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Hydrogel-Based Sensors for Human−Machine Interaction

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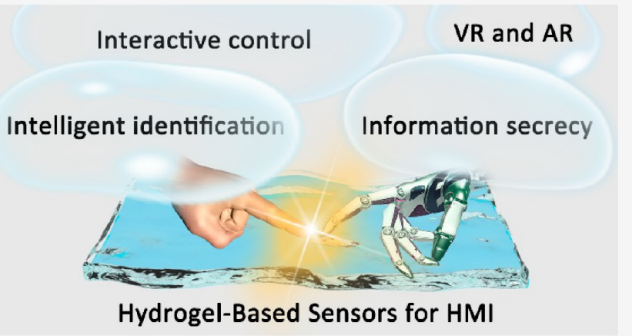
ABSTRACT: In the past decades, remarkable progress has been made in the field of human−machine interaction. The need for accurate sensing devices with satisfactory user experiences has propelled the development of flexible, stretchable, biocompatible, and imperceptible hydrogel-based interfaces. These innovative interfaces facilitate direct interactions between humans and machines while receiving detected input signals from sensors and giving output commands to controllers, thus motivating accurate real-time responsiveness. This Perspective discusses the sensing mechanisms for the two categories of hydrogel-based sensors and summarizes the recent progress in the development of different representations of human−machine interactions, including intelligent identification, information secrecy, interactive control, and virtual reality and augmented reality technologies. The advantages of hydrogel-based systems over conventionally used rigid electrical components are explicitly discussed. The conclusion provides a perspective on current challenges and outlines a future roadmap for the realization of state-of-the-art hydrogel-based smart systems.



INTRODUCTION

Human−machine interaction provides opportunities for bidirectional communication between humans and automated systems while satisfying application requirements and enriching user experiences, and it has emerged as a rapidly expanding state-of-the-art research topic. The concept of a human−machine interface (HMI) has attracted considerable social attention since the third industrial revolution and has gone through three revolutionary generations until now with the advent of industry 4.0. Human−machine interfaces have evolved from buttons and diodes (HMI 1.0) through desktop and touch panel visualizations (HMI 2.0) to web applications and portable devices (HMI 3.0). Furthermore, the integration of virtual reality (VR) and augmented reality (AR) provides future promise for enhancing cognitive and intelligent spaces (HMI 4.0).[1] At the current generation of HMI 3.0, wearable and implantable electronic sensors and devices are the most prevalent types of human−machine interfaces, functionalizing in the fields of health and safety monitoring, medical rehabilitation, robotics, and entertainment.[2,3] However, despite the exceptional operating performance and durability provided by these conventional electronic devices containing hard semiconductors, their inflexibility and lack of conformability have limited the suitability of these devices in certain applications where intimate contact with the human body is required.

Different from the HMIs that involve inorganic and nonbiodegradable rigid devices, hydrogel-based devices offer the benefits of being adhesive, flexible, stretchable, imperceptible, biocompatible, and self-healable, and they have emerged as one of the most extensively interdisciplinary research fields.[4] Hydrogels do not dissolve or disintegrate during swelling as a result of their cross-linked structures. In chemical cross-linking, the polymer chains are covalently bonded via cross-linking agents, while in physical cross-linking, the chains are dynamically linked by building blocks based on noncovalent interactions. Inspired by the flexible natural hydrogels found in biological soft tissues, synthetic hydrogels have been developed and have been validated to be superior to conventional electronic components, thanks to their mechanical properties and biocompatible nature with relatively similar levels of Young's moduli to biological tissues.[2,5] These close similarities in Young's moduli help to minimize the mechanical mismatch with biological neural tissues and stimulate intimate interactions with biological organisms and the human body[6] with the presence of electronic and ionic activities.[7] In recent decades, synthetic hydrogels have been developed to be innovative materials for versatile functions in devices, including sensors, actuators, coatings, optics, electronics, and water harvesters.[8]

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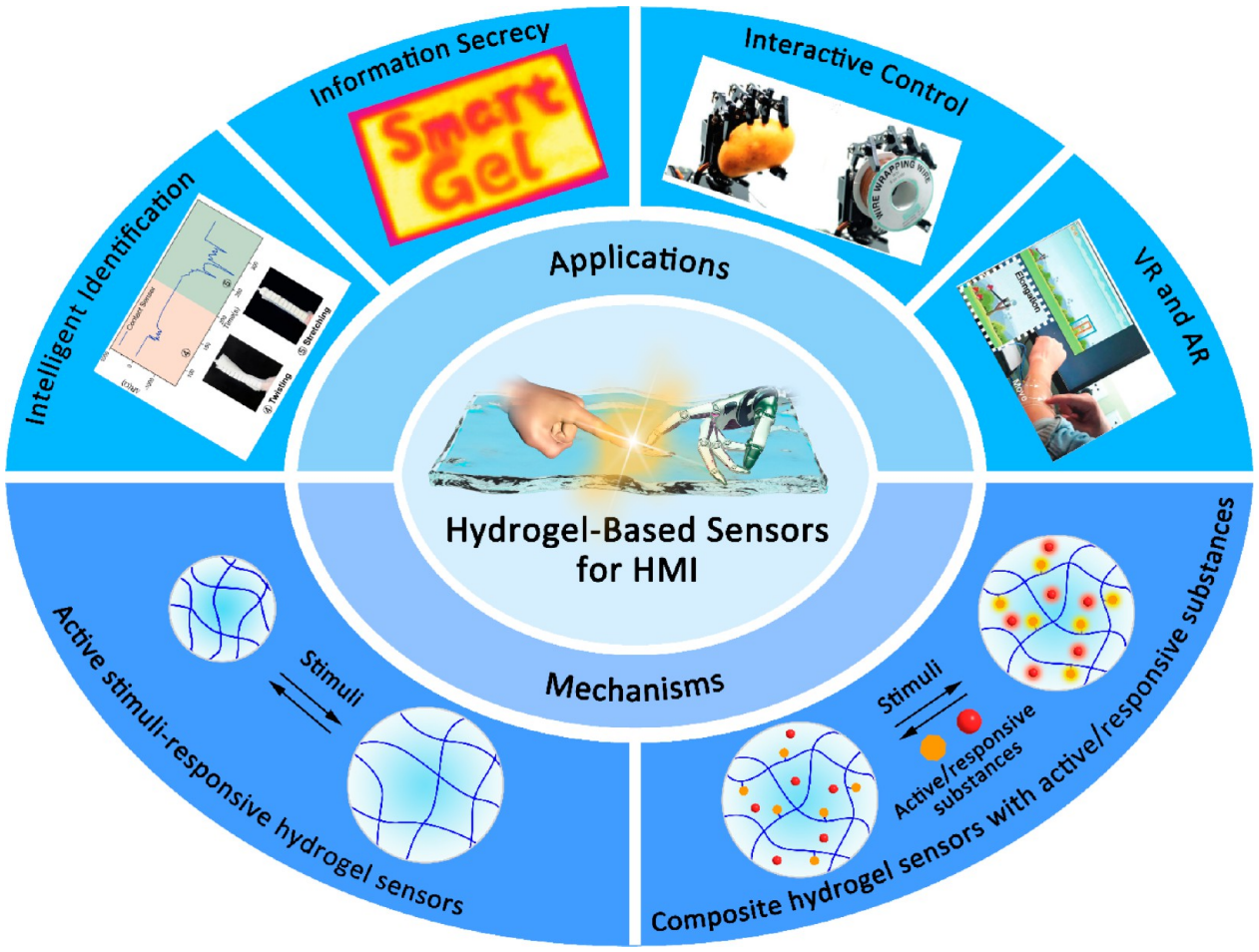

Figure 1. Mechanisms and applications of hydrogel-based sensors in the field of human−machine interaction. The sensors are categorized based on their sensing mechanism as either active hydrogel sensors or composite hydrogel sensors, and the applications mainly focus on intelligent identification (reproduced with permission from ref 12 (copyright 2022 John Wiley & Sons)), information secrecy (reproduced with permission from ref 13 (copyright 2022 Taylor & Francis)), interactive control (reproduced with permission from ref 14 (copyright 2022 John Wiley & Sons)), and virtual reality (VR) and augmented reality (AR) (reproduced with permission from ref 15 (copyright 2020 John Wiley & Sons)).

While individual synthesis techniques, sensing mechanisms of hydrogel-based sensors, and multivariate perspectives in the field of human−machine symbiosis have been reviewed,[9−11] a comprehensive overview of various categories of hydrogel-based sensors to be used for human−machine interactions is still absent. Herein, this Perspective highlights the latest progress in synthetic hydrogel-based sensors in the domain of human−machine interactions (Figure 1). First, explicit explanations of hydrogel-based sensors are given with a focus on the detailed sensing mechanisms of the two categories of hydrogel sensors: active stimuli-responsive hydrogel sensors and composite hydrogel sensors with active/responsive substances. Then, the uses of hydrogels as human−machine interfaces in the fields of intelligent identification, information secrecy, interactive control, and VR/AR-supplemented applications are discussed. Lastly, we conclude by outlining the key challenges and delineating the future feasibility of hydrogel-based systems for human−machine interactions.

HYDROGEL-BASED SENSORS

Sensors are utilized to convert received input signals to understandable and interpretable output signals. The accurate real-time perception of external stimuli dominates in interactive systems, and lightweight hydrogel-based sensors that depend on molecular-level changes in polymer chains and networks have emerged as innovative alternatives to comfortably interact with humans. Depending on the functionalities of the hydrogels in the sensors, hydrogel-based sensors can be categorized into two categories: active stimuli-responsive hydrogels sensors, which exhibit spontaneous changes of volumes and phases in response to physical or chemical stimul,I[16,17] or composite hydrogel sensors, which contain hydrogel matrices and active or responsive substances.[8,18,19] The sensing process of hydrogel-based sensors starts with the detection of external environmental inputs, including chemical and physical stimuli through chemical diffusion or a physical field, followed by the transduction of the input signals to measurable outputs. Interestingly, the output signals of hydrogel-based sensors can be represented in the form of geometrical, optical, biological, and electrical responses.

Active Stimuli-Responsive Hydrogel Sensors. Stimuli-responsive hydrogels are often referred to as smart hydrogels and can experience micro/macroscopic volume changes and phase transitions, which result from the molecular-level variations of the polymer chains and networks with conformational transformations triggered by chemical (e.g., pH, ions, and biomolecules)[20−24] or physical (e.g., temperature, light, and mechanical forces)[25,26] stimuli. Interestingly, these hydrogel-

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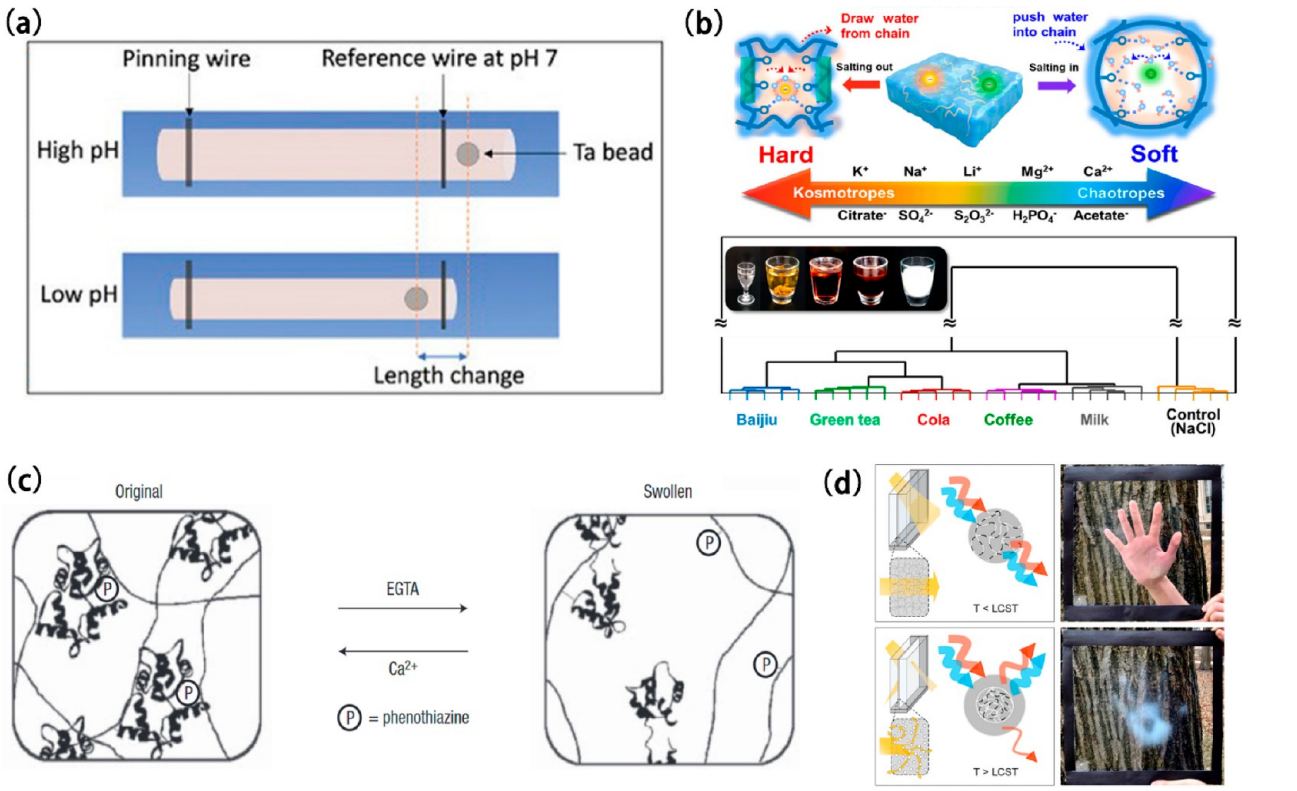

Figure 2. Active stimuli-responsive hydrogel sensors. (a) Implantable pH-responsive hydrogel sensor based on a poly(acrylic acid)-based hydrogel. Reproduced with permission from ref 29. Copyright 2021 John Wiley & Sons. (b) Hofmeister effect on an ion-responsive hydrogel, and the discrimination of different beverages by a gel iontronic sensor. Reproduced with permission from ref 31. Copyright 2023 American Chemical Society. (c) Swelling mechanism of a stimulus-responsive protein hydrogel in response to ethylene glycol-bis(β-aminoethyl ether)-*N*,*N*,*N*′,*N*′-tetraacetic acid (EGTA). Reproduced with permission from ref 23. Copyright 2005 Springer Nature. (d) Thermochromic smart windows based on the phase transition of PNIPAm−AEMA thermo-responsive hydrogel microparticles. Reproduced with permission from ref 25. Copyright 2019 Elsevier.

based sensors can be simultaneously activated by various stimuli, resulting in different responses.

pH-responsive hydrogels are the most studied stimuli-responsive hydrogels, allowing for the reversible change of their volumes, masses, and elasticities in response to variations in pH. The equilibrium constants pK_a and pK_b, which describe the level of ionization of an acid or a base, respectively, are defined to be thresholds for the geometrical change in pH-responsive hydrogels. The volumetric variation of hydrogels is affected by different osmotic swelling forces generated by the electrostatic repulsive forces between the ionized groups. Remarkably, the geometrical changes of pH-responsive hydrogels always occur with electrical signal fluctuations because of the different levels of ion concentrations[27] and optical color shifts, owing to the changes of cross-linking densities and corresponding holographic diffraction wavelengths in Bragg grating sensors.[28] Wijayaratna et al.[29] developed an implantable sensor made of a pH-responsive poly(acrylic acid)-based hydrogel to detect the pH of synovial fluid. When monitoring the length of this hydrogel-based sensor in response to pH, it exhibited a sensitivity of 3 mm/pH unit between a pH of 4 and 8 (Figure 2a).

Ion-responsive hydrogel-based sensors that depend on ionic and electronic conduction have a mechanism similar to that of pH-responsive sensors while also meeting high-sensitivity and high-adhesion requirements due to the various functional groups within the hydrogel for specific ionic responses and adhesion interactions. Polyelectrolyte hydrogels have been adopted to detect variations in pH and ion concentration.[8] Comparably, geometrical and electrical responses are the two most intuitionistic and essential criteria for the sensors. Guenther et al.[30] used hydrogels as chemical−mechanical transducers to monitor metal ions for online analytical systems. Recently, by utilizing a Hofmeister effect-regulated chemical−mechanical interface, Li et al.[31] developed a flexible gel iontronic sensor for discriminating, classifying, and quantifying various cations, anions, amino acids, and saccharides (Figure 2b).

Tremendous progress has been made in the development of biomolecule-based stimuli-responsive hydrogels, including the use of responsive elements such as nucleic acids, saccharides, peptides, and proteins. For the hydrogels centered on nucleic acids, encoded base sequences are functionalized to deliver significant structural and functional information, with the presence of catalytic amplification by polymerization or ligation. As for saccharide-based hydrogels, the stiffness, elasticity, toughness, and swelling properties can be regulated either by the polysaccharide components in response to external triggers during Schiff base cross-linking reactions and ionic interactions or by the nonsaccharide polymer chains, which act as anchoring scaffolds for oligosaccharide receptor units through supramolecular ligand−receptor interactions. Similarly, peptide- and protein-based hydrogels are generally cross-linked by supramolecular structures and specific recognition complexes, with enzymes acting as biocatalysts.[5] In conclusion, the geometric characteristics of these hydrogels would be impacted by biomolecules that are triggered by the variations of polymer chains and cross-linking densities. Miyata et al.[22,23] presented a

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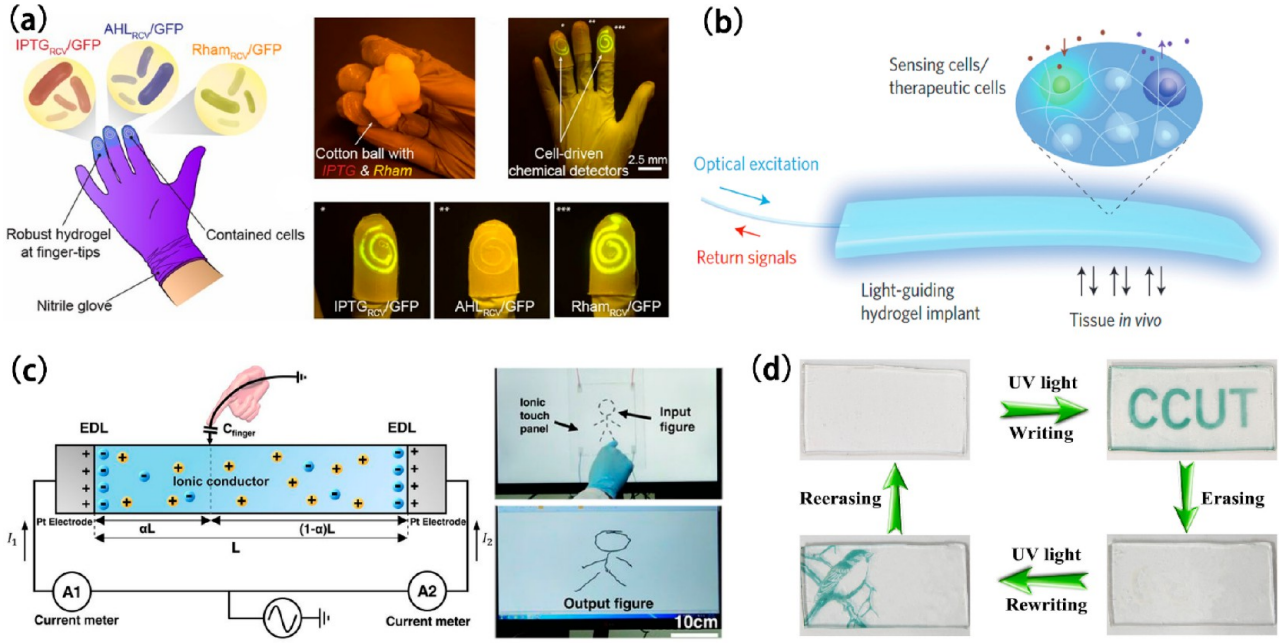

Figure 3. Composite hydrogel sensors with active or responsive substances. (a) Glove with living hydrogel chemical detectors robustly integrated at the fingertips. Reproduced with permission from ref 37. Copyright 2017 National Academy of Science. (b) Light-guiding hydrogel with encapsulated cells establishes bidirectional optical communication with the cells, allowing for the real-time interrogation and control of the biological system in vivo. Reproduced with permission from ref 38. Copyright 2013 Springer Nature. (c) PAAm−LiCl hydrogel-based ionic touch panel. Reproduced with permission from ref 18. Copyright 2016 American Association for the Advancement of Science. (d) Process of rewritable imaging on a hybrid PAAm−Mo3 hydrogel using UV light. Reproduced with permission from ref 41. Copyright 2019 Elsevier.

hydrogel with intrachain antigen−antibody bindings that was capable of detecting free antigens in solution (Figure 2c).

Thermo-responsive hydrogel-based sensors can be categorized as either positive or negative temperature-stimulated sensing systems, which have contrary transition sensing mechanisms determined by the hydrogel's critical solution temperature.[32] Taking negative temperature hydrogels as an example, when the surrounding temperature is lower than the lower critical solution temperature (LCST), hydrogen bonds can be formed by the interaction of hydrophilic materials and water, thus improving the dissociation of the polymer chains and the swelling of the hydrogels. As the temperature increases to be >LCST, hydrophobic contents dominate the operation over the hydrophilic parts, resulting in the association of interpolymer chains and the shrinking of the hydrogels.[33] In contrast, for positive temperature hydrogels, dehydration and shrinking occurs at temperatures lower than the upper critical solution temperature (UCST), where solvents are contracted and released from the matrices of the hydrogels with the formation of complex structures by hydrogen bonds. Other contributing variables for thermo-responsive hydrogels include ionic interactions, van der Waals interactions, system communications of entropy, and energy.[34]

The most widely studied poly(*N*-isopropylacrylamide) (PNIPAm)-based negative thermo-responsive hydrogels that are used in the aqueous phase show dramatic swelling properties at an LCST of 32 °C due to the subtle equilibrium between their hydrophilic and hydrophobic nature. Furthermore, for detection purposes at lower temperatures, the LCST can be decreased with the addition of cellulosic materials and plasmonic nanoparticles. For example, the PNIPAm hydrogels containing gold nanoparticles (AuNPs) proposed by Choe et al. achieved temperature detection through color changes between 25 and 45 °C with a resolution of 0.2 °C.[35] According to Li et al.,[25] PNIPAm−AEMA microparticles synthesized using PNIPAm hydrogels and 2-aminoethyl methacrylate (AEMA) hydrochlorides experienced phase transitions under solar irradiance and were evidenced to have an unprecedented infrared transmittance modulation of 75.6% and a high luminous transmittance of 87.2% at 25 °C (Figure 2d). However, the monomers and cross-linkers for the PNIPAm hydrogels and their derivatives were nonbiodegradable and nonbiocompatible, potentially leading to toxic, carcinogenic, and teratogenic impacts. As an alternative, Pluronic, a thermo-reversible hydrogel reported by Chatterjee et al. that undergoes sol−gel transformations, exhibited environmentally friendly properties and has been applied in food additives, medicinal compounds, and tissue engineering.[36]

Composite Hydrogel Sensors with Active or Responsive Substances. In addition to stimuli-responsive hydrogels with the collapse or extension of their polymer chains, hydrogels can also be adopted as inert matrices to accommodate active or responsive substances such as free ions, fluorescence/luminescence probes, biomolecules, antigens/antibodies, and living cells. These active substances are chemically associated with or physically entrapped in the passive hydrogel matrices and are responsive to matrix-permeable chemical (e.g., biomolecules)[24,37−40] or physical (e.g., light[41] and mechanical forces)[18,42,43] stimuli. For this sensing strategy, the volume and phase of these hydrogel matrices during the sensing process remain constant but have novel and desirable properties and constructions, including permeability, conductivity, and adhesive and mechanical properties.[8,44]

Active and responsive substances can be accommodated in various hydrogel-based living materials and devices to specifically interact with target biomolecules, giving various output responses. Liu et al.[37] covalently anchored polyacrylamide (PAAm) networks on passive hydrogel−elastomer

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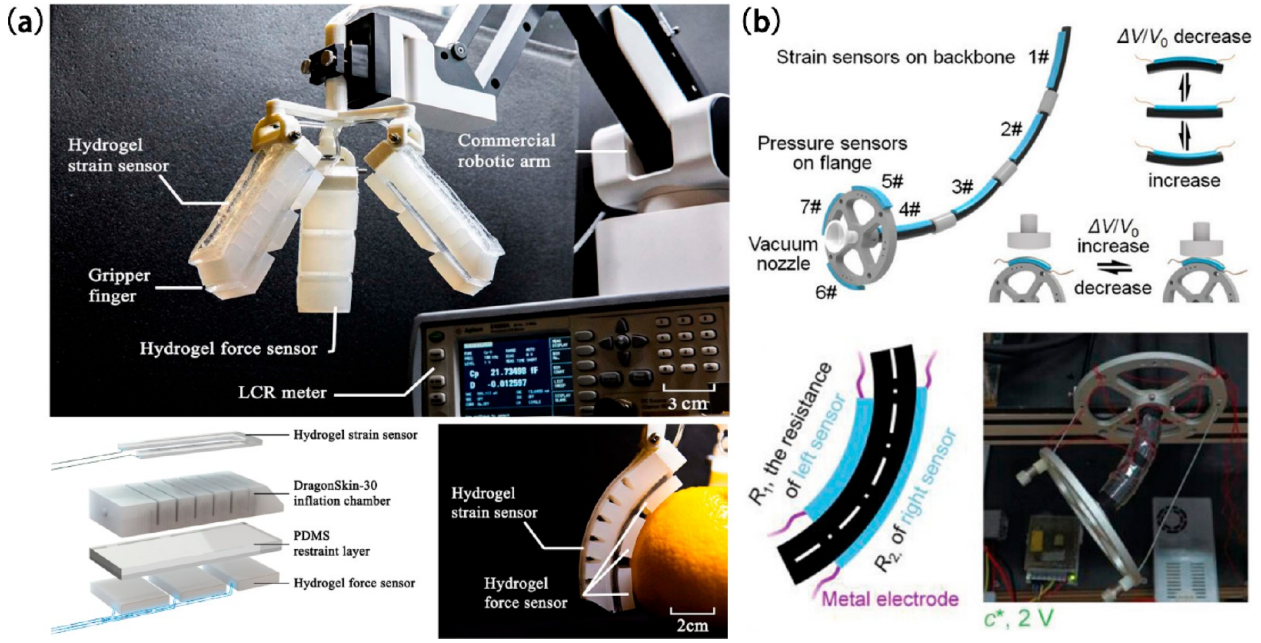

Figure 4. Intelligent identification. (a) Soft gripper with hydrogel-based force and strain sensors for object recognition and force-controlled grasping. Reproduced with permission from ref 47. Copyright 2022 Institute of Electrical and Electronics Engineers. (b) Skin-like hydrogel sensor-based sensing feedback system of a CDCR for motion identification. Reproduced with permission from ref 48. Copyright 2022 John Wiley & Sons.

matrices and used *Escherichia coli* bacterial strains to express the output green fluorescence proteins (GFPs) in response to the input cognate chemicals, giving optical responses. Furthermore, to achieve the preferred sensing selectivity and sensitivity, passive hydrogel matrices for living sensors were designed, allowing for the rapid penetration of water, nutrients, and chemical inputs to the bacteria while preventing bacteria leakage into the environment (Figure 3a). The research team also validated that the versatile living materials and devices could be fabricated as wearable patches, living gloves, or skin-adhesive tattoos[24] to detect chemicals on the skin, with anticipated detection of human sweat or blood in the future. Additionally, Choi et al.[38] reported that polyethylene glycol (PEG)-based hydrogels with incorporated fluorophores could be used as cell scaffolds during the sensing process. The living cells hosted in the hydrogel matrices were able to respond to the input environmental stimuli and produce the corresponding luminescence as output signals. Remarkably, the porous aqueous networks of the PEG-alginate-based matrices served as bidirectional optical communication channels for the encapsulated cells, allowing for the real-time interrogation of in vivo biomedical applications (Figure 3b). The researchers also validated the real-time cell-based toxicity sensing in live mice and optical sensing of blood oxygenation levels by cadmium-based quantum dots and absorbance spectroscopy, respectively.[39]

Even though such hydrogel matrices are supposed to have no autonomous volume change resulting from the variation of polymer chains or networks, they are frequently subjected to external disturbances and mechanical forces, leading to undesirable deformation. However, the deformation of the matrices can, in turn, be processed into useful information, indicating the level of external disturbances and mechanical forces. With mechanical forces as inputs and electrical signals as outputs, hydrogel matrices have surprisingly shown potential in the fields of strain and pressure sensing.[45,46] Kim et al.[18] presented an ionic hydrogel-based epidermal touch panel that was highly stretchable, biocompatible, and transparent. The hydrogel touch panel revealed negative linear correlations between the induced touching current and the interval between the corresponding electrode and the touch point, suitable for both the undeformed and stretched hydrogels. The data sets of the measured current flow in the ionically conductive hydrogels could be further processed with signal processing algorithms, yielding more straightforward and explicit information (Figure 3c). In addition, Pu et al.[42] demonstrated the tactile sensing capabilities of PAAm−LiCl hydrogels, which exibited transparency and an ultrahigh stretchability of 1160%. Recently, Lin et al.[43] fabricated dual-channel flexible strain sensors based on mechanofluorescent and conductive hydrogel matrices, taking mechanical forces as inputs and outputting electrical and fluorescent signals. Intriguingly, the high sensitivity and good linearity of the sensors demonstrate advancements in accurate human motion detection.

Photochromic hydrogels have gained substantial interest because of their prospective applications in optical devices. Guan et al.[41] introduced a hybrid PAAm-based hydrogel with rapid photochromic abilities and robust mechanical properties. Particularly, the novel photochromic hydrogel supplemented with ammonium molybdate (Mo) exhibited rapid discoloration within 3 s, a high fracture elongation of up to 3000%, and an increased fracture stress threshold of 130 kPa. The fading process and self-recovery capabilities of the hydrogel could be accomplished in an air environment. Moreover, the researchers highlighted potential developments in the field of flexible and elastic hydrogel-based devices for recording media for visual display by implementing rewritable and erasable optical information (Figure 3d).

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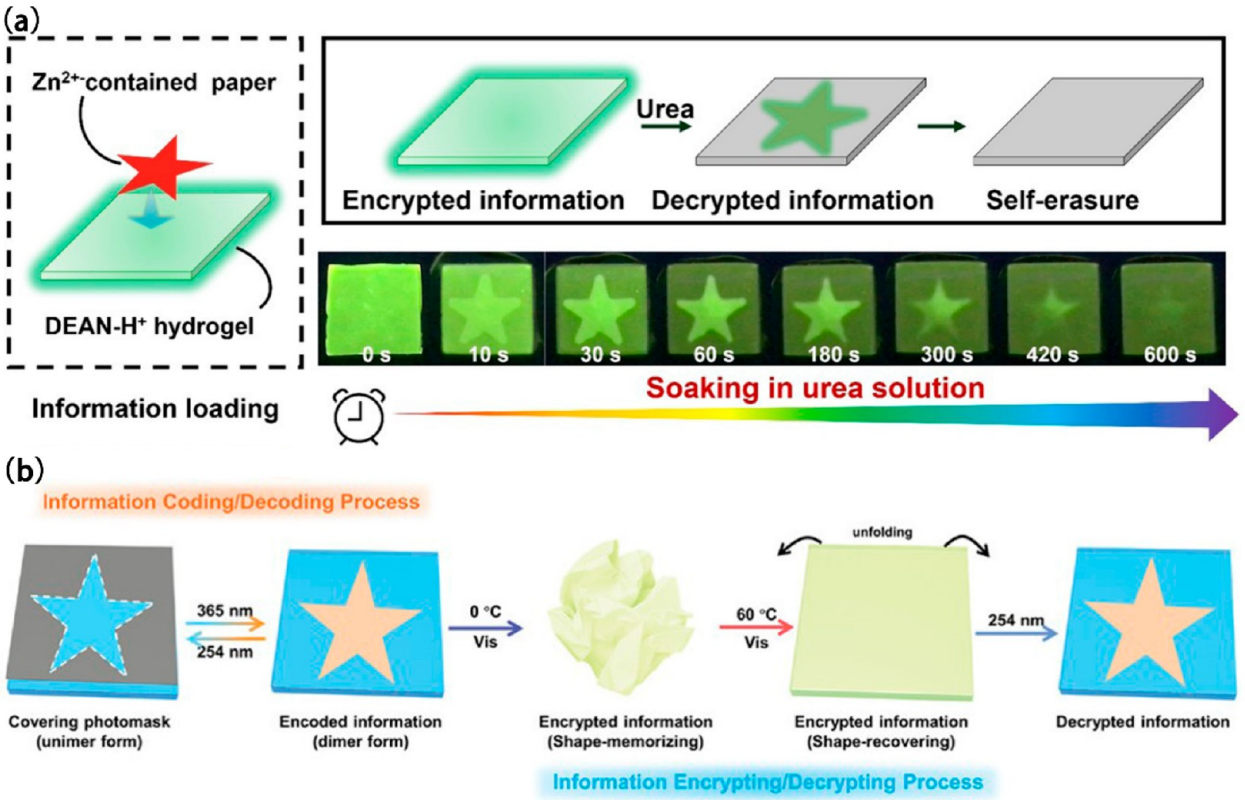

Figure 5. Information secrecy. (a) Information encrypted via H^+ and decoded through urea. Reproduced with permission from ref 51. Copyright 2020 John Wiley & Sons. (b) Information encrypted and decoded through wavelength changes of UV light and fluctuations of ambient temperatures. Reproduced with permission from ref 52. Copyright 2022 John Wiley & Sons.

APPLICATIONS OF HYDROGEL-BASED SENSORS IN HMI

With the emergence and evolution of artificial intelligence (AI) and the Internet of Things (IoT), hydrogel machines are expected to systematically merge the superior properties of hydrogels with the versatile functions of machines, including the sensing, actuating, communication, and visualization capabilities of a complete HMI system. Particularly, hydrogel-based sensors, which are superior in their softness, responsiveness, flexibility, stretchability, imperceptibility, biocompatibility, and self-healing abilities, provide new possibilities in different wearable application scenarios and have the promise of enhancing the user experience of HMI technology. In this section, four representative wearable hydrogel-based devices are described, along with their respective build strategies and specific examples.

Intelligent Identification. Intelligent identification technology has become one of the most prevalent and mature topics in the field of HMIs, ranging from object recognition tasks in the presence of image, haptic, or electrical changes to movement identification applications. Interestingly, intelligent identification has manifested success in providing individuals with unique transdisciplinary insights that enhance their understanding of the world, and the utilization of hydrogels has presented numerous benefits, including increased wearing comfort and improved user experiences with HMI identification devices.

Previous studies have demonstrated various applications in the realm of learning-based object recognition. For example, Wang et al.[49] presented a smart electronic skin (e-skin) system for wearable sensing, which was obtained by immersing a combination of strong, aligned cellulose scaffolds and weak, flexible polyvinyl alcohol (PVA)@MXene hydrogels in a glycerinum solution for solvent exchange. The smart e-skin system demonstrated the ability to identify different objects corresponding to distinct electrical signals. Intriguingly, the sensing response and recovery capability under 6% of the applied strain were measured to be approximately 120 and 90 ms, respectively, which are values that are suitable for rapid indentation and real-time monitoring. Similarly, Zhou et al.[47] presented a three-finger soft robot gripper integrated with hydrogel-based strain and tactile sensors. By employing a long short-term memory (LSTM) deep learning neural network that analyzed the data sets of the resistive and capacitive changes that were detected separately with the hydrogel strain and tactile sensors, object recognition could be achieved (Figure 4a).

In addition to object recognition, hydrogel-based sensors can also be used for real-time motion identification. We proposed a motion identification approach for a cable-driven continuum robot (CDCR) based on the perception of PAAm−alginate−NaCl composite hydrogels.[48] Specifically, the electrical resistances of the composite hydrogels showed a positive correlation with strain and a negative correlation with pressure, referring to the predictable voltage changes. With the sensing feedback from the four strain sensors and three pressure sensors, which were adhered to the four backbone segments and the rim of the terminal flange, respectively, the CDCR was able to estimate the bending directions and degrees and could perceive the surrounding space (Figure 4b). Furthermore, hydrogel-based sensors show amazing potential in the field of instant communication, having been used for the real-time identification for vocalization[21] and finger bending.[50]

Information Secrecy. Cybersecurity and information authentication have become a critical issue with the proliferation

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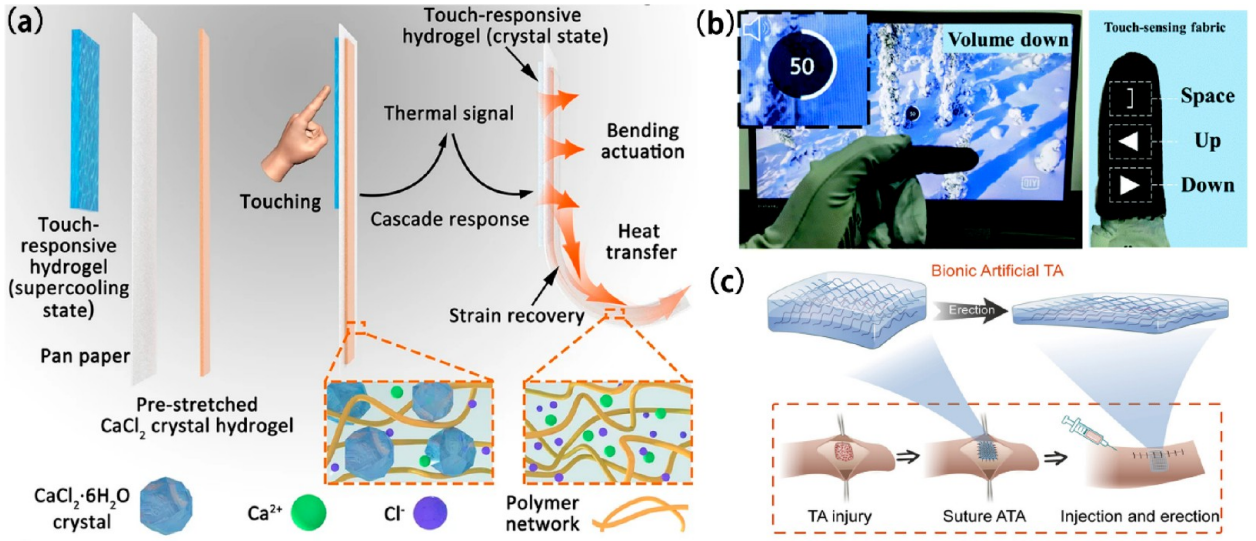

Figure 6. Interactive control. (a) Biomimetic flytrap-like actuator. Reproduced with permission from ref 54. Copyright 2022 Springer Nature. (b) Touch-sensing glove. Reproduced with permission from ref 53. Copyright 2021 Royal Society of Chemistry. (c) Bionic artificial tunica albuginea. Reproduced with permission from ref 55. Copyright 2023 Elsevier.

of IoT over the last two decades, and the public is becoming increasingly aware of the significance of information security in HMI systems. Surprisingly, the conventionally used keyboard- and touchscreen-based HMIs are considered to be susceptible to information leakage.

To address this issue, alternative HMI systems using innovative hydrogel-based devices with encryption capabilities have been intensively studied. Fluorescent hydrogels have emerged as the dominant focus of investigation in this research area. Our group presented an enzyme-involved and urease-exposed fluorescent hydrogel, which was copolymerized with acrylamide and the fluorescent monomer 4-(*N*,*N*-dimethylaminoethylene) amino-*N*-allyl-1,8-naphthalimide (DEAN).[51] The introduction of metal ions (e.g., Zn^{2+} and Al^{3+}) led to an increase in fluorescence, whereas the presence of NH_3, which was generated through urea decomposition, caused a subsequent decrease, endowing the hydrogel with programmable information decryption capabilities (Figure 5a). We also presented a fluorescent organohydrogel with dual-encryption information capabilities.[52] The organohydrogel was fabricated through the interpenetration of hydrophilic poly(*N*,*N*-dimethylacrylamide) (PDMA) polymer chains and hydrophobic poly(methacrylate stearate) (PSMA) polymer chains, which were composed of naphthalimide moieties (e.g., DEAN) for emitting yellow-emitting fluorescent units and photoswitchable anthracen-9-yl acrylates (9-ANA), respectively. The anthracene organohydrogel in this system demonstrated dual encryption with decodable fluorescent patterns and reconfigurable stereo structures under the stimulus of wavelength changes of ultraviolet (UV) light and fluctuations of ambient temperatures (Figure 5b).

To further improve the covertness of the information decryption process, we proposed another smart hydrogel for information authentication with dual imaging modes.[13] Benefiting from the inhibition effect of ethylene glycol (EG) on the crystal growth of sodium acetate trihydrate (SAT), information could be encrypted by incorporating EG patterns into supercooling SAT-filled hydrogels. The encrypted information could be further decrypted based on differences in the crystallinity and variances in the temperature maps influenced by spatial resolution, leading to identification by optical imaging technology in the light and infrared thermal imaging technology in the dark. Novel hydrogel-based HMI systems such as these provide profound potential for information secrecy applications.

Interactive Control. In addition to the physical or electrophysical output signals from sensors, decision algorithms and control commands should also be taken into consideration for a complete human−machine interaction system. Hydrogel-based systems have demonstrated tremendous success in the field of wearable and portable devices.

For example, Xu et al.[53] developed a smart touch-sensing glove encapsulated with PAAm−LiCl hydrogels for remote control. The touch-sensing area of the fabricated hydrogel was divided into three sections with different predefined functions, namely the interactive control of volume up, volume down, and pause, by identifying the corresponding functions inferred from the detected current amplitude. Additionally, real-time commands were intuitively visualized on a display screen with improved user experiences (Figure 6b).

Hydrogels can also be applied for various biomimetic purposes. We developed a flytrap-like hydrogel-based actuator based on biomimetic cascade response strategies.[54] Benefiting from the metastability of the supersaturated sodium acetate (NaAc) solution incorporated in the polymer matrix, the hydrogel was endowed with a new stimulus of touch, and this touch-responsive hydrogel exhibited startling responsive crystallization and excellent exothermal and electric behaviors. Interestingly, the heat generated by the touch-induced exothermal crystallization could be transferred to another temperature-responsive hydrogel, resulting in a phase transition and subsequent flytrap-like bending actuation (Figure 6a). Chai et al.[55] demonstrated a bionic artificial tunica albuginea (ATA) that incorporated isotropic PVA hydrogels with stretch-induced orientation, which was specifically designed for male patients with TA-damaged penile tissues to restore normal erection capabilities. The ATA, which had a bionic double-layer structure, exhibited TA-like strain hardening characteristics in both the axial and rail axes. Additionally, the possibility of ATA as a TA repair patch was demonstrated in the analogous in vitro

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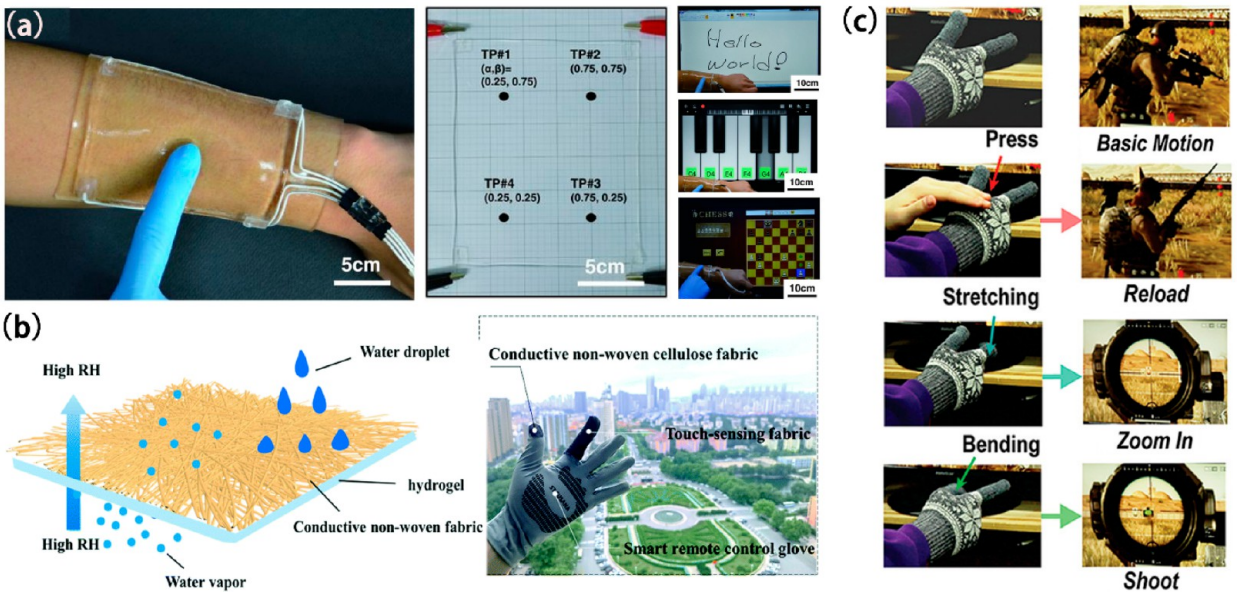

Figure 7. VR/AR-supplemented applications. (a) Smart touch panel for entertainment. Reproduced with permission from ref 18. Copyright 2016 American Association for the Advancement of Science. (b) Touch-sensing glove for volume adjustment and media control. Reproduced with permission from ref 53. Copyright 2021 Royal Society of Chemistry. (c) Smart glove designed for virtual shooting games. Reproduced with permission from ref 58. Copyright 2019 John Wiley & Sons.

porcine penis models (Figure 6c), providing future potential for implanted devices for humans. Other design concepts for human−machine cooperation[11] and human−brain collaboration[56] will be further investigated and developed.

Virtual Reality and Augmented Reality. While virtual reality (VR) immerses the user in a completely artificial world with substantial vividness and interactivity, augmented reality (AR) augments the real world with additional virtually generated information. Currently, the widely used commercial VR systems that adopt one of the five perception channels (i.e., auditory, haptic, taste, smell, and visual) with high quality have become inadequate. There is a growing demand for the simultaneous engagement of multiple perception channels despite the low individual feedback quality.[57] Both comprehensive and immersive VR and AR technologies with multisensory capacities in the field of human−machine interactions have the potential for social media, entertainment, rehabilitation, and recovery applications. Thus far, many portable and compatible wearable VR and AR devices made from hydrogels have been developed to enrich the senses of the virtual world, consequently optimizing user experiences.

As the first sensation acquired by humans in their prenatal development, haptics allows for the perception of self-sense and further plays a crucial role in the perception of the physical world and the social function of interpersonal bonding. Kim et al.[18] and Xu et al.[53] reported critical haptic human−machine interactions involving VR and AR technologies with the fabrication of a touch screen demonstrating real-time outputs. As depicted in Figure 7a,b, haptic-triggered input signals were utilized to control the VR interfaces for entertainment purposes and human-comprehensible visualization using hydrogels. In addition, as exhibited by the work by Choi et al., in which a conductive fiber-based glove capable of controlling the virtual interface with a simulated shooting game responded to real-time hand movements through a virtual game interface, VR interfaces can further be controlled with hydrogel-based gloves with analogous strain sensing mechanisms (Figure 7c).[58]

CONCLUSION AND FUTURE PROSPECTS

In this Perspective, we have highlighted the recent advances of hydrogel-based sensors in the domain of human−machine interaction. In addition to the widely recognized stimuli-responsive hydrogel-based sensors that experience spontaneous geometrical changes with the extension or collapse of polymer chains during the sensing operation, another conceptual idea of passive sensors related to hydrogels has been raised and developed, which utilizes hydrogels as inert matrices to host active and responsive substances. These passive hydrogel matrices have the ability to chemically associate with or physically encapsulate substances, which imparts them with properties such as permeability, conductivity, adhesiveness, and mechanical strength. Both categories of hydrogel-based sensors were evaluated in regard to their corresponding sensing mechanisms and potential applications. The diverse applications have demonstrated the feasibility of using hydrogel-based sensors for human−machine interactions with rapid responses and high accuracy for real-time intelligent identification, including object identification and motion detection. Also, fluorescent hydrogels have demonstrated a promising approach for information protection and anti-counterfeiting. Moreover, the interactive control applications of hydrogel-based wearable and portable devices have advantages over rigid electronic systems because of their intimate skin-adhered and biocompatible properties. VR/AR-assisted systems have also proven to be effective in improving user experiences during human−machine interactions.

However, despite the fact that numerous application scenarios have been investigated and the potential of hydrogel-based sensors in human−machine interactions has been demonstrated, there are still several challenges that need to be resolved before hydrogel-based systems can be produced commercially. The current challenges and future prospects are summarized as follows:

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1. The most crucial criterion of sensors is their sensing capabilities, and the development of novel hydrogels with exceptional properties represents a central need and a significant future trend in the field. Compared to well-developed, commercial, rigid electronic sensors, hydrogel-based sensors exhibit inferior performances in terms of sensitivity, accuracy, response time, and signal transmission efficiency. The slow sensing systems and long response times of fluorescent hydrogels, the extremely low electrical conductivity of conductive hydrogels, and the high light attenuation in hydrogel optical fibers strongly restrict the sensing performances of hydrogels.
2. Hydrogels always suffer from limited mechanical strength and toughness. A lack of durability and stability exists as another challenge for hydrogel-based sensors, in addition to poor repeatability, which is induced by the inherent senseing performances of hydrogels. The loss of water content over time due to evaporation, the degradation of the hydrogel networks due to prolonged exposure to harsh environments, and the poor adhesion between the hydrogel and the substrate are the leading causes of these issues. Improvement strategies include the modification of the cross-linking structures, the application of protective coatings, the addition of stabilizing agents, and the development of novel materials and fabrication methods. Moreover, an improved design of structures and selection of the materials of hydrogels should be further investigated with robust mechanical properties and comfortable user experiences.
3. Hydrogel-based sensors face challenges in sensing different stimuli/analytes and integrating with electronic devices. One significant challenge is the limited selectivity toward individual stimuli/analytes. Strategies such as incorporating specific recognition elements can help overcome this limitation. Another challenge is interfacing hydrogel-based sensors with traditional hard electronic components due to their soft and flexible nature. To enhance the capabilities of multifunctional hydrogel sensors, further developments should focus on integrating them into a comprehensive system.
4. The final and most inevitable step is the translation of laboratory discoveries into marketable products. Nevertheless, the fabrication and synthesis of hydrogel-based sensors are still complicated and lack industrial production machinery. A multidisciplinary approach that involves collaboration between researchers and industry partners can help overcome these challenges associated with fabrication difficulties while possibly reducing the cost.

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