39].

DESIGN AND PREPARATION OF LEATHER COMPOSITES

Currently, there have been many studies on the preparation of leather composites. In this section, five common and effective methods are introduced in detail, including vacuum-assisted filtration, spraying, laser direct writing (LDW), *in-situ* growth, and multilayer assembly.

Vacuum-assisted filtration

The method of vacuum-assisted filtration utilizes a vacuum environment to provide a pressure difference between the upper and lower sides of leather, which drives functional materials to enter the interior of the leather and combine with collagen fibers. Then, the samples were dried to obtain leather composites. Due to its unique layered structure, there are significant differences between the two sides of leather. The grain side is composed of collagen fibers and elastic fibers, and the fiber bundles are finer and more tightly woven, presenting an uneven shape[40]. The fiber side is composed of interwoven bundles of collagen fibers of different thicknesses, forming a three-dimensional network, and its tightness is positively correlated with the mechanical properties of leather. Ma *et al.* utilized this characteristic to put the fiber side of the leather upwards, and AgNWs were dissolved and infiltrated into the porous structure of the leather through vacuum-assisted filtration [Figure 1A][41]. Strong interactions between AgNWs and collagen fiber bundles could be generated through hydrogen bonding, forming an efficient three-dimensional conductive network in leather. The pores of leather range from tens to hundreds of nanometers, so nanoscale functional materials, such as MXene, CNTs, GO, poly(3,4-ethylenedioxythiophene) nanofibers (PEDOT NFs), and ionic liquids, within this scale can be used to prepare leather composites by vacuum-assisted filtration[42].

Spraying and soaking

The methods of spraying and soaking are attaching functional materials to leather through physical processes, relying solely on the binding ability of functional materials and leather collagen fibers, which is relatively simple and convenient in terms of process. In general, some polymer solutions, non-metal, and metal particles are used to spray onto the leather substrate to form functional coatings, which can strengthen the interfacial bonding between the material and leather due to soaking during the spraying process[24,43]. Wilson *et al.* used bimetallic copper-iron oxide nanoparticles to spray on the surface of the leather as an electromagnetic coating[15]. The formed electrically conductive and magnetically active bifunctional leather demonstrated the application possibilities in operating intelligent screens and magnetic switches. Li *et al.* soaked La_2O_3 and Bi_2O_3 nanoparticles in sheep leather and sprayed them on the upper and lower surfaces of leather as coatings, greatly increasing the particle load and enhancing its X-ray protection performance[44]. Mo *et al.* also combined spraying and vacuum-assisted filtration to filter acidified CNTs on the fiber side and sprayed porous cellulose acetate on the corium side to obtain multifunctional double-layer leather composites, greatly utilizing the layered structure of leather [Figure 1Ba][45]. It is obvious that one side of the leather becomes black after filling acidified multiwalled carbon nanotubes (a-MWCNTs), and the other side turns white after spraying porous cellulose acetate. The resulting double-layer leather composites maintain good mechanical performance and breathability [Figure 1Bb-f].

Laser direct writing

In recent years, LDW has gradually become a high-precision and efficient processing technology[46,47]. Many fabric-based flexible electronics were developed by the LDW technique, and the various carbon precursors were converted into graphene during the laser scanning in the textile. Based on maskless, design flexibility, and pattern editable characteristics of LDW, Yang *et al.* used a femtosecond laser on Kevlar fabric to induce graphene for various electronic textile applications[48]. It is simple to construct wearable sensors in various textile structures by LDW. Leather, as an emerging biomaterial, contains a large amount of carbon elements. Various flexible electronic devices also can be manufactured without the need for other functional materials by combining computer control with micro-processing technology. Local high temperatures are generated by laser irradiation on the leather substrate to achieve carbonization. Wang *et al.* used the LDW method to induce the carbonization of collagen fiber on the surface of the leather for fabricating wearable sensors. The high control accuracy could directly characterize complex structures, such as arrays, on the surface [Figure 1Ca-d][49]. In this case, the collagen fibers transformed from insulation to conductive materials after carbonization and could directly serve as strain sensors to detect tensile and compressive strains. Zhang

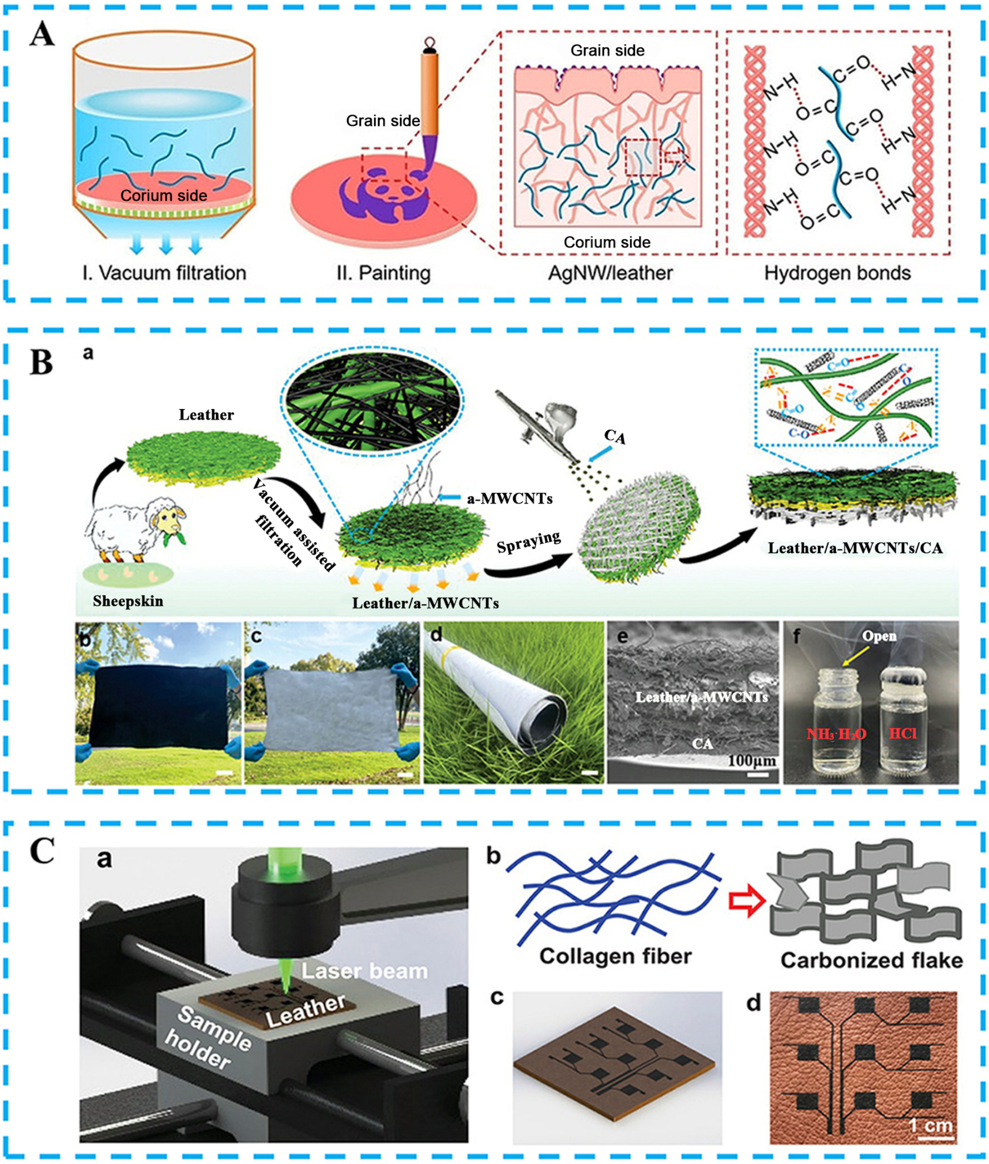

Figure 1. The preparation methods of leather composites. (A) Schematic illustration for fabricating AgNW/leather nanocomposites by vacuum-assisted filtration[41]. Reprinted with permission. Copyright 2022, Wiley-VCH; (Ba) Schematic diagram of the fabrication of leather/a-MWCNT/CA fabric by spraying and vacuum-assisted filtration; (Bb-f) Photographs of the color, bendability, cross-sectional characterization, and breathability of leather/a-MWCNT/CA fabric[45]. Reprinted with permission. Copyright 2023, Wiley-VCH; (Ca) A schematic shows the sensor fabrication on the leather by laser direct writing and (Cb) another schematic shows the conversion of collagen fibers in the leather to carbon flakes by the LDW; (Cc and d) Schematic and photograph of a 3 × 3 sensor array[49]. Reprinted with permission. Copyright 2020, Wiley-VCH. AgNW: Silver nanowire; a-MWCNT: acidified multiwalled carbon nanotubes; LDW: laser direct writing.

et al. prepared the cross-sensing arrays on the leather substrate with the assistance of LDW technology[21]. The final product exhibits effective control for mechanical hand movements and human-machine interaction switches, which indicates the high convenience and application potential of the LDW method.

In-situ polymerization

Originally, *in-situ* polymerization was the filling of reactive monomers into the interlayer of nanolayered materials, allowing them to undergo polymerization reactions between the layers. The natural hierarchical 3D network structure of leather provides an interpenetrating fiber network for the *in-situ* growth of polymers[50]. By configuring different precursor solutions, they can easily flow in the porous structure of the leather and conduct gelation[28]. Fan *et al.* used natural goat skin as the substrate and dipped the acrylic monomer, Zr^{4+} ions, carbon quantum dots@nanosilver particles (CQDs@AgNPs), and 1,3-propanediol (PDO) into the fiber skeleton of leather. Then, the AA monomer was *in-situ* polymerized to obtain a multifunctional hydrogel containing the leather skeleton [Figure 2Aa][27]. The 3D network of leather in ionic gel became an effective flow channel for loading CQDs@AgNPs and PDO, which endowed the hydrogels with excellent mechanical properties, self-adhesiveness, transparency, UV shielding, antibacterial, biocompatibility, and conductivity. Furthermore, the $Zr(SO_4)_2$ could form a strong interfacial bonding with collagen fibers to enhance the network structure; therefore, the mechanical properties of leather composites were strengthened [Figure 2Ab]. This method provides effective design ideas for the development of intelligent, flexible electronic skins.

Multilayer assembly

Sandwich structures can enhance the overall mechanical properties of composites through the synergistic coordination of different components[51,52]. Fan *et al.* proposed a flexibility-toughness coupling design strategy to develop intelligent anti-impact leather. By assembling flexible shear stiffening gel (SSG), tough leather, and nonwoven fabric (NWF) into a Leather/SSG/NWF sandwich structure, the mechanical properties of the resulting leather composite were greatly improved [Figure 2B][53]. At the same time, the leather layer could also be designed with special functions. For example, the MXene nanosheets could be combined with leather fibers through vacuum-assisted filtration, and then the wearable Leather/MXene/SSG/NWF safeguarding leather composite with excellent sensing, thermal management, and electromagnetic interference shielding was obtained. Obviously, this idea can be further expanded for the multifunctional design and application of intelligent leather [Figure 2C][54].

MULTIFUNCTIONAL APPLICATIONS OF LEATHER COMPOSITES

By combining various functional materials with leather, a variety of leather composites, including conductive leather, electromagnetic shielding leather, flame retardant leather, thermal management leather, *etc.*, have been successfully developed[6,26,55]. Then, intelligent leather composites are further obtained by structural design and assembly based on single-function leather, which can be widely used for flexible sensors, electromagnetic shielding devices, safety protection, flame retardant, intelligent displays, and intelligent thermal management. As shown in Table 1, the preparation methods and functionalities of different leather composites based on various materials are summarized to understand intuitively. Obviously, intelligent leather composites provide an important research direction for the development of wearable electronic devices.

Flexible sensors

Flexible sensors have broad application prospects in human motion monitoring, human-machine interactions, and the intelligent wearable field[56,57]. Natural leather materials have a hierarchical structure and elemental composition similar to human skin; thus, they can be used as an excellent substrate material for flexible sensors. To date, various leather composites have been widely used in flexible sensors, which can be divided into different working mechanisms, including piezoresistive sensing[13,16], strain sensing[58,59], triboelectricity[17,21], and so on. Ma *et al.* prepared AgNW/leather composites and assembled them with interdigitated copper electrodes to form a piezoresistive sensor [Figure 3Aa][41]. The piezoresistive sensor showed different sensitivities in three distinct pressure stages, indicating excellent piezoresistive sensing ability. When the pressure is lower than 2.5 kPa (stage I), the piezoresistive sensor shows a low sensitivity due to the slight compression deformation of AgNW/leather composites. As the pressure increases to 10 kPa (stage II), the collagen fiber bundles undergo densification, resulting in a more efficient conductive network under larger compression deformation, so the piezoresistive sensor shows a high sensitivity. If the

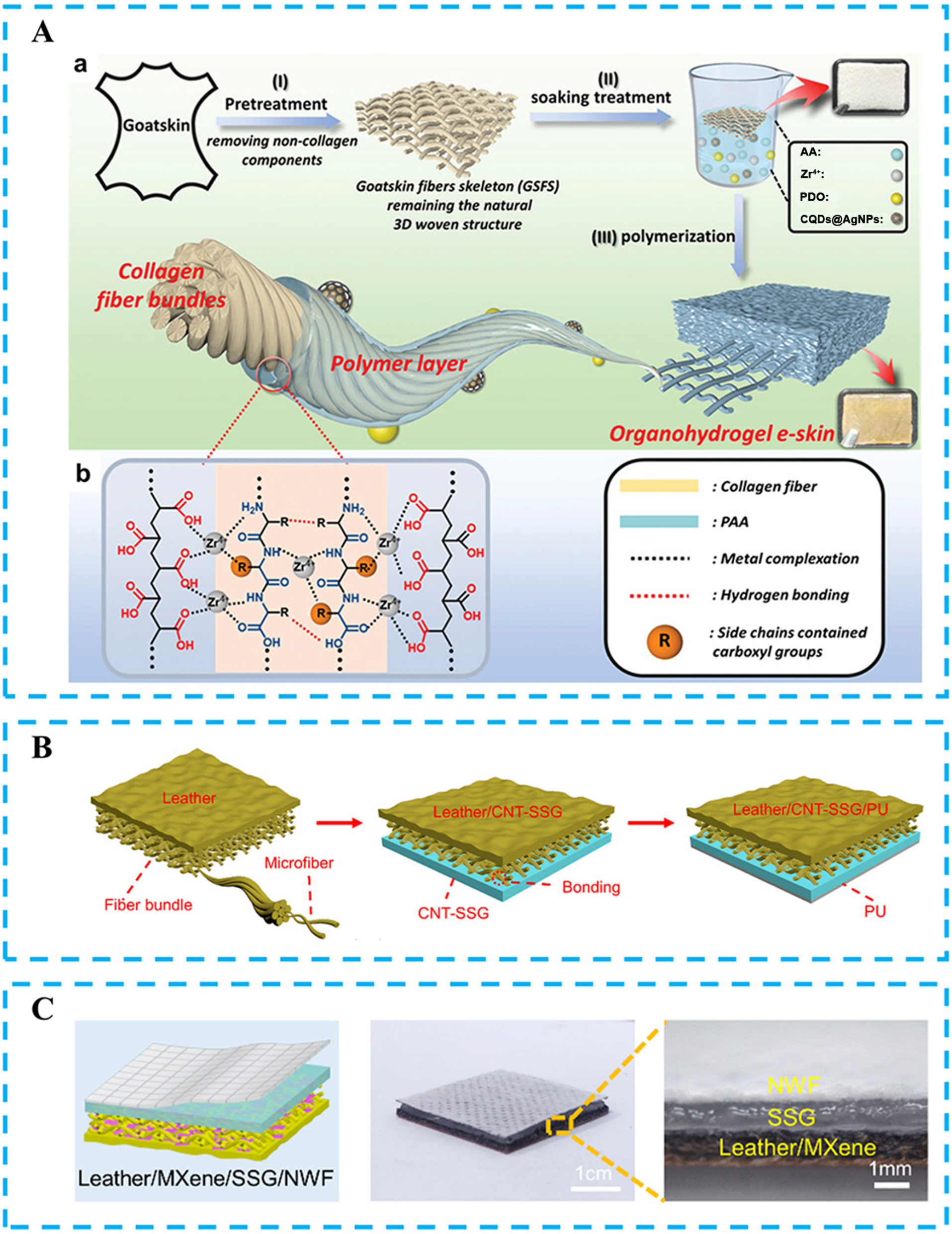

Figure 2. The preparation methods of leather composites. (Aa) Schematic fabrication route of leather composite by *in-situ* polymerization; (Ab) the crosslinking mechanism of leather composite[27]. Reprinted with permission. Copyright 2023, Wiley-VCH; (B) The schematic of Leather/CNT-SSG/NWF; and (C) Leather/MXene/SSG/NWF by multilayer assembly[53,54]. CNT: Carbon nanotube; SSG: shear stiffening gel; NWF: nonwoven fabric.

applied pressure is larger than 10 kPa (stage III), the sensitivity of the piezoresistive sensor begins to decrease because the efficient conductive networks have been constructed [Figure 3Ab and c]. Furthermore, the sensing performance of AgNW/leather composites is stable under multiple pressure cycling current changes and has a short response time (100 ms) [Figure 3Ad and e]. The leather pressure sensor can also monitor finger bending and changes in the throat during speech in real-time, demonstrating excellent responsiveness and stability in human motion detection [Figure 3Af-h].

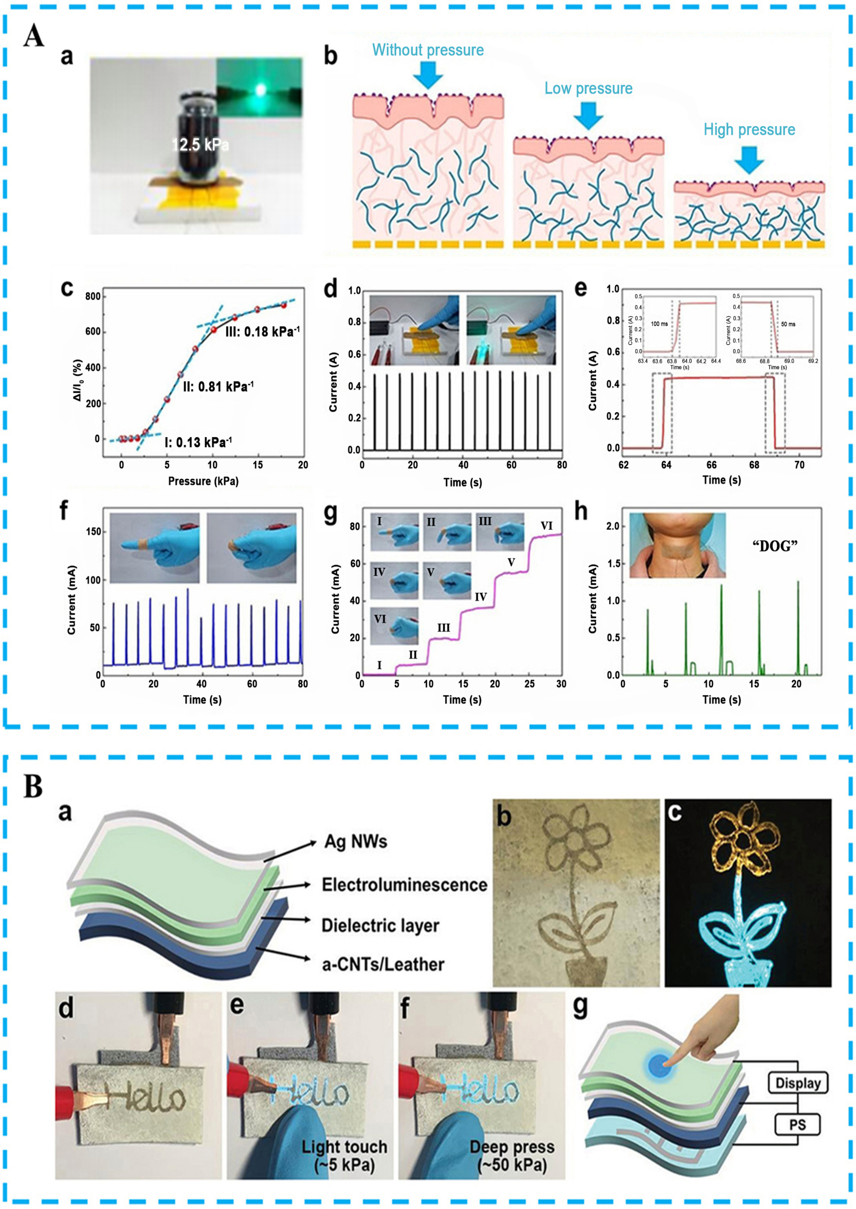

Figure 3. The application directions of leather composites. (Aa) The LED lamp lighting upon pressure; (Ab) illustration for the electric current response mechanism upon externally applied pressures; (Ac) relative current changes of the AgNW/leather nanocomposites upon various pressures, (Ad and e) the piezoresistive sensing performances of the AgNW/leather nanocomposites; (Af-h) human motion detection of the AgNW/leather nanocomposites[41]. Reprinted with permission. Copyright 2022, Wiley-VCH; (Ba) Structure of leather-based display; (Bb-g) smart display based on electroluminescence[42]. Reprinted with permission. Copyright 2019, Wiley-VCH. AgNW: Silver nanowire; CNT: carbon nanotube.

Actually, fabric-based flexible electronics also have good sensing performance; however, their mechanical strength is weaker than the leather composites[60]. Yan *et al.* prepared a polytetrafluoroethylene (PTFE)/AgNW/silk fibroin fabric sensor, and it showed wonderful monitoring[61]. Obviously, its application can be enlarged after improving the low tensile strength. Xie *et al.* introduced carbon black into leather, and then the Kirigami structure was designed[58]. The leather-based strain sensor can accurately identify the bending angle and direction. More importantly, the resistance changes maintained the stability of the strain sensor under 12,000 bending cycles, which showed good durability and mechanical performance. Finally, the shapes also can be recognized by using this sensor. Due to the unique surface structure of leather, the rough fiber surface provides natural advantages for frictional power generation. Moreover, Zhang *et al.* constructed rough MXene films on leather fiber surfaces to improve the performance of frictional power generation. The output voltage of the leather-based triboelectric nanogenerator (TENG) was stable under cyclic impact for 4,000 s, which showed excellent durability. Furthermore, the array sensor was fabricated to achieve the motion control of a mechanical hand, which demonstrated the potential in human-computer interaction applications[21].

With the development of intelligent leather composites, conductive leather is widely used in various industries[30]. By utilizing the sensing capabilities of conductive leather, “dead skin” is “revitalized”, providing a new electronic device design strategy for intelligent sensing, display, and interaction devices[42]. In the dyeing and finishing process of leather making, personalized design is carried out on leather composites to obtain multi-stimuli responsive chromic devices[62]. Zou *et al.* applied an electroluminescent layer on the surface of the conductive leather composite and successfully illuminated them by designing complex patterns such as flowers and words, demonstrating the excellent visual display ability of electronic devices[42]. Furthermore, the brightness of leather-based electronic devices could be varied with the amount of pressure applied, which further controlled the light intensity of the device through pressure and provided real-time visual feedback [Figure 3B]. This design strategy is simple and efficient; thus, it is expected to be intensively applied to develop artificial intelligence and interactive electronic devices.

Electromagnetic interference shielding

With the popularization of electronic devices, concerns about electromagnetic radiation pollution and electromagnetic shielding have become important[31]. Generally, the main ways to shield electromagnetic waves include reflection, absorption, and multiple reflections[63]. It is beneficial for enhancing the electromagnetic shielding effect by designing the structure and conductivity of the material. Leather has the natural dielectric property, which enables dipoles to relax, resulting in dielectric loss to electromagnetic wave energy under the action of electromagnetic waves[43]. Secondly, after functionalization by various conductive materials, the 3D collagen fiber network of the leather will induce electromagnetic waves to undergo multiple reflections in the conductive network, resulting in Ohmic loss to consume electromagnetic wave energy. For example, Bai *et al.* used polypyrrole (PPy), superconductive carbon black (SCB), 1*H*, 1*H*, 2*H*, 2*H*-perfluorodecyltriethoxysilane (PFDTES), and PDMS to nano-engineer the design of leather[64]. The resulting PPy/SCB@PP-CFs with high conductivity (6.5 S/m) show significant electromagnetic shielding ability [Figure 4A]. It also indicates that the thickness of leather composites is positively correlated with electromagnetic shielding performance. As the thickness increases, the time for multiple reflections of electromagnetic waves on leather increases; then, more electromagnetic wave energy is consumed. At the same time, leather, as a promising natural material, has excellent X-ray protection capabilities due to its multilayer woven structure that complements other functional materials[65-67].

Flame retardant

Leather is inherently flammable since it is composed of a large number of collagen fibers and contains elements such as carbon, nitrogen, hydrogen, and oxygen[68], which limits its applications. Therefore, it is very important to develop leather composites with flame-retardant properties[69]. Recently, many intelligent fire-safe fabrics have been developed by modifying the surface of textiles with flame retardants[70-72]. The preparation of leather requires tanning and fat-liquoring, which greatly facilitates the addition of flame retardants and can be directly introduced into the leather-making process[73]. Wang *et al. in situ* grew silica particles on the 3D framework of leather and sprayed silica particles on the surface to obtain a thermal insulation layer[37]. The leather composite did not ignite after direct contact with the flame, demonstrating its excellent flame retardancy [Figure 4B]. Lyu *et al.* added montmorillonite and layered double hydroxide to leather during the fat-liquoring process to form a synergistic flame retardant system that enhances the

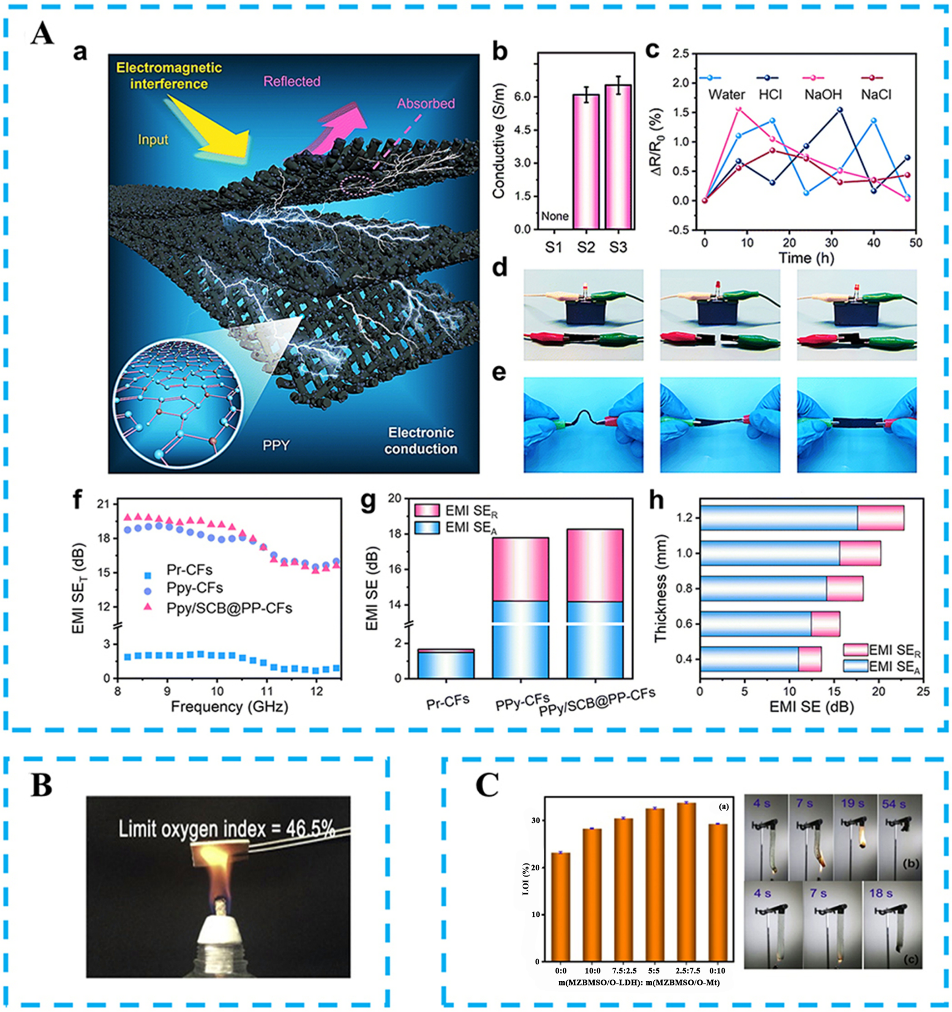

Figure 4. The application directions of leather composites. (Aa) Schematic illustration of electromagnetic interference shielding and electronic conductivity of PPy/SCB@PP-CFs; (Ab-e) the characterization of electrical characteristics; (Af-h) the EMI effectiveness of PPy/SCB@PP-CFs[64]. Reprinted with permission. Copyright 2023, Royal Society of Chemistry; (B) and (C) The flame retardant performance of leather composites[25,37]. Reprinted with permission. Copyright 2018, Elsevier. Reprinted with permission. Copyright 2021, Elsevier. PPy: Polypyrrole; SCB: superconductive carbon black.

protection of leather fibers[25]. The ultimate limiting oxygen index of leather composites reaches 33.8%, and leather composites can quickly self-extinguish after leaving the flame, greatly improving the fire safety of leather [Figure 4C].

Safeguarding

Leather was used as a popular armor material in ancient times, but its protective performance had not been further improved as time went by[74]. Considering its wear resistance and mechanical strength, the development of leather composites with excellent protective properties has gradually attracted attention[24]. Surianarayanan *et al.* treated leather with silane to improve its load distribution and impact resistance, which enabled leather composite to absorb more impact energy[75]. The multilayer structure assembled by leather composites greatly improves its load-bearing and fatigue performance. Fan *et al.* used a rate-dependent SSG to strengthen the natural leather, and then the cold flow effect of SSG allowed it to penetrate slightly into the porous structure of the leather, resulting in a flexible-tough coupled leather composite. It exhibits excellent impact resistance under different impacts[53]. More recently, to enlarge its application, the leather layer was treated by MXene nanosheets for functional design. The treated leather composite has excellent conductivity and no reduced impact resistance[54], which can provide sensing feedback under different impacts. Finally, an intelligent impact resistance device based on the wearable Leather/MXene/SSG/NWF safeguarding leather composite was obtained by integrating a wireless transmission system; therefore, the final system could monitor the impact status of the leather composite in real time [Figure 5A]. In short, the development of intelligent leather composites opened up new avenues in the field of intelligent protection.

Thermoregulatory clothing

Recently, many studies have reported the development of multifunctional textiles for intelligent thermoregulation, which could maintain human body temperature in a comfortable area[76]. Most animal fur benefits from the low thermal conductivity brought by their multilayer and porous skin structure; thus, they can effectively reduce heat loss and have a natural insulation effect[37]. In this case, the most common use of leather in daily life is in the field of clothing[5]. It is worth noting that during sudden changes in weather or harsh environments, a single insulation performance may not be able to meet the temperature needs of the human body[77]. Mo *et al.* developed an asymmetric double-layer leather composite. A porous cellulose acetate layer achieved passive radiation cooling on the corium side, and a highly connected CNT network exerted passive radiative heating and Joule-heating ability on the other side[45]. The two different functions can switch between cooling and heating modes according to environmental conditions by turning over two sides of the fabric, thus achieving high adaptability to weather changes [Figure 5B]. Fan *et al.* designed a conductive MXene array on a leather substrate, resulting in a leather vest that could achieve regional electric heating[54]. By controlling voltage, the leather vest could precisely regulate the temperature regulation, which could be used to cope with extreme cold environments by providing the required temperature for the human body [Figure 5C]. As a result, the above analysis demonstrates that the leather composites exhibit enormous application potential in intelligent thermoregulatory clothing.

SOFT WEARABLE ROBOTICS FOR FUTURE APPLICATIONS BASED ON LEATHER COMPOSITES

In recent years, soft wearable robotics has received increasing attention due to the rapid development of flexible sensors and human-robot interface applications[78]. Xiloyannis *et al.* developed a soft and textile-based robotic exoskeleton for assisting hand opening and closing [Figure 6A][79]. Schmidt *et al.* introduced a soft and wearable device to provide gravity support to the user's knee and hip joints [Figure 6B][80]. These resulting soft robotic devices based on textiles could help the wearer engage in simple daily activities. Leather composites have good biocompatibility and natural fiber structure, but research on the application of soft robots is scarce. Dong *et al.* prepared a smart conductive leather skin to imitate the functions of human skin[50]. Furthermore, a smart glove based on leather skin was designed to monitor complex hand movements. At the same time, a human-computer interaction function has been developed based on the smart glove, which can control the movement of the robotic arm in real time by using gestures [Figure 6C and D]. A broad research foundation of leather composites has been built in intelligent applications. Intelligent leather composites will expand their research gaps in the field of soft robots in the future by combining their biocompatibility and mechanical strength. Ultimately, it is expected to develop soft robots that can cope with different complex environments, such as biomimetic soft robots, radiation-resistant robots, safety protection robots, *etc*.

CONCLUSION AND OUTLOOK

Natural leather has a unique multilayer structure and collagen fiber network, which is easy to combine with

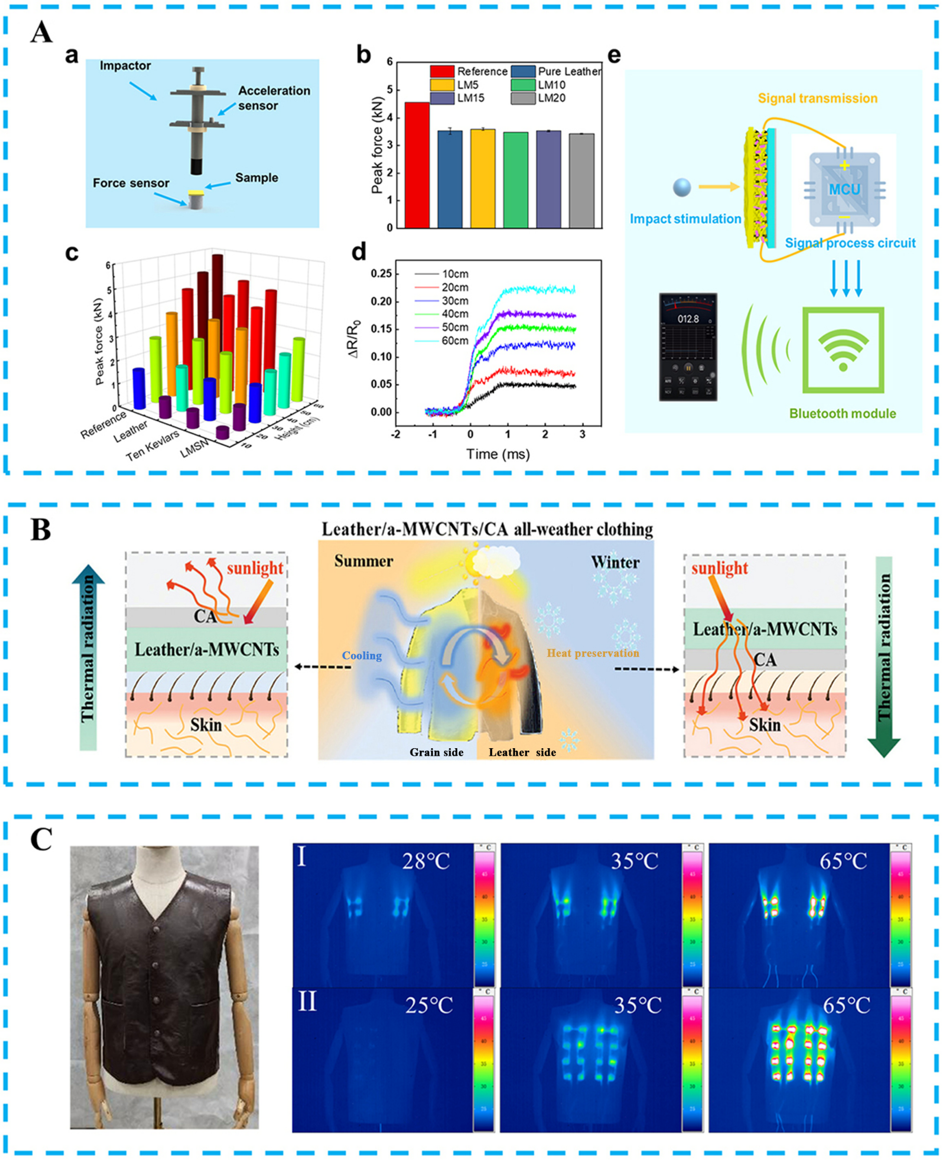

Figure 5. The application directions of leather composites. (A) The impact-resistant and impact-sensing performances of Leather/MXene/SSG/NWF[54]; (B) The cooling and heating modes of leather/a-MWCNTs/CA all-weather fabric[45]. Reprinted with permission. Copyright 2023, Wiley-VCH; (C) The optical image and IR camera images of intelligent thermoregulatory clothing[54]. SSG: Shear stiffening gel; NWF: nonwoven fabric; a-MWCNTs: acidified multiwalled carbon nanotubes.

various functional materials, taking advantage of each other to obtain soft, wear-resistant, and editable intelligent leather composites. This perspective reviews the recent research progress of leather composites, summarizing and analyzing the existing preparation methods of leather composites. Furthermore, this work also emphasizes various popular application directions based on the different functionalities of leather composites, such as flexible sensors, electromagnetic interference shielding, flame retardant protection, impact-resistant safety protection devices, and intelligent thermoregulatory clothing. In addition, although great application prospects have been achieved on leather composites, there are still significant difficulties and challenges in practical applications.

Figure 6. The application of soft wearable robotics. (A) Soft robotic glove for grasping assistance[79]. Reprinted with permission. Copyright 2017, SAGE; (B) The Myosuit for supporting the user's knee and hip joints. Reprinted with permission. Copyright 2017, Frontiers Media[80]; (C) Schematic diagrams of the smart glove and its controlling circuits and (D) various hand gestures of smart gloves in real-time[50]. Reprinted with permission. Copyright 2017, Elsevier.

Firstly, economical, environmentally friendly, safe, and efficient functional materials can be developed to reduce the production cost of leather composites and promote their faster application from the source. Secondly, it is necessary to strengthen the stability and durability of leather collagen fibers combined with functional materials, maintaining the breathability and comfort of leather to ensure the stable performance and advantages of leather composites during the application process. Thirdly, the preparation process of leather composites should be improved. The filtration, spraying, *in-situ* growth, and other methods of the leather-making process should be industrialized, mechanized, and simplified so that intelligent leather composites can embark on the path of industrialization. Fourthly, specific performance in the application direction can be further deepened. Intelligent leather, such as conductive leather, flame-retardant leather, and electromagnetic shielding leather, should be developed toward similar industry standards, and a performance standard system for functional leather can be established to standardize production and performance standards. Finally, developing multifunctional sensing for leather composites can enable leather to perceive the world similarly to human skin by combining machine learning to carry out human-computer interactions.

In conclusion, as an emerging flexible intelligent biomaterial, leather composites have demonstrated enormous potential for various applications, especially in the fields of sensing and safety protection. This coincides with the safety and real-time sensing of some soft robots in hazardous environments. Therefore, leather composites can be developed towards biomimetic, intelligent, and thermo-mechanical coupled safety protection in the future. It is believed that this result can open up new research directions for the design and safe use of soft robots.

DECLARATIONS

Authors' contributions

Wrote the original draft: Fan Z, Sang M

Supervised, reviewed, and revised the manuscript: Leung KCF, Gong X, Xuan S

Availability of data and materials

Not applicable.

Financial support and sponsorship

Financial support from the National Natural Science Foundation of China (Grant Nos. 12072338, 12132016, and 12202435), the Anhui's Key R&D Program of China (202104a05020009), the Aviation Science Foundation of China (20200029079004), the Fundamental Research Funds for the Central Universities (WK2480000007) is gratefully acknowledged.

Conflicts of interest

All authors declared that there are no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

Copyright

REFERENCES

1. Falcão L, Araújo MEM. Vegetable tannins used in the manufacture of historic leathers. *Molecules* 2018;23:1081. DOI PubMed PMC
2. Ariram N, Madhan B. Development of bio-acceptable leather using bagasse. *J Clean Prod* 2020;250:119441. DOI
3. Shi H, Jiang M, Zhang T, Chen S, Li L, Li Y. Multifunctional silver decorated leather solid waste/poly(vinyl alcohol) nanocomposites for electromagnetic interference shielding, joule heating and crude-oil cleaning. *ACS Sustain Chem Eng* 2022;10:13165-75. DOI
4. Jima WD, Dada TK, Palanisamy T. Cool garment leathers for hot environment. *J Therm Anal Calorim* 2019;135:3289-95. DOI
5. Memon H, Chaklie EB, Yesuf HM, Zhu C. Study on effect of leather rigidity and thickness on drapability of sheep garment leather. *Materials* 2021;14:4553. DOI PubMed PMC
6. Wang Y, Zheng M, Liu X, Yue O, Wang X, Jiang H. Advanced collagen nanofibers-based functional bio-composites for high-value utilization of leather: a review. *J Sci Adv Mater Dev* 2021;6:153-66. DOI
7. An W, Ma J, Xu Q, Zhang H. Nanoarchitectonics of flame retardant leather: current status and future perspectives. *Compos Part A Appl Sci Manuf* 2023;165:107327. DOI

8. Li S, Zhang J, He J, et al. Functional PDMS elastomers: bulk composites, surface engineering, and precision fabrication. *Adv Sci* 2023;10:e2304506. DOI PubMed PMC
9. Zhang M, Wang L, Xu H, Song Y, He X. Polyimides as promising materials for lithium-ion batteries: a review. *Nanomicro Lett* 2023;15:135. DOI PubMed PMC
10. Kim DH, Kim YS, Wu J, et al. Ultrathin silicon circuits with strain-isolation layers and mesh layouts for high-performance electronics on fabric, vinyl, leather, and paper. *Adv Mater* 2009;21:3703-7. DOI
11. Xu S, Shi B. A green and sustainable strategy for leather manufacturing: endow dehydrated hide with consistent and durable hydrophobicity. *J Clean Prod* 2023;383:135526. DOI
12. Tian Z, Ma J, Liu Q, Zhang H. Preparation and application of novel amphoteric acrylic retanning agents to improve dye absorption. *React Chem Eng* 2023;8:645-55. DOI
13. Ke L, Wang Y, Ye X, Luo W, Huang X, Shi B. Collagen-based breathable, humidity-ultrastable and degradable on-skin device. *J Mater Chem C* 2019;7:2548-56. DOI
14. Pei Y, Yang W, Tang K, Kaplan DL. Collagen processing with mesoscale aggregates as templates and building blocks. *Biotechnol Adv* 2023;63:108099. DOI PubMed
15. Wilson NH, Ragothaman M, Palanisamy T. Bimetallic copper-iron oxide nanoparticle-coated leathers for lighting applications. *ACS Appl Nano Mater* 2021;4:4055-69. DOI
16. Qin R, Luo X, Feng J, et al. A novel eco- and user-friendly graphene/leather-based composite for real-time mechano-monitoring of human motion. *J Clean Prod* 2022;371:133360. DOI
17. Guo Q, Guo J, Chen H, et al. Multi-functional graphene/leather for versatile wearable electronics. *J Mater Chem A* 2023;11:11773-85. DOI
18. Wang X, Yue O, Liu X, Hou M, Zheng M. A novel bio-inspired multi-functional collagen aggregate based flexible sensor with multi-layer and internal 3D network structure. *Chem Eng J* 2020;392:123672. DOI
19. Stanca M, Gaidau C, Alexe CA, et al. Multifunctional leather surface design by using carbon nanotube-based composites. *Materials* 2021;14:3003. DOI PubMed PMC
20. Xie R, Du Q, Zou B, et al. Wearable leather-based electronics for respiration monitoring. *ACS Appl Bio Mater* 2019;2:1427-31. DOI
21. Zhang S, Xiao Y, Chen H, et al. Flexible triboelectric tactile sensor based on a robust MXene/leather film for human-machine interaction. *ACS Appl Mater Interfaces* 2023;15:13802-12. DOI
22. Kebede ZT, Tadesse MG, Chane TE, Mengistie DA. Application of PEDOT:PSS conductive polymer to enhance the conductivity of natural leather: retanning process. *J Nanomater* 2023;2023:1-9. DOI
23. Hu X, Liu M, Cheng Y, Hu J. Light-responsive mesoporous silica nanoparticles loaded with osmanthus fragrance for improving leather odor. *ACS Appl Nano Mater* 2022;5:1317-26. DOI
24. P Bhasi A, Hanna Wilson N, Palanisamy T. Nanosized hexagonal boron nitride and polyethylene glycol-filled leathers for applications demanding high thermal insulation and impact resistance. *ACS Omega* 2022;7:45120-8. DOI PubMed PMC
25. Lyu B, Luo K, Gao D, Wang Y, Ma J. Synergistic effect of layered double hydroxide and montmorillonite: towards super-efficient fireproofing of leather. *Appl Clay Sci* 2021;212:106215. DOI
26. Renganath Rao R, Sathish M, Raghava Rao J. Research advances in the fabrication of biosafety and functional leather: a way-forward for effective management of COVID-19 outbreak. *J Clean Prod* 2021;310:127464. DOI PubMed PMC
27. Fan X, Ke T, Gu H. Multifunctional, ultra-tough organohydrogel e-skin reinforced by hierarchical goatskin fibers skeleton for energy harvesting and self-powered monitoring. *Adv Funct Mater* 2023;33:2304015. DOI
28. Bai Z, Wang X, Zheng M, et al. Mechanically robust and transparent organohydrogel-based e-skin nanoengineered from natural skin. *Adv Funct Mater* 2023;33:2212856. DOI
29. Chen YY, Xie RJ, Zou BH, et al. CNT@leather-based electronic bidirectional pressure sensor. *Sci China Technol Sci* 2020;63:2137-46. DOI
30. Wegene JD, Thanikaivelan P. Conducting leathers for smart product applications. *Ind Eng Chem Res* 2014;53:18209-15. DOI
31. Liu C, Huang X, Zhou J, et al. Lightweight and high-performance electromagnetic radiation shielding composites based on a surface coating of Cu@Ag nanoflakes on a leather matrix. *J Mater Chem C* 2016;4:914-20. DOI
32. Lyu B, Luo K, Wang Y, Gao D, Ma J. Sodium alginate oxide assembly layered double hydroxide and its structure-activity relationship to anti-fogging properties and flame retardancy of leather. *Appl Clay Sci* 2020;190:105559. DOI
33. An W, Ma J, Xu Q, Zhang H, Wei L, Yuan L. Construction of hetero-structured fillers to significantly enhance the fire safety of bio-based nanocomposite coating. *Appl Surf Sci* 2022;575:151767. DOI
34. Xiang J, Yu R, Yang L, et al. Breathable, antibacterial, and biocompatible collagen fiber network decorated with zwitterionic silver nanoparticles for plantar pressure monitoring. *ACS Appl Mater Interfaces* 2022;14:21645-56. DOI
35. Zhang K, Kang N, Zhang B, et al. Skin conformal and antibacterial PPy-leather electrode for ECG monitoring. *Adv Elect Mater* 2020;6:2000259. DOI
36. Nguyen NT, Vu TH, Bui VH. Antibacterial and antifungal fabrication of natural lining leather using bio-synthesized silver nanoparticles from *piper betle* L. leaf extract. *Polymers* 2023;15:2634. DOI PubMed PMC
37. Wang X, Tang Y, Wang Y, et al. Leather enabled multifunctional thermal camouflage armor. *Chem Eng Sci* 2019;196:64-71. DOI
38. Sun X, Wang Q, Zhan J, et al. Superhydrophobic conductive suede fabrics based on carboxylated multiwalled carbon nanotubes and polydopamine for wearable pressure sensors. *ACS Appl Nano Mater* 2023;6:10746-57. DOI

39. Omoloso O, Mortimer K, Wise WR, Jraisat L. Sustainability research in the leather industry: a critical review of progress and opportunities for future research. *J Clean Prod* 2021;285:125441. DOI
40. Liu X, Zheng C, Luo X, Wang X, Jiang H. Recent advances of collagen-based biomaterials: multi-hierarchical structure, modification and biomedical applications. *Mater Sci Eng C Mater* 2019;99:1509-22. DOI
41. Ma Z, Xiang X, Shao L, Zhang Y, Gu J. Multifunctional wearable silver nanowire decorated leather nanocomposites for joule heating, electromagnetic interference shielding and piezoresistive sensing. *Angew Chem Int Ed Engl* 2022;61:e202200705. DOI PubMed
42. Zou B, Chen Y, Liu Y, et al. Repurposed leather with sensing capabilities for multifunctional electronic skin. *Adv Sci* 2019;6:1801283. DOI PubMed PMC
43. Liu C, Wang X, Huang X, Liao X, Shi B. Absorption and reflection contributions to the high performance of electromagnetic waves shielding materials fabricated by compositing leather matrix with metal nanoparticles. *ACS Appl Mater Interfaces* 2018;10:14036-44. DOI
44. Li Q, Zhong R, Xiao X, Liao J, Liao X, Shi B. Lightweight and flexible Bi@Bi-La natural leather composites with superb X-ray radiation shielding performance and low secondary radiation. *ACS Appl Mater Interfaces* 2020;12:54117-26. DOI
45. Mo C, Lei X, Tang X, et al. Nanoengineering natural leather for dynamic thermal management and electromagnetic interference shielding. *Small* 2023;19:2303368. DOI
46. Ye R, James DK, Tour JM. Laser-induced graphene: from discovery to translation. *Adv Mater* 2019;31:e1803621. DOI PubMed
47. Luo Y, Miao Y, Wang H, et al. Laser-induced Janus graphene/poly(p-phenylene benzobisoxazole) fabrics with intrinsic flame retardancy as flexible sensors and breathable electrodes for fire-fighting field. *Nano Res* 2023;16:7600-8. DOI
48. Yang D, Nam HK, Le TSD, et al. Multimodal E-textile enabled by one-step maskless patterning of femtosecond-laser-induced graphene on nonwoven, knit, and woven textiles. *ACS Nano* 2023;17:18893-904. DOI
49. Wang Z, Chen B, Sun S, Pan L, Gao Y. Maskless formation of conductive carbon layer on leather for highly sensitive flexible strain sensors. *Adv Elect Mater* 2020;6:2000549. DOI
50. Dong D, Yang Y, Zhang H, et al. Nanocatalysts induced self-triggering leather skin for human-machine interaction. *Chem Eng J* 2023;454:140269. DOI
51. Zou X, Wang X, Gou M, et al. Ultra-strong adhesive, self-healing and electroactive bio-based hydrogels for the on-demand fabrication of sandwich-inspired smart electronic sensing floors. *J Mater Chem A* 2022;10:14555-67. DOI
52. Sang M, Zhang J, Liu S, et al. Advanced MXene/shear stiffening composite-based sensor with high-performance electromagnetic interference shielding and anti-impacting Bi-protection properties for smart wearable device. *Chem Eng J* 2022;440:135869. DOI
53. Fan Z, Zhao C, Wu J, et al. Intelligent safeguarding leather with excellent energy absorption *via* the toughness-flexibility coupling designation. *Compos Part A Appl Sci Manuf* 2022;161:107078. DOI
54. Fan Z, Lu L, Sang M, et al. Wearable safeguarding leather composite with excellent sensing, thermal management, and electromagnetic interference shielding. *Adv Sci* 2023;10:e2302412. DOI PubMed PMC
55. Ayyappan VG, Prakash D, Jaisankar SN, Sadhukhan N, Alam MS, Samanta D. Nanoconjugates of methacrylic polymers: synthesis, characterization, and immobilization to leather. *J Appl Polym Sci* 2020;137:48627. DOI
56. Shajari S, Ramakrishnan S, Karan K, Sudak LJ, Sundararaj U. Ultrasensitive wearable sensor with novel hybrid structures of silver nanowires and carbon nanotubes in fluoroelastomer: multi-directional sensing for human health monitoring and stretchable electronics. *Appl Mater Today* 2022;26:101295. DOI
57. Khalid MAU, Chang SH. Flexible strain sensors for wearable applications fabricated using novel functional nanocomposites: a review. *Compos Struct* 2022;284:115214. DOI
58. Xie R, Hou S, Chen Y, et al. Leather-based strain sensor with hierarchical structure for motion monitoring. *Adv Mater Technol* 2019;4:1900442. DOI
59. Xie R, Zhu J, Wu H, et al. 3D-conductive pathway written on leather for highly sensitive and durable electronic whisker. *J Mater Chem C* 2020;8:9748-54. DOI
60. Wang Q, Sheng H, Lv Y, et al. A skin-mountable hyperthermia patch based on metal nanofiber network with high transparency and low resistivity toward subcutaneous tumor treatment (Adv. Funct. Mater. 21/2022). *Adv Funct Mater* 2022;32:2270123. DOI
61. Yan X, Chen S, Zhang G, et al. Highly breathable, surface-hydrophobic and wet-adhesive silk based epidermal electrode for long-term electrophysiological monitoring. *Compos Sci Technol* 2022;230:109751. DOI
62. Jia L, Zeng S, Ding H, et al. Leather-based multi-stimuli responsive chromisms. *Adv Funct Mater* 2021;31:2104427. DOI
63. Al-saleh MH, Saadeh WH, Sundararaj U. EMI shielding effectiveness of carbon based nanostructured polymeric materials: a comparative study. *Carbon* 2013;60:146-56. DOI
64. Bai Z, Wang X, Huang M, et al. Versatile nano-micro collagen fiber-based wearable electronics for health monitoring and thermal management. *J Mater Chem A* 2023;11:726-41. DOI
65. Wang Y, Ding P, Xu H, et al. Advanced X-ray shielding materials enabled by the coordination of well-dispersed high atomic number elements in natural leather. *ACS Appl Mater Interfaces* 2020;12:19916-26. DOI
66. Li H, Zhou J, Yan L, et al. Barbican-inspired bimetallic core-shell nanoparticles for fabricating natural leather-based radiation protective materials with enhanced X-ray shielding capability. *Chem Eng J* 2023;466:143355. DOI
67. Li Q, Wang Y, Xiao X, et al. Research on X-ray shielding performance of wearable Bi/Ce-natural leather composite materials. *J Hazard Mater* 2020;398:122943. DOI
68. Yang L, Liu Y, Ma C, et al. Kinetics of non-isothermal decomposition and flame retardancy of goatskin fiber treated with melamine-

based flame retardant. *Fibers Polym* 2016;17:1018-24. DOI

69. Lyu B, Luo K, Gao D, Wang Y, Ma J. Modified layered double hydroxide/zanthoxylum bungeanum seed oil composites to improve the flame retardant of leather. *Polym Degrad Stab* 2021;183:109430. DOI
70. Tang T, Wang S, Jiang Y, et al. Flexible and flame-retarding phosphorylated MXene/polypropylene composites for efficient electromagnetic interference shielding. *J Mater Sci Technol* 2022;111:66-75. DOI
71. Liu L, Ma Z, Zhu M, et al. Superhydrophobic self-extinguishing cotton fabrics for electromagnetic interference shielding and human motion detection. *J Mater Sci Technol* 2023;132:59-68. DOI
72. Ma Z, Zhang J, Maluk C, et al. A lava-inspired micro/nano-structured ceramifiable organic-inorganic hybrid fire-extinguishing coating. *Matter* 2022;5:911-32. DOI
73. Duan B, Wang Q, Wang X, Li Y, Zhang M, Diao S. Flame retardance of leather with flame retardant added in retanning process. *Results Phys* 2019;15:102717. DOI
74. Perini N, Mercuri F, Thaller MC, et al. The stain of the original salt: red heats on chrome tanned leathers and purple spots on ancient parchments are two sides of the same ecological coin. *Front Microbiol* 2019;10:2459. DOI PubMed PMC
75. Surianarayanan P, Balaji N, Balasubramanian K. Effect of silane-treated chitosan carbohydrate polymer and tanned leather/areca fiber hybrid epoxy composites on mechanical, drop load, and fatigue properties. *Biomass Conv Bioref* 2023. DOI
76. Lei L, Shi S, Wang D, et al. Recent advances in thermoregulatory clothing: materials, mechanisms, and perspectives. *ACS Nano* 2023;17:1803-30. DOI
77. Xie L, Wang X, Bai Z, et al. Facile "synergistic inner-outer activation" strategy for nano-engineering of nature-skin-derived wearable daytime radiation cooling materials. *Small* 2023;19:2207602. DOI
78. Zannat A, Uddin MN, Mahmud ST, Prithu PSS, Mia R. Review: textile-based soft robotics for physically challenged individuals. *J Mater Sci* 2023;58:12491-536. DOI
79. Xiloyannis M, Cappello L, Binh KD, Antuvan CW, Masia L. Preliminary design and control of a soft exosuit for assisting elbow movements and hand grasping in activities of daily living. *J Rehabil Assist Technol Eng* 2017;4:2055668316680315. DOI PubMed PMC
80. Schmidt K, Duarte JE, Grimmer M, et al. The myosuit: Bi-articular anti-gravity exosuit that reduces hip extensor activity in sitting transfers. *Front Neurorobot* 2017;11:57. DOI PubMed PMC