

Review

Revolutionizing biosensing with superwettability: Designs, mechanisms, and applications



Zhong Feng Gao^a, Hai Zhu^b, Yanlei Li^a, Xiaochen Yang^a, Xiang Ren^a, Dan Wu^a, Hongmin Ma^a, Qin Wei^{a,c,*}, Fan Xia^{d,**}, Huangxian Ju^{a,e,*}

^a Key Laboratory of Interfacial Reaction & Sensing Analysis in Universities of Shandong, School of Chemistry and Chemical Engineering, University of Jinan, Nanxinzhuan West Road, Jinan 250022, PR China

^b Department of Civil Engineering, The University of Hong Kong, Pokfulam 999077, Hong Kong Special Administrative Region of China

^c Department of Chemistry, Sungkyunkwan University, Suwon 16419, Republic of Korea

^d Engineering Research Center of Nano-Geomaterials of Ministry of Education, Faculty of Materials Science and Chemistry, China University of Geosciences, 388 Lumo Road, Wuhan 430074, PR China

^e State Key Laboratory of Analytical Chemistry for Life Science, School of Chemistry and Chemical Engineering, Nanjing University, Nanjing 210023, PR China

ARTICLE INFO

Keywords:

Superwettable surface
Nanosensor
Nucleic acid
Single-cell trapping
Wearable bioelectronics

ABSTRACT

Interfacial superwettability and biosensors are constantly evolving and being recognized as ubiquitous components of interfacial science. Superwettability provides new insight from micro/nano scale to revolutionize the study of biosensors to improve the sensitivity, selectivity, accuracy, and practicality in clinical diagnosis and environmental monitoring. However, these superwettability-boosted biosensors require complex design and rigorous optimization of parameters due to the variation in surface topography and modulation of surface recognition. To reduce the confusion and barriers to entry in controlled tailoring of the interfacial superwettability of biosensors, this review presents an overview of the fundamental understanding of superwettability, and the classification of superwettability biosensors, which can be divided into homogeneous and heterogeneous superwettability biosensor. The biosensing mechanisms based on superwettability surfaces are reviewed in detail, including stimuli-responsive superwetting mechanism, droplet evaporation-enhanced enrichment mechanism, and liquid phase-regulated sensing mechanism. The emerging applications, such as nucleic acid analysis, immunoassay, single-cell trapping, bacteria-related study, and wearable electronics, are highlighted. The limitations and potential research viewpoints in superwettability biosensors are also discussed. By introducing the state-of-the-art of this important and rapidly expanding area, this review will motivate industrial circles and multidisciplinary scientific communities to create superwettability biosensors that can fulfil the demands of different practical applications.

Introduction

Biosensors have gained considerable attention as a promising platform for detecting biomolecules with high sensitivity and selectivity [1–5]. Recent advancements in electrochemistry, surface-enhanced Raman scattering (SERS), magnetic resonance, mass spectrometry, colorimetry, and fluorescence have led to an upsurge in their applications across various research areas, including biomedical diagnosis, environmental monitoring, and food safety [6–14]. However, the wetting properties of biosensors, such as hydrophobicity or hydrophilicity,

are often ignored, despite their significant impact on detection performance by regulating the interaction between the biological components and the target molecules. [15–19]. In this regard, superwettability has emerged as a promising approach to regulate biosensor performance. Superwetting surfaces exhibit extreme hydrophobicity or hydrophilicity [17,20], enabling them to interact with liquids in unique and controllable ways. Consequently, superwettability regulated biosensors have been quickly developed and exhibited enhanced sensitivity, selectivity, and stability, leading to the discovery of new applications across diverse fields.

* Corresponding author at: Key Laboratory of Interfacial Reaction & Sensing Analysis in Universities of Shandong, School of Chemistry and Chemical Engineering, University of Jinan, Nanxinzhuan West Road, Jinan 250022, PR China.

** Corresponding author.

E-mail addresses: sdjndxwq@163.com (Q. Wei), xiafan@cug.edu.cn (F. Xia), hxju@nju.edu.cn (H. Ju).

<https://doi.org/10.1016/j.nantod.2023.102008>

Received 12 July 2023; Received in revised form 12 September 2023; Accepted 25 September 2023

Available online 30 September 2023

1748-0132/© 2023 Elsevier Ltd. All rights reserved.

Nature stimulates human development and advancement, and learning from nature is a perennial theme [21–24]. One of common natural phenomena that have received considerable interest is special wetting behaviors [25,26]. Barthlott and his colleagues found that the self-cleaning and superhydrophobic properties of lotus leaves are attributed to their micro-structures and wax-made low surface energy material, which enable them to stay clean even in dirty situations [27]. Jiang and co-workers observed smaller structures, convexities on the nanoscale, on the micropapillae [28]. As a result, the presence of both micro/nano hierarchical structures and a waxy layer on the lotus leaf have been proven to be the reason of its superhydrophobicity, allowing water droplets to easily and randomly roll off the surface, thus removing impurities. This discovery has led to the study of several impressive organisms and plants that possess superwettability properties [29–31]. The lovely rose petals are also extremely water repellent. In contrast to lotus leaves, water droplets may be suspended on rose petal surfaces even upside down due to high adhesive pressures [32]. Besides bio-inspired superhydrophobicity, superamphiphobicity inspired by bacterial (*Bacillus subtilis*) biofilm colonies, pellicles, springtails, and leafhoppers, slippery inspired by *Nepenthes* pitcher plant, and patterned wettability inspired by Namib Desert beetles (*Stenocara sp.*) have all received a lot of attention recently [33–38]. Biomimetic superwettability materials, inspired by these natural organisms, have been developed in a variety of methods, including textural constructions and chemical alterations [21,39,40]. Meanwhile, various bioinspired applications based on wettability have emerged, which include antifouling, liquid separation, self-cleaning, anti-icing, energy harvesting, and antifogging [41–49]. Nevertheless, little attention has been dedicated to biosensing applications that govern surface superwettability, which also plays an essential role in the development of the superwettability biosensors.

The wettability of sensing interfaces is critical in biosensors and has significant effect on the sensitivity, stability, selectivity, and dynamic ranges. Compared with conventional methods, the merits of superwettability in the perspective of biosensing strategy include, but are not limited to: (1) expanding active sensing regions owing to the larger specific surface area-volume ratio, (2) precisely regulating locations and geometries of sample droplet, (3) allowing for simultaneous high-throughput detection of multiple targets, (4) expediting desorption of nonspecific molecule, (5) promoting the enrichment of signal probe or target to amplify the sensing signal, and (6) low droplet usage down to nanoliter level that facilitate the investigation of rare biological and clinical samples. These droplets may enclose a variety of analytes, including DNA, enzyme, proteins, cells, and microorganism. When fabricating biosensors for colorimetric, fluorescence, and electronic detection, the surface can be functionalized with the modifiers ranging from chemical molecules to nanoparticles for specific requirements. These superwettability biosensors integrate the characteristics of biochips with high throughput, allowing parallel conduct of hundreds of fabrications, screens, and experiments. Therefore, the chemical reaction rates and thermodynamics may be fine-tuned to improve biosensor performance by tuning the surface wettability.

Superwettability is a rapidly emerging field owing to its significance in resolving fundamental scientific challenges and commercial concerns. Superwettability surfaces have demonstrated exceptional droplet control and are appearing as valuable platforms in a wide range of applications, such as water/oil separations [50,51]. Several excellent review articles have extensively covered the surface fabrication strategies in detail [52, 53]. In terms of the type of materials, such as superhydrophobic surfaces, slippery liquid-infused porous surfaces, and wettability-patterned surfaces, there are several high-quality reviews summarized their biomedical applications, including biosensing as a section [54–56]. In terms of the type of biosensors, several excellent reviews about micro-patterned microchips, electrochemical biosensors, and microfluidics have been published [57–59], which verified the thriving development in this field. However, most of these reviews are focused on patterned wettability, which are heterogeneous superwettability surfaces. A

systematic review of the biosensor with homogeneous and heterogeneous wettability from the viewpoints of rational design, sensing mechanism, and application remains absent.

In this review, we first provide a brief overview of superwettability fundamentals, including superhydrophilicity, superhydrophobicity, superamphiphobicity, slippery superwettability, micropatterned superwettability, responsive superwettability, Janus superwettability, to help understand the role of wettability for constructing functional surface. Focusing on the homogeneous and heterogeneous wettability of sensing surfaces, the classification of superwettability biosensors is summarized. The biosensing mechanism for pursuing sensitive superwetting system from the perspectives of the stimuli-responsive superwetting mechanism, droplet evaporation-enhanced enrichment mechanism, and liquid phase-regulated sensing mechanism are critically analyzed. Subsequently, the emerging applications of superwettability biosensors including nucleic acid analysis, immunoassay, single-cell trapping, bacteria-related study, and wearable electronics are presented (Fig. 1). Finally, an outlook is offered concerning the challenges and opportunities accompanied with the basic research and industrial application prospects. The purpose of this review is not only to highlight progresses and challenges of biosensors associated with the existing superwetting techniques but also to advocate the promises and evaluate the opportunities of harnessing surface superwettability and manipulating droplet for next generation biosensing devices.

Fundamental definition of superwettability

The behavior of wettability is primarily determined by the chemical composition and surface structure of an interface. To estimate the wettability performance, static contact angle (CA) and contact angle hysteresis (CAH) are presented as measurement criteria to represent the static and dynamic wettability properties of the surface, respectively. Several old-fashioned and contemporary hypotheses have been proposed to explain the mechanics behind wettability during the research process.

The CA of water defines the surface wetting behavior [60–62]. Historically, the CA of 90° was regarded to be the threshold of hydrophilicity and hydrophobicity based on Young's equation, which is described

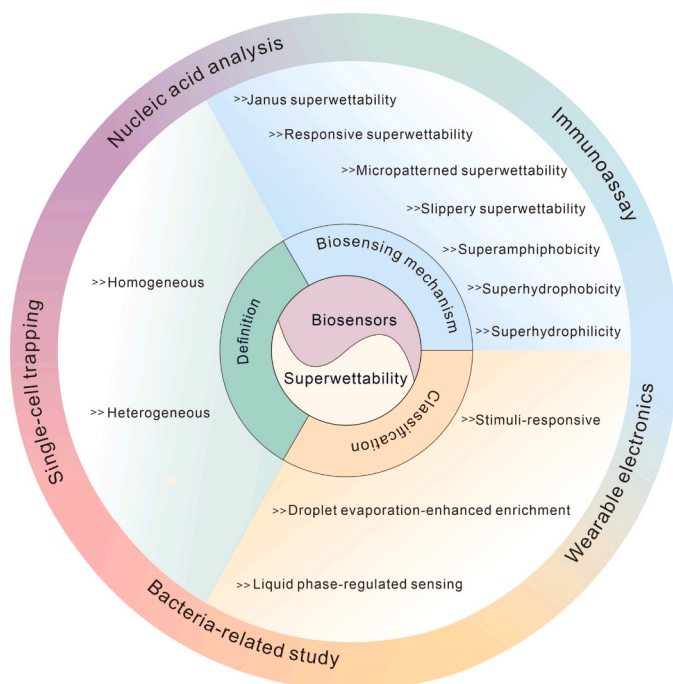


Fig. 1. Schematic illustration for superwettability-boosted biosensors.

below (Fig. 2a) [63]:

$$\cos\theta = \frac{\gamma_{SA} - \gamma_{WS}}{\gamma_{WA}} \quad (1)$$

where, γ_{WA} and γ_{SA} represent the interfacial tensions of a water against air and a solid against air, which are commonly referred to as surface tensions. γ_{WS} refers to the interfacial tension between solid and water. θ is the water CA on the surface. When θ is $< 90^\circ$, a hydrophilic surface is identified.

Hydrophobicity, on the other hand, is characterized by $\theta > 90^\circ$. However, an absolutely smooth surface is necessary for Young's equation, ignoring the chemical compositions and rough structures. Following that, Jiang and his colleagues suggested that a water contact angle of 65° could be used as a criterion for distinguishing between hydrophobicity and hydrophilicity (Fig. 2b) [64–66]. Materials with a CA ranging from 10° to 65° are considered hydrophilic. By increasing the roughness of the surface, a CA lower than 10° can be achieved, resulting in superhydrophilicity. Hydrophobicity is defined within a CA range of 65 – 150° . When the surface roughness is enhanced, the CA

increases to over 150° , which is referred to as superhydrophobicity.

Superhydrophilicity

To explore superhydrophilicity, nature provides plenty of specific cases. Researchers discovered that tears may spread and create a layer to protect human eyes [67]. The finding of superhydrophilic fish scales paved the way for bioinspired superhydrophilic investigation [68]. The unique micro/nanostructure structure reduces surface adhesion [69], leading to exceptional underwater self-cleaning ability. Similar to fish scales, shark skin also has significant superhydrophilicity [70]. Tree-frog toe secretion may moisten and stick to a wide range of surfaces due to the microstructure and superhydrophilicity [71]. These findings inspired researchers to exploit and fabricate innovative superhydrophilic surfaces for various applications including biosensing. The superhydrophilic character attributes to the antifouling performance [72–74], which can be used to minimize nonspecific adsorption on the sensing surface, including proteins, cells, enzymes, and other biomolecules.

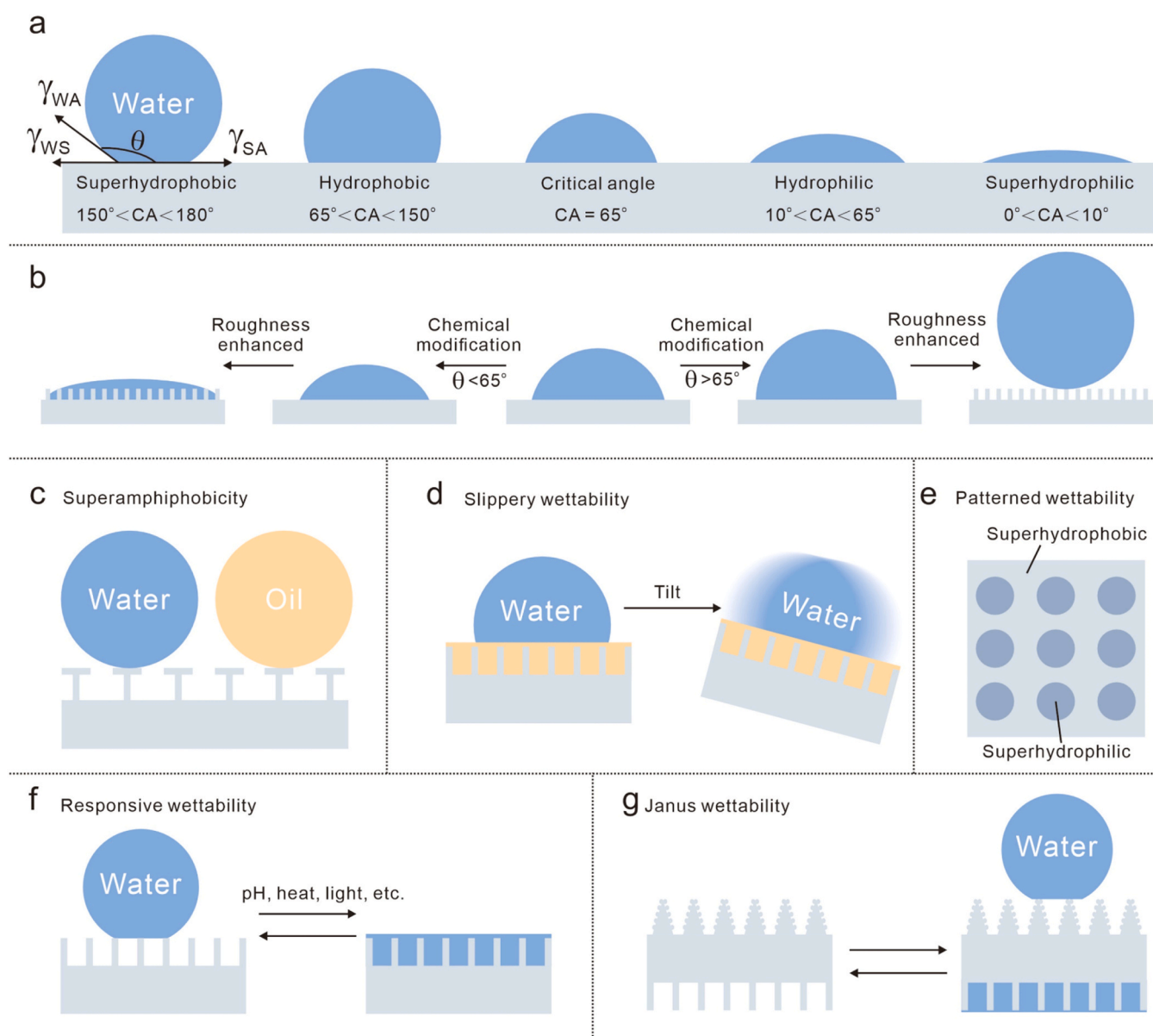


Fig. 2. Definition and classification of wettability. (a) Definition of several common wettability. (b) Rules for superwetable materials design. Special wettability, including (c) superamphiphobicity, (d) slippery wettability, (e) patterned wettability, (f) responsive wettability, and (g) Janus wettability.

Superhydrophobicity

Lotus was a representation of purity and magnificence in ancient Eastern societies due to its ability to self-clean in contaminated water [75–77]. A water droplet on the lotus surface has a high CA greater than 160° and readily rolls off the surface in any direction. Jiang and colleagues discovered that micro/nano-structures and the wax shell are critical for the self-cleaning superhydrophobic surface [28]. In contrast to the lotus leaf's highly water-repellent nature, rose petals are superhydrophobic and possess exceptional adhesion for water droplets [33]. The crimson rose petals have micro papillae arrays embedded in nano-scale wrinkles. When water droplets are placed on it, spherical drops are presented. Interestingly, water droplets have been found to adhere to these surfaces even when turned upside down. *Salvinia* is another unusual superhydrophobic plant identified. A unique technique to keep air for a prolonged period is provided by the *Salvinia* leaf. It is surprising that the terminal hair does not have wax crystals but produces hydrophilic patches, which scientists have revealed could stabilize the air layer and pin the air-water contact, a phenomenon called the "Salvinia effect". The finding of these distinctive creature surfaces has advanced our knowledge of superhydrophobicity. To explore wetting characterizations, five typical models of superhydrophobicity, including the Cassie state [78], Wenzel state [79], Wenzel-Cassie transition state [80], "gecko" state [81,82], and "lotus" state [83], have been established. The discovery of an inherent wet-ting process has improved theory and driven designs of artificial superhydrophobic surfaces. For the bio-sensing application, superhydrophobicity makes it difficult for the droplet to wet the sensing interface, which increases sensitivity by evaporating the sample droplet and specificity due to the antifouling feature.

Superamphiphobicity

Superamphiphobic materials have been extensively reported inspired by the oil-repelled creatures, such as springtails, leafhoppers, pellicles, and bacterial biofilm of *Bacillus subtilis* [33–37,84]. When the contact angles of water and low surface tension oils on a solid surface are above 150°, the phenomenon of superamphiphobicity occurs (Fig. 2c). The oil-repellent material requires lower surface energy and high roughness. Complex micro/nano-structures, such as T-like structures, mushroom-like structures, fibrous structures, matchstick-like structures, overhangs, and so on, are used to create roughness [85–89]. Meanwhile, fluorides are frequently chosen as chemical components to decrease surface energy. For the reactions involving non-oil phase, the role of superamphiphilic surface and superhydrophobic surface is almost the same as that in biosensing applications, which can effectively enrich analytes by evaporating sample droplets and remove contaminations.

Slippery superwettability

The *Nepenthes* pitcher plant has unusual slippery wetting characteristics [23]. Inspired from pitcher plant, slippery liquid-infused porous surfaces (SLIPS) are created by fixing lubricating lubricants on textured rough samples (Fig. 2d) [90]. Because of the immiscible solvents and low adhesion forces, water, polymers, insects, and paraffin can readily slide off SLIPS [91–93]. The fabrication of SLIPS requires three aspects: (1) the lubricating liquid may penetrate into, wet, and firmly stick the substrate; (2) the selected lubricating liquid entirely distributes over the substrate; and (3) the liquids being tested must not be able to mix with lubricating liquids. Slippery surface has significant self-clean and anti-fouling properties, which reduces the nonspecific interference on the sensing interface. It is also an excellent substrate for SERS biosensing due to its significant role for enriching analytes.

Micropatterned superwettability

In the Namib Desert, the *Stenocara* beetles have incredible hydrophilic–hydrophobic patterned surfaces, which collect foggy water micro-droplets from their back to feed themselves [38]. Because of the heterogeneous surface energy, water drops can be moved from hydrophobic regions to hydrophilic regions in this unique wetting. The superhydrophilic dots on a superhydrophobic substrate, known as superwettable micropatterns (Fig. 2e), can impressively hold micro-droplets due to their strong anchoring ability [57,58,94–96]. Patterned superwettability indicates two states of superhydrophilicity and superhydrophobicity in properly structured 2D arrays. Patterned superwettability that are sometimes used to construct superwettable micropatterns. The targeted droplets can be anchored and enriched on superhydrophilic microwells surrounded by superhydrophobic regions and then treated, recognized, and detected in following procedures. These micropatterns have demonstrated exceptional capabilities in regulating and patterning microdroplets, and are evolving as powerful platforms to cooperate with SERS, fluorescence, colorimetric, and electrochemical detections for biosensing and disease diagnosis.

Responsive superwettability

Responsive superwettability can be regulated by designing the stimulate surface, switching between superhydrophobicity and superhydrophilicity (Fig. 2f). External stimuli are classified as pH [96–98], gas [99,100], magnetic field [101–103], temperature [104–106], light [107–110], electric field [111,112], and so on. For example, photofunctionalization-based titanium biomaterial is a kind of typical light-responsive material [113–116]. Due to molecular water adsorption and hydroxyl groups, fluorinated TiO₂ nanoparticles exhibit superhydrophilicity after extended UV exposure [117–119]. In addition, the superhydrophobicity remains stable when shielded from light or kept in darkness, as the hydroxyl free radicals remove surface-absorbed water. Consequently, when the storage in darkness and exposure to light are repeated, the reversible switching between superhydrophilicity and superhydrophobicity occurs rapidly. Responsive superwetting behaviors, including contact angle, rolling angle, adhesive force, droplet transportation, etc., provide a straightforward approach for the visual biosensing. Semi-quantitative detection can be achieved using our naked eye, which is simply and user-friendly.

Janus superwettability

The non-wetting top surface and wetting reverse surface of lotus leaves make them difficult to blow over, even under windy conditions [120,121]. The superhydrophilic surface produces a high capillary force, enabling that lotus leaves stick securely to water. Generally, Janus superwettability exhibits two types of wetting behaviors: superhydrophilic/omniphobic, (super)hydrophilicity/ (super)hydrophobicity, and patterned slippery/wetting (Fig. 2g) [122–125]. Janus materials, with unique superwettability, present an ideal substrate for creating wearable biosensors that can monitor personalized health in a non-invasive way [126,127]. Janus wetting allows for efficient and thorough transportation of sweat from the skin to the sensing interface, ensuring comfort even during an outbreak. Using Janus superwettable materials, sweat may be fully and unidirectionally transported from the skin to the sensing interface providing great comfort.

Classification of superwettable biosensing interface

Superwettable biosensors have been demonstrated as an effective tool for the selective and sensitive detection of a wide range of biochemical targets with environmental and clinical implications. Based on the specified biorecognition components aimed at diverse targets, several superwettable biosensing interfaces have been developed. To

represent current progress in superwetable biosensors, this section focuses on two major elements of superwetable biosensing interfaces with homogeneous or heterogeneous wettability.

Homogeneous interface

The principles used in designing superwetable biosensors is highly related to interfacial biosensors of varying dimensions, including 0D nanoparticles, 1D channels, 2D surfaces, and 3D membranes. Consequently, it is possible to create multiscale functional superwetable biosensors by incorporating interfacial materials with different dimensionalities.

0D nanoparticles

The absorption of nanoparticles at liquid/liquid interfaces can provide effective stabilization against disproportion and coalescence, particularly when the nanoparticles with homogeneous hydrophilicity or hydrophobicity. Such nanoparticle-stabilized liquid/liquid systems offer potential as templates for the preparation of diverse wetting materials, including bulk structures and liquid marbles [128]. Interfacial wetting behaviour of nanoparticles, as indicated by the CA measured through the water phase, is a key parameter influencing the sensing performance (Fig. 3a). For air-liquid system, relatively hydrophilic nanoparticles are preferred for stabilizing air-in-water system, while relatively hydrophobic nanoparticles are suited for water-in-air system. This concept extends to solid-nanoparticle-stabilized oil-liquid dispersed systems, where relatively hydrophilic or hydrophobic nano-particles are employed to achieve oil-in-water or water-in-oil materials, respectively.

Generally, the homogenous wettability of nanoparticle can be controlled by surface chemistry and roughness. In principle, metal oxide, ceramic, and metallic nanoparticles are completely wettable by most liquids due to their high surface free energies while liquids hardly wet nanoparticles. Generally, the homogenous wettability of nanoparticle can be controlled by surface chemistry and roughness. In principle, metal oxide, ceramic, and metallic nanoparticles are completely wettable by most liquids due to their high surface free energies while liquids hardly wet nanoparticles with low surface free energies, such as polymeric and carbon nanoparticles [128]. By selectively modifying hydrophobic or amphiphilic chemicals on the surface of nanoparticles, their CA at liquid/liquid interface may be altered from 0° to 180° . This method is applicable to a broad range of nanoparticles, including

polymers, metals, and inorganic oxides [129–131]. Liquid marble, a unique non-wetting and non-adhesive material, may be created by coating liquid droplets with a hydrophobic powder layer [132]. By including stimulus-responsive nanoparticles in the outer shell, liquid marbles that responsive to external stimuli including light, pH, temperature, an electric or magnetic field may be constructed [133–135]. Liquid marbles have been reported for a variety of applications such as reactors, gas detection, catalysis, and controlled medication release [136–138]. It also provides a promising platform for blood-typing assays and sensing chemical molecules [139–141].

Apart from chemical modification, an alternate technique for creating sensing interface between a solid particle and liquid is surface structure, which results in distinct wetting behavior. Generally, hydrophilic nanoparticles in oil and hydrophobic nanoparticles in water can aggregate. Nonetheless, a type of particle known as 'hedgehog', which comprises both micro- and nanostructural features, displays consistent colloidal stability in both hydrophobic and hydrophilic solvents [142]. The surface of the nanoparticles contains nanoscale spikes which significantly reduce the contact area and attractive forces between adjacent particles. Specifically, when dispersed in water, the hydrophobic hedge-hog particles tend to exhibit a Cassie-Baxter wetting mode, as evidenced by the formation of an air shell surrounding the microsphere core. Particle dispersion behaviour can be customized leveraging either micro- or nanostructures to develop efficient nano-sensors capable of operating in complex liquid environments.

1D channels

Micro/nanoscale channels are an important type of 1D material that are instrumental in the advancement of biosensing interfaces. Channels with special wettability enables selective and directional transportation of water, indicating a potential distance-based visual sensing method (Fig. 3b). The capillary force is the key factor to drive a droplet transport on a non-sticking surface. The movement of a droplet is due to an energy transfer that overcomes the energy barrier of the non-sticking surface through the application of external stimuli such as light, chemical reaction, vibration, heat, magnetic force, or electricity. The distance the liquid travels depends on the energy supplied to the droplet and the rate of energy dissipation during its motion. To create a sensing interface, a responsive material (substrate or droplet) is necessary to convert inputs into energy that drives the droplet through gravity, viscous flow, and capillary force. Once the driving forces on the surfaces are greater than

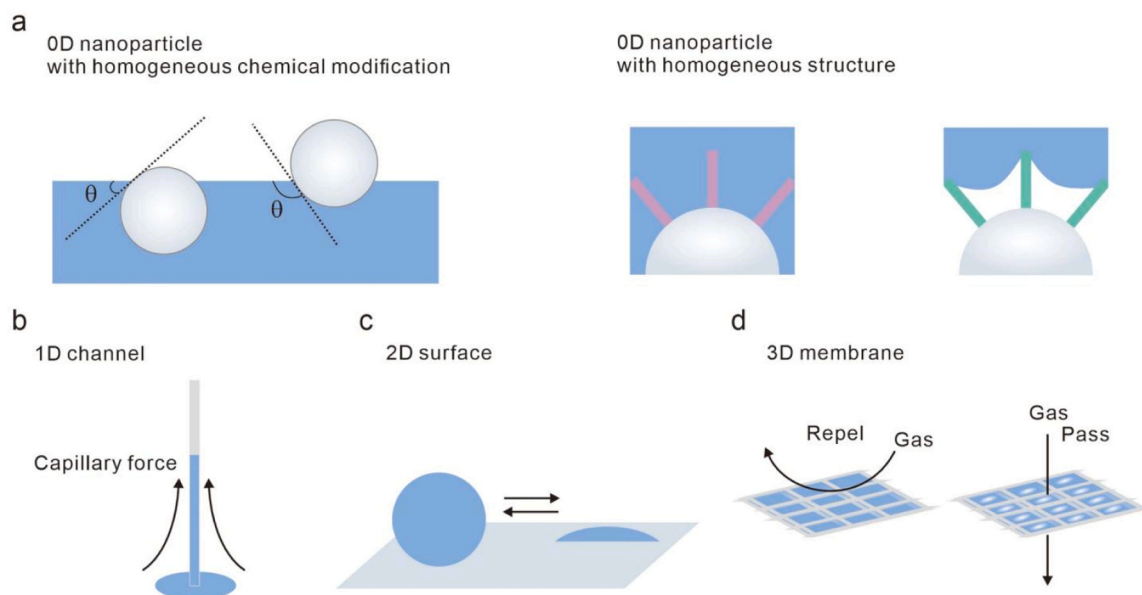


Fig. 3. Homogeneous superwetable systems with different dimensionalities, including (a) 0D nanoparticle, (b) 1D channel, (c) 2D surface, and (d) 3D membrane.

the frictional forces, the droplet initiates its motion. After the energy is dissipated, the drop-let motion comes to a halt. Capillary tube is a commonly used sensing device with homogenous hydrophilic property. The formation and degradation of a responsive DNA hydrogel will result in a varied liquid flow distance. The visible signal is proportional to the analyte concentration. Responsive hydrogel is a star material for investigation. A method for quantitative detection was developed by Yang and colleagues using a distance-based approach [143]. This method involves combining a target-responsive hydrogel that is encapsulated with Au core/Pt shell nanoparticles (Au@PtNP) with a volumetric bar-chart chip (V-Chip). When the target is introduced, the hydrogel dissolves quickly and releases Au@PtNPs which results in a substantial amount of oxygen being produced via the breakdown of hydro-gen peroxide. This oxygen is then used to move an ink bar in the V-Chip. Recording the distance of ink bar allows the user to visually quantify the concentration of the target in the sample. This method is universal for various target, such as cocaine, lead ion, adenosine, and Ochratoxin A [144–147].

Using two nearly parallel plates with homogeneous hydrophilicity, Taylor rising, a well-known capillary behavior, can be formed. It demonstrated better sensitivity for liquid transportation compared with the capillary tube [148]. Taylor rising-based sensing devices has been seldom developed, which might be a promising filed for developing point-of-care devices. Embedding surfactants in hydrogels is also a feasible strategy. Trypsin can degrade gelatin-based hydrogel to release surfactants and reduce the surface tension of the solution [149]. The low surface tension hinders the bouncing and sliding performance of liquid droplets on superhydrophobic substrates. Based on the surface tension, rolling angle, contact angle, and bouncing performance, trypsin and its inhibitor can be visually detected with various dynamic ranges. The input of matter and output of wetting signals can be used to construct Boolean logic trees and perform information encryption [150,151]. Moreover, a recent partnership of researchers concentrating on the usage of one-dimensional ion channels and nanopores has significantly advanced this area. Since the properties and applications of ion channels and nanopore have been well-described in detail by several excellent reviews [152–155], thus they are not be discussed here.

2D surfaces

This section involves surfaces with uniform surface structure, energy, or chemistry, which is simple to design and easy to use. Superhydrophobic surface is the most widely used sensing substrate with micro/nano structures and high roughness (Fig. 3c). The micro/nano structure on these surfaces prevents droplets from penetrating, leading to a decreased contact area between the droplet and surface. However, the droplet appears spherical with a high CA exceeding 150° , which allows it to roll off easily with minimal critical rolling angle and contact angle hysteresis, thereby enhancing visual performance. Superhydrophobic surfaces are commonly used for the development of superwetable biosensors for the determination of diverse targets such as nucleic acid, proteins, ions, and cells [156–160]. On a superhydrophobic surface, the droplet contact area can be dramatically in a tiny spot greatly improving sensitivity [58,161]. Superoleophobic interface indicates that oil forms a spherical shape and slides off the surface in both air and water [162,163]. The special wettability is realized under the condition that the droplets and the liquid surrounding them are incapable of being mixed. The advantage of this approach is the possible regulation of surface wettability in inert liquids, which is preferable for the extraction and detection of target from oil [164–166]. However, the biological sensing system seldom involves oil phase. The superoleophobic surface-based sensing method is promising for the detection of chemicals and the detection of biomolecule remains to be explored. SLIPS provides another well-performed sensing interface due to its self-cleaning and self-healing properties [167,168]. The lubricant is capable of promptly wetting the surface and spreading out to recover its original lubricating ability, even if the porous structure is impaired. The

low surface energy of the SLIPS surface makes it highly resistant to biofouling, reducing the likelihood of false positives or reduced sensitivity due to fouling. However, SLIPS-based biosensors require multiple steps to prepare the surface, which can make it difficult to scale up production or manufacture at a low cost. Limited analyte compatibility is also an impact factor that restrain its application. The SLIPS coating may not be compatible with all analytes due to its lubricant layer, and its hydrophobic properties may limit the detection of certain biomolecules. Homogeneous wettability enables well-performed responsive super-wetting switch and droplet evaporation-enhanced enrichment, which significantly improved the sensitivity for biosensing.

3D membranes

Porous membranes, serve as classic 3D materials, have been extensively explored and used in various fields including catalysis, water treatment, food production, chemical separation, and energy harvesting. Recently, liquid-based porous membrane (LPM) has shown great promise in sensing as an alternative to traditional membranes due to its desirable attributes including antifouling, omniphobic, dynamic, diffusive, self-healing, and adaptive properties (Fig. 3d) [169–172]. Imbuing a functional liquid into a permeable membrane is necessary to create LPM, which can stabilize the structure through capillary force. This membrane consists of two critical constituents: the infused liquid and the porous matrix. They work together synergistically to active interfacial functions exhibit a wide range of dynamics. The infused liquids, including water, oil, liquid metals, ionic liquids, and other responsive liquids, have been extensively used while the porous matrix usually prepared using biological materials, inorganic materials, organic materials, or composite materials. Transport mechanisms across LPM can be broadly categorized into physically-driven, chemically-driven, and physicochemically-driven mechanisms. The functional liquid in a liquid-based porous membrane may undergo chemical diffusion, which can impact the gating threshold pressure when transporting fluids due to changes in its chemical properties. A new chemical detection mechanism based on visual liquid gating has been developed through the infusion of water into a porous matrix [173,174]. By incorporating surfactants into the functional liquid, the threshold pressure of transport gas is lowered. Moreover, diffusion of the target cation results in further reduction of the threshold pressure as the surfactant rearranges itself at the liquid/gas interface. The distinct movements of marker droplets released due to gas production indicate the presence of various cation types. Improvements in the detection system's sensitivity can be achieved through pore size refinement and the anchoring of specific molecules.

Heterogeneous interface

Controllable droplet manipulation is crucial for biosensing. Controlling droplets is achievable by applying driving forces to them, and at the same time, avoiding them from sticking to the surface they come in contact with. This requirement is fulfilled by creating sensing interfaces with heterogeneous wettability, which has a significant impact on sensing performance. There are two main types of reported techniques for heterogeneous wettability: gradient and anisotropic super-wettability. Here, we elaborate on the concepts and design methodologies by outlining previous efforts.

Gradient surface topography/chemistry

Based on gradient surface structure, energy, or chemistry, the droplet movement can be accurately controlled [175], indicating a straightforward and simple visual sensing method. When an energy gradient is applied to the droplet contact interface (Fig. 4a), it spontaneously flows towards the lower energy. Thus, the droplet is propelled along the direction of the energy gradient through capillary forces. To identify droplets that exhibit different surface tensions, a simple sensing device has been suggested, which has precisely calibrated discrete surface

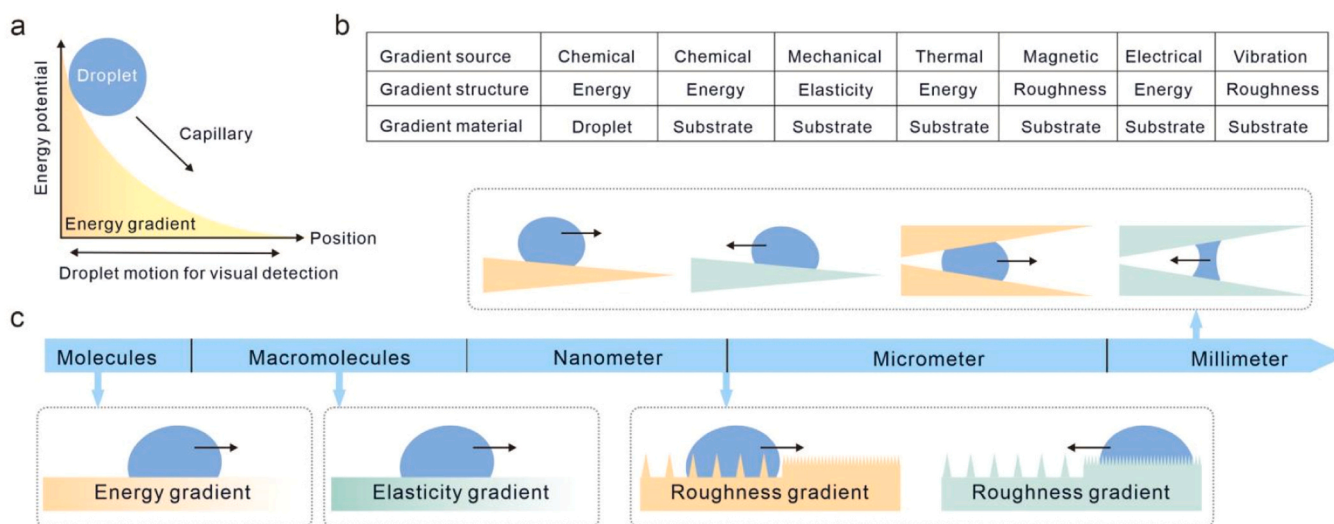


Fig. 4. Rational design of gradient superwetable surface topography/chemistry. (a) The energy potential on gradient superwetable surface. (b) Composition of gradient superwettability. (c) Gradient superwetable surfaces in various scales.

energy domains [176,177]. When the target changes the surface tension, the associated transportation distance can be employed for visual detection. Apart from manipulation of surface gradient, external stimuli with spatial gradient intensity, such as light, heat, magnetic, and electrical, are also essential sources to create droplet or structure gradients (Fig. 4b) [178–181]. When subjected to stimuli, the droplet, sensing interface, or field reacts by creating a gradient structure and altering its surface energy. Different from the homogenous superwetable surface, the pre-established gradient moves the droplet a specific distance without external influence, which is favorable in precise biosensing.

Gradient superwetable surfaces can be rational designed with different length scales ranging from molecular to macroscopic scale (Fig. 4c). The classic molecular-scale design is surface energy gradient. The magnetic/electrical interactions and surface chemical composition, involving electron, spin, and molecular modification, have great influence on this wettability. A differential in surface tension leads droplets to spontaneously migrate from non-wettable to wettable region. Introducing elasticity gradient is a macromolecular level design, including the cross-linking degree of polymer networks. Fabricating sensing substrate using polymer networks with different cross-linking degree and elastic modulus, an interfacial tension gradient can be formed from the flexible to hard sides. If a droplet is placed on a substrate with a Young's modulus that is equal to or lower than the interfacial tension, the tension will alter the interface and lead to a change in the direction of the surface tension. The sensing substrate designed with gradient wettability provides the sample-harvesting mechanism, which can collect and detect trace target for various applications, such as wearable biosensing in sweat and visual biosensing in clinical samples. For example, Zhang et al. developed a multichannel superwetable microspine (SMS) biosensor with significant sample droplet-collecting behaviors [182]. By combining surface superhydrophilicity and geometric asymmetry, the SMS device enabled directional and spontaneous droplet transportation. The asymmetrical geometric design of the SMS sensor creates a Laplace pressure gradient that controls the direction of droplet transportation. The nanomaterial-based microspine provides significant superhydrophilicity, which also plays an essential role for droplet self-transportation. The multichannel SMS sensor achieved sensitive detection of blood prostate-specific antigen (PSA) with a low limit of detection (LOD) of 1 pg/mL, indicating an efficient and sensitive strategy for clinical applications. The development of gradient superwetable biosensors is still in its infancy stage, and we expect researchers could pay more attention to this area of research for multiplex biosensing and clinical detections.

Anisotropic surface topography/chemistry

Numerous plant leaf surfaces in nature feature directional groove patterns with special wettability, such as rice leaves [183–185]. Inspired by this structure, Anisotropic tuning of the energy potential is achieved through the utilization of a surface that has undergone asymmetric chemical modification and morphology (Fig. 5a) [186]. Applying external forces such as capillary, pressure, or gravity to the system, the flow direction of the droplet is towards the area with lesser potential energy [185,187–189]. Typical rules for designing anisotropic sensing interfaces are summarized in Fig. 5b. As a result, programmable droplet mobility is conceivable. This potential acts as a stumbling block for the droplet migration. According to the distance that droplet moved, the target concentration can be determined simply by naked eye. The design of anisotropic superwetable surface covering from molecular to millimeter scale is shown in Fig. 5c.

At the molecular scale, the surface heterogeneously modified with hydrophobic or hydrophilic chemicals results in the anisotropic surface energy. When a droplet on a hydrophobic region flows across a hydrophilic region, the droplet flows to the hydrophilic region spontaneously. This is attributed to the hydrophilic region having a much lower energy potential than the hydrophobic region. As a droplet goes from the hydrophilic region to the hydrophobic region, a high driving energy is required for droplet transportation. At the macromolecular level, surface viscoelasticity is critical in droplet mobility. Lubricating the surface with low-viscous liquids, such as fluorinated oil, organic alkane, or silicone oil, the slippery surface allows immiscible droplets to slide off easily with an ultralow contact angle hysteresis. Nevertheless, the droplet mobility is limited on the surface infused with cross-linked or viscous lubricant. Thus, a high energy is required to drive the transportation of liquid droplets. As lubricant viscosity increases, the energy potential required to move droplets also increases. To adjust droplet mobility from nanometer to micrometer scale, introducing roughness to the surface is a powerful technique. The increase of potential energy contributes to the anchor effect on the rough surface, pinning the droplet and limiting its motion. At the Cassie state, droplet cannot wet the rough surface with small contact area, weak interfacial adhesion, and low rolling angle. On the contrary, the droplet fully invades the rough surface and tightly adheres to it at Wenzel state. By introducing external stimuli, such as heating, light, pressure, or chemicals, to the droplet, the transition between Cassie and Wenzel state can be achieved. The pinning effect inhibits droplet mobility at the micrometer to millimeter scale. When a droplet contacts with the inclined surface, the CA ($\theta + \alpha$) is crucial for droplet transportation. Only in the case that the CA of

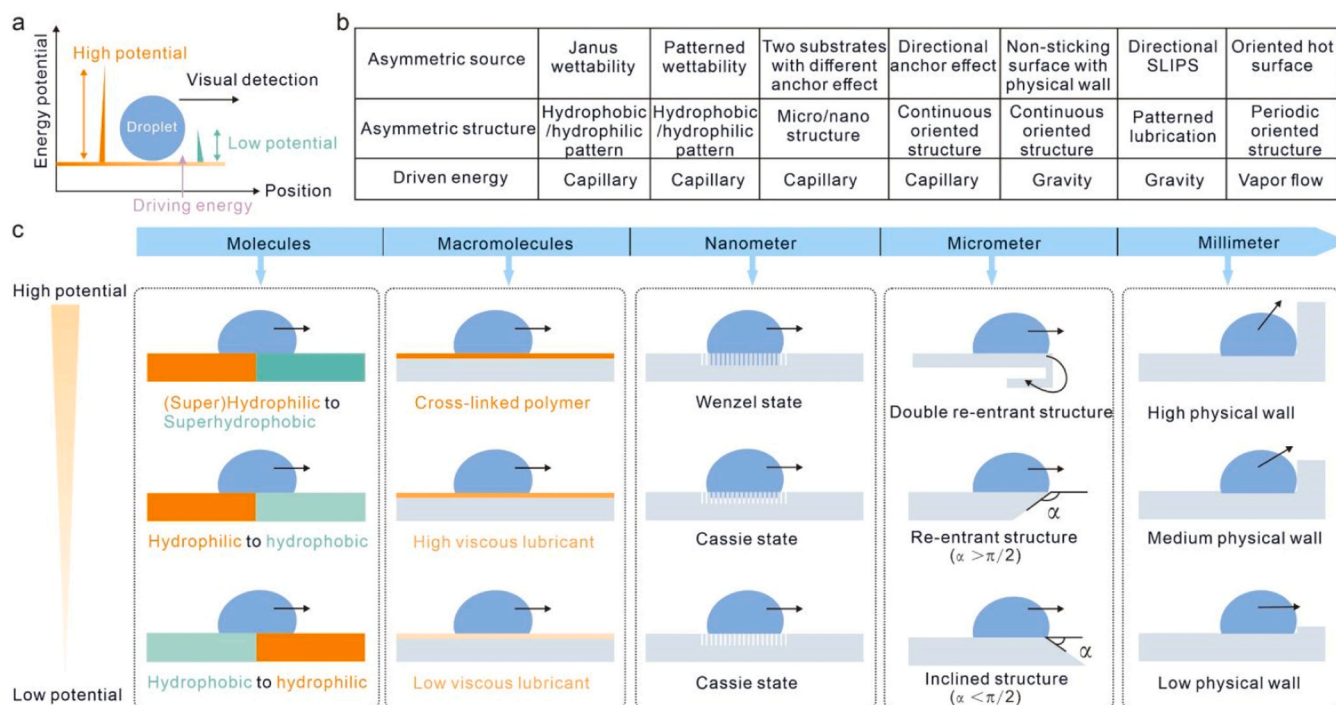


Fig. 5. Rational design of anisotropic superwettable surface topography/chemistry. (a) The energy potential on anisotropic superwettable surface. (b) Composition of anisotropic superwettability. (c) Anisotropic superwettable surfaces in various scales.

droplet exceeds $\theta + \alpha$ enables the droplet transportation. The energy potential increased with increasing the α value. When the α is higher than $\pi/2$, it possesses high energy and prevents droplet from spreading, which is defined as re-entrant structure. To further improve the water-repellent performance, the double re-entrant structure is developed with superamphiphobicity. At millimeter-scale, the gravity-restricted potential energy is the key element that needs to be explored, which is essentially governed by the macroscopic physical grooves. Due to its anisotropic wetting property, liquid droplets are more prone to rolling off in the direction parallel to the grooves. Based on the droplet transportation on anisotropic superwettable surface, diverse visual biosensing methods can be developed theoretically.

Up to now, few examples have been reported, leaving a huge space for researchers to explore. Anisotropic SLIPS display significant sliding capabilities for droplets with varying concentration of potassium chloride [190]. The provision of voltage increases the electrostatic attraction between the conductive droplet and the substrate, leading to a transitional phase between the Cassie and Wenzel models. The critical sliding angle rose as the adhesion force increased, which is proportional to the concentration of potassium chloride. We recently provided an orthogonal dual-regulation technique for accurate droplet motion control, which serves as a sensitive sensing strategy with tunable dynamic sensing ranges for various targets detection, including DNA, ATP, thrombin, miRNA, and kanamycin [191]. DNA involved droplet was moving over a micro-grooved SLIPS to control resistance from the liquid and solid phases individually. The periodic micro-grooves dictated the energy barrier from the solid surface while the hydrophobic interaction between DNA and lubricant dominated the resistance from the liquid phase. This orthogonal dual-regulation technique revealed the capacity to accurately regulate the motion behaviors of droplets as well as sensitive detection with tunable dynamic ranges for a variety of targets. The anisotropic sensing interface holds great potential for superwettable biosensors and beyond.

Fundamental biosensing mechanism by superwettability

As for the biosensing mechanism in the design of sensitive and selective superwettable biosensor, three primary types have been commonly used: (1) stimuli-responsive mechanism. It involves the bio-recognition events to alter the droplet wetting behavior for visual signaling, such as contact angle, rolling/sliding angle, droplet transportation, etc.; (2) droplet evaporation-enhanced enrichment mechanism. It can be used to increase the relevant concentration of signal probe, thus serving as a novel signal amplification method to improve the sensitivity. This method is open and universal to various conventional signals including electrochemical, fluorescent, colorimetric, SERS, etc.; and (3) liquid phase-regulated sensing mechanism. To rationally design biochemical reactions to alter the surface tension or viscosity of droplet is an important way to trigger the superwetting performance for biosensing.

Stimuli-responsive mechanism

Stimuli-responsive superwettability plays an important role in constructing a sensitive biosensor. Stimuli-responsive superwettability that switches between nonwetting state and wetting state under external sources has attracted considerable interest [91,192,193]. By using external stimuli, including electrical field, magnetic field, heat, light, pH, or target molecules (Fig. 6a), wettability and motion performances of sample droplets can be timely and reversibly regulated, providing insightful and innovative sensing mechanisms for preparing naked eye-based visual biosensors (Fig. 6b). The core mechanism is the stimuli-responsive superwetting switch between Cassie state and Wenzel state (Fig. 6c). Typical examples include the pH-responsive surface, which presents superhydrophilic character (CA approaches 0°) at acidic conditions and superhydrophobic character (CA $> 150^\circ$) at alkaline environments [194]. The CA alteration serves as a signal for output, which can be used for quantitative detection of pH. Combining with glucose oxidase-catalyzed reactions, it can be extended to detect glucose and other targets, such as PSA [195]. The host-guest reactions are also designed to tune the interfacial interaction, which can be used for the

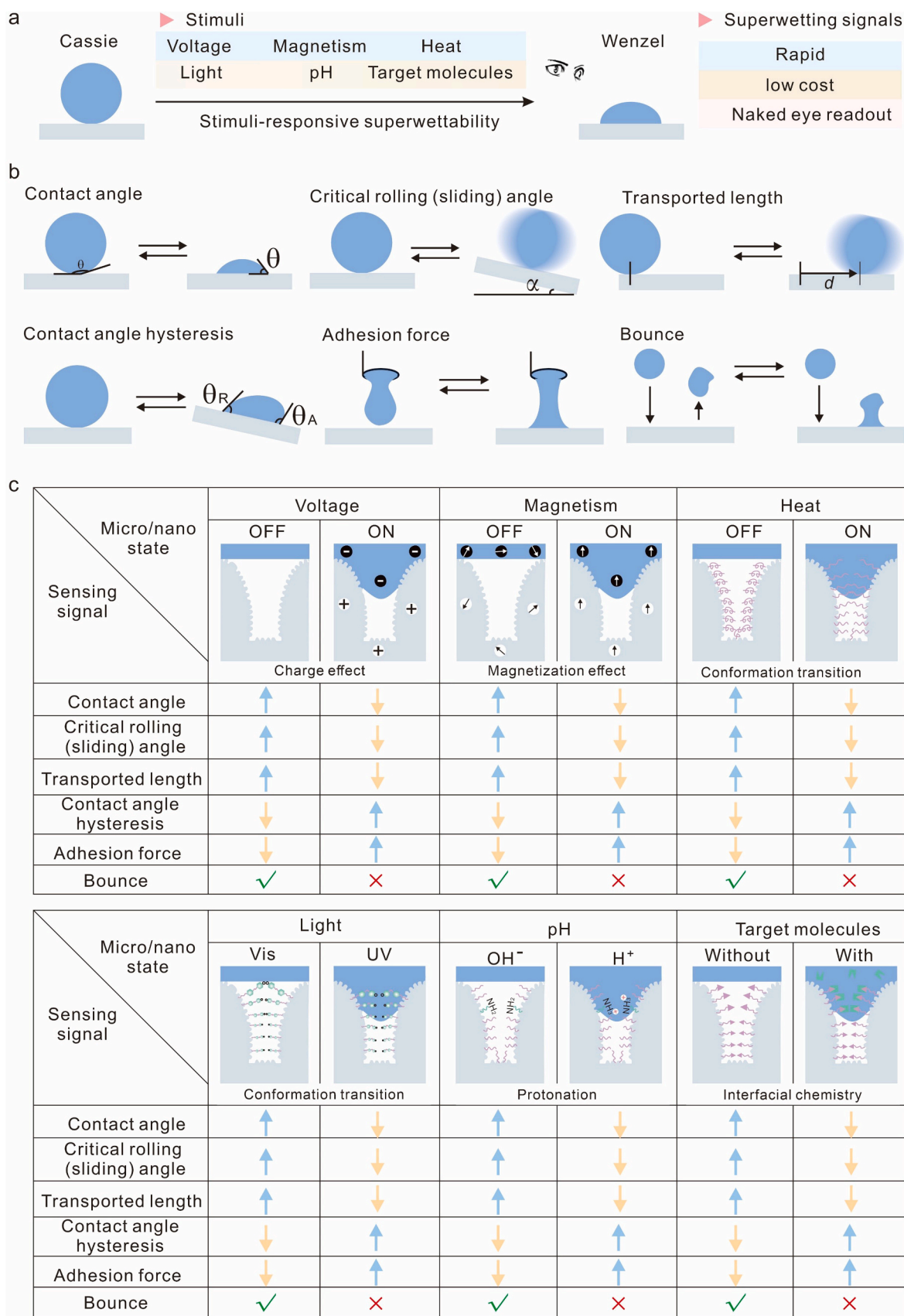


Fig. 6. Fundamental biosensing mechanism based on stimuli-responsive superwettability. (a) Interfacial sensing principle using different stimuli. (b) Various superwetting switches detected by naked eye. (c) The interfacial mechanism switched between Cassie state and Wenzel state under various external stimuli.

control of droplet movement [196,197]. Thus, the critical sliding angle is employed for signaling the concentration of targets. Other wetting behaviors, such as droplet displacement, speed, contact angle hysteresis, and bouncing, are demonstrated sensitive to reflect the concentration change of target [193]. These wetting signals are droplet-based responses, which can be easily detected using naked eye. Thus, semi-quantitative detection is achieved without any complex instruments. For accurate quantitative detection, the use of an automatic contact angle instrument or other equipment is still necessary.

Responsive surface wettability holds great potential in the field of bioimaging, with significant implications for biosensing research. A typical example is the use of photoinduced superhydrophilic Gd-doped TiO₂ ellipsoidal nanoparticles (GdT_i-SC NPs) to enhance the sensitivity of MRI imaging.[119] By reducing the water contact angle and increasing the number of surface hydroxyl groups, the efficiency of paramagnetic relaxation enhancement (PRE) is greatly improved. Therefore, these photo-responsive GdT_i-SC NPs can be utilized as highly effective T₁ contrast agents for magnetic resonance imaging. Additionally, ultrasound-responsive hydrophobic nanoparticles are capable of producing micrometer-sized bubbles, leading to a blinking ultrasound signal. [198] By capturing and localizing these pulsing events with acquisition times as short as 11 ms, background-free ultrasound images can be obtained. This advancement in wettability-regulated nanoparticles will greatly facilitate the clinical translation of localization-based ultrasound imaging, enabling more sensitive detection of cancer and other diseases.

Droplet evaporation-enhanced enrichment mechanism

As the second category, droplet evaporation-enhanced sample consumption [199]. Superwettable surfaces, including superhydrophobic, SLIPS, and superwettable patterns, are designed as to decrease the contact area between the droplet and surface [57,200,201]. Bio-recognition events mainly designed to occur in the liquid phase rather than the surface. Their applications in diverse arenas such as national security, food safety, disease diagnosis, and environmental engineering have garnered significant attention for this unique approach.

The precise and accurate detection of targets is strongly depended on the evaporation process of sessile droplets (Fig. 7a). The way in which the liquid flows during evaporation plays a crucial role in determining the movement of nonvolatile solute particles and the patterns they form after deposition. During droplet evaporation, several physical mechanisms, including the gravitational force, Deegan flow, Marangoni flow, evaporation flux, and Brownian motion, are involved (Fig. 7b) [58]. When dealing with a droplet that can be affected by gravity, it is critical to consider the force of gravity. Additionally, during the process of evaporation, particles that cannot dissolve may come together to form aggregates. Two important flow regimes within sessile droplets are Deegan flow and Marangoni flow. Deegan flow is an outward radial flow that occurs within the droplet [202]. The coffee-ring effect occurs when insoluble particles or nonvolatile solutes deposit at the periphery of a droplet.

Another flow regime, known as the Marangoni flow [203], can be opposite to the Deegan flow, and is caused by surface tension thinning contact line and recirculates with the vortex, insoluble particles or nonvolatile solutes deposit at the center of the droplet and form a uniform spot upon evaporation. Random movements of insoluble particles/nonvolatile solute are produced by Brownian motion through interactions with liquid molecules. Even though it is often ignored in droplet evaporation, it should be taken into account for droplets that contain sub-micro solutes. Moreover, in situations where there is evaporation occurring on the surface of a droplet, there is a gradient resulting from temperature and concentration gradients. As the droplet moves towards the periphery of the diffusion of solute from the outermost layer towards the middle of the droplet. The adhesion between the droplet and substrate significantly influences the deposited patterns of

sessile droplets. The coffee-ring pattern is observable on almost all hydrophilic and hydrophobic substrates [204,205]. Nevertheless, superhydrophobic surfaces, liquid-infused surfaces and superwettable micropatterns provide easy and efficient platforms for enhancing spot homogeneity and preconcentrating samples. As a result, these methods present great potential for creating sensitive and accurate biosensors. One can create multifunctional microchips based on superwettable micropatterns by combining their open character with various signal output approaches. For instance, glucose, calcium, pH, and other parameters can be monitored by using superwettable colorimetric tapes to analyze sweat samples (Fig. 7c) [206]. Using droplet evaporation-enhanced enrichment mechanism, the signal probe can be enriched, which is favourable for the biorecognition process thermodynamically and kinetically, resulting in a significant amplification of the SERS (Fig. 7d) [207] and fluorescence (Fig. 7e) [161] signal. This amplification enhances the sensitivity and reduces the LOD of the superwettable microwell chip. The micropatterns which can hold droplets in a superwettable manner may also prove beneficial for amplifying reactions such as polymerase chain reaction (PCR) and gene sequencing, with significant implications in gene regulation and disease development. (Fig. 7f) [208]. Paper-based substrates provided a universal and sensitive platform for multiple targets detection by mass spectrometry. [209–211] For example, Pradeep and colleagues reported a superhydrophobic preconcentration paper spray ionization mass spectrometry (SHPPSI MS) for a diverse array of analytes with a low LOD down to sub-parts-per-trillion level.[212] The process involves the initial preparation of a superhydrophobic filter paper through chemical modification. A small hole is then created in the paper using external force, which serves as an anchor for the sample droplet. The sample droplet is allowed to evaporate completely, and this sampling and drying process is repeated at least three times to enhance the concentration of the target analytes. Subsequently, SHPPSI MS is performed to detect substances like urea, adenine, isoleucine, melamine, and caffeine. This method offers several advantages, including the ability to work with small sample volumes (10 μL) and simplified sample preparation procedures. Therefore, it holds significant potential for applications in food safety and environmental monitoring.

With the evaporation of droplets, the increased probability of molecular collision increases the sensitivity of detection, but reduces the selectivity, which poses a high challenge for the detection of nucleic acid mutation, especially single nucleotide polymorphism. As research on this aspect is still in its early stages, it is imperative that future efforts are dedicated towards it.

Liquid phase-regulated sensing mechanism

The two mechanisms mentioned above mainly depend on surface treatment. However, liquid phase has equal significance to regulate the sensing performance. The method that is typically utilized involves managing the characteristics of the liquid phase, which includes regulating its surface tension, viscosity, and interfacial tension (Fig. 8a).

The droplet pendant is frequently utilized in tensiometry for measuring the interfacial tension of liquids, indicating a sensitive tool for biodetection by monitoring the droplet surface tension. Yu and co-workers proposed a pendant droplet-based sensor for visual detection of acetylcholinesterase (AChE) and its inhibitor (Fig. 8b) [213]. When a droplet of liquid crystal (4-cyano-4'-pentylbiphenyl) is mixed with a cationic surfactant solution (Myr), Myr is adsorbed onto the interface between the LC and the solution. This results in a reduction in the interfacial tension of the droplet, causing it to fall when the gravitational force of droplet exceeds the tension. When the concentration of Myr decreases, the waiting time of LC droplet increases. The droplet cannot fall into the AChE and Myr mixed solution because Myr is enzymatically cleaved by AChE into myristic acid (MA) and choline. The inhibitors prevent AChE activity, causing the 5CB droplet to fall. As a result, visual detection AChE is successfully performed, and the detection of AChE

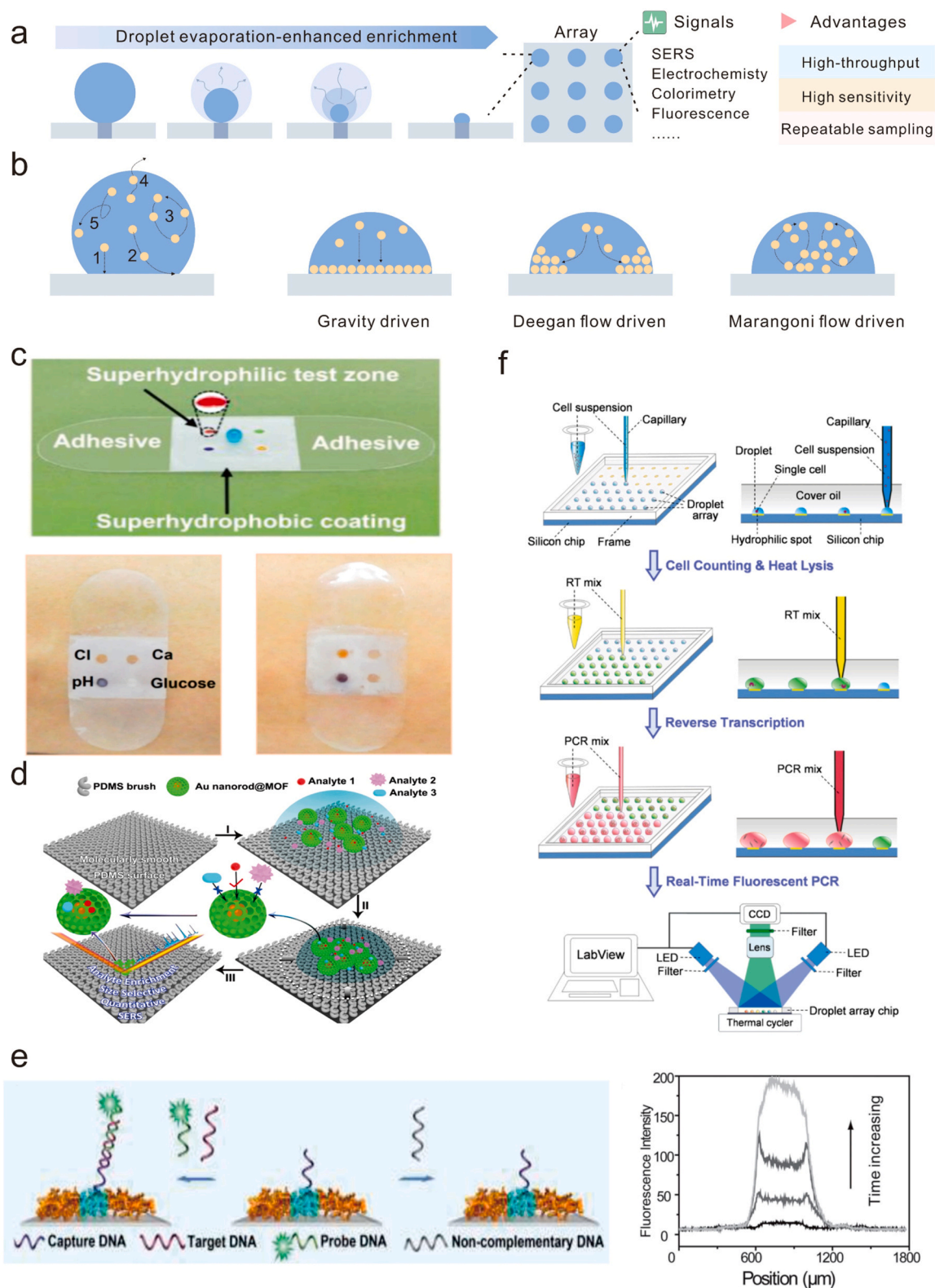


Fig. 7. Fundamental biosensing mechanism based on droplet evaporation-enhanced enrichment mechanism. (a) The diffusion and sensing mechanisms. (b) Driving forces of the sensing probe during the droplet evaporation: gravitational force, Deegan flow, Marangoni flow. The droplet evaporation-enhanced enrichment as universal sensing mechanism to prepare various superwetable biosensor for (c) colorimetric,[206] (d) SERS,[207] (e) fluorescent,[161] and (f) PCR detection.[208] Part (c) Reproduced with permission [206]. Copyright 2019, ACS Publishing Group. Part (d) Reproduced with permission [207]. Copyright 2020, ACS Publishing Group. Part (e) Reproduced with permission [161]. Copyright 2015, Wiley Publishing Group. Part (f) Reproduced with permission [208]. Copyright 2015, Nature Publishing Group.

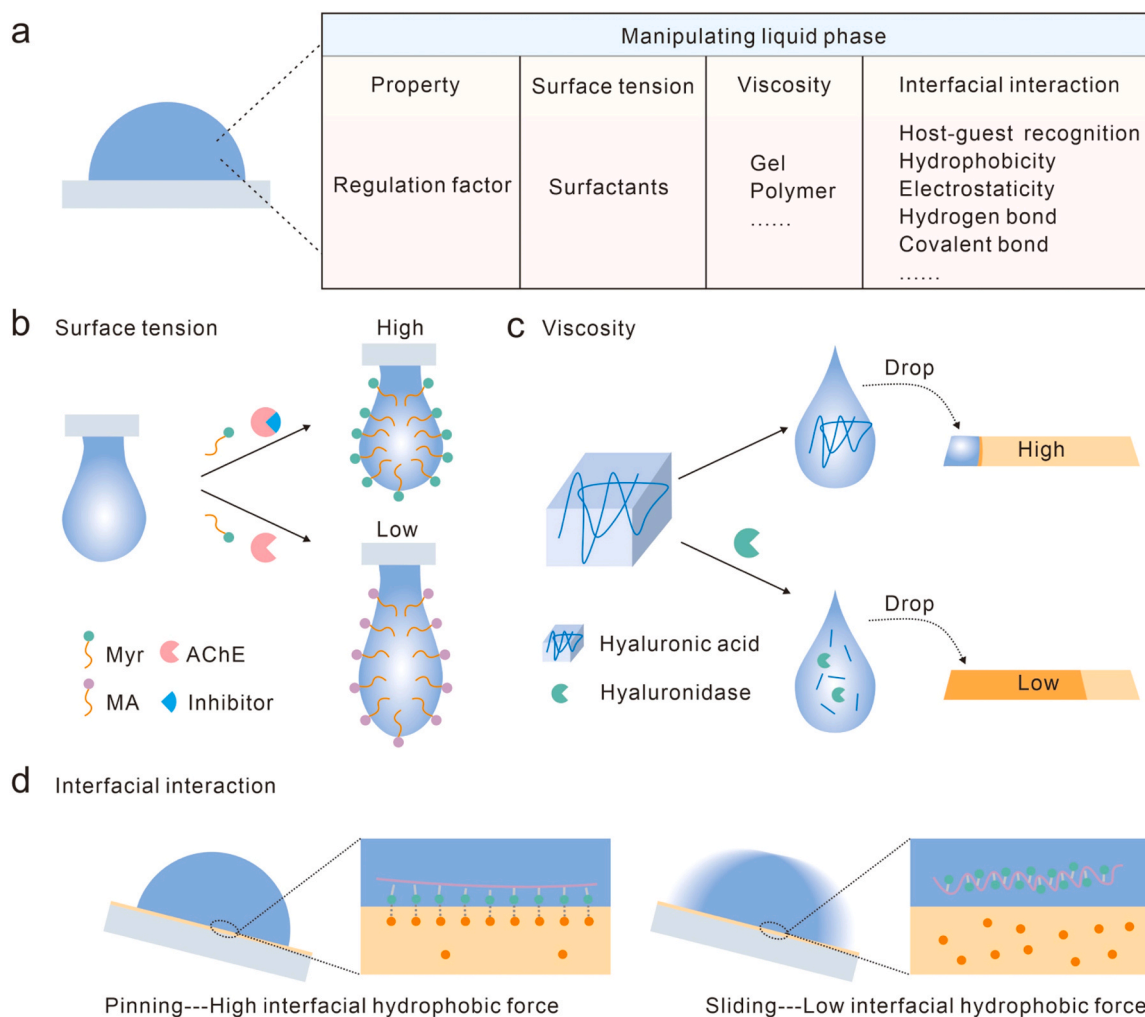


Fig. 8. Fundamental biosensing mechanism based on liquid phase-regulated sensing mechanism. (a) The sensing mechanisms. (b) Pendant droplet-based sensor for the detection of acetylcholinesterase (AChE) and its inhibitor. [213] (c) Paper-based flow sensor for the detection of hyaluronidase based on viscosity change. [216] (d) Controlling droplet motion by regulating interfacial interaction. [222] Part (b) Reproduced with permission [213]. Copyright 2021, RSC Publishing Group. Part (c) Reproduced with permission [216]. Copyright 2022, ACS Publishing Group. Part (d) Reproduced with permission [222]. Copyright 2019, ACS Publishing Group.

inhibitors is demonstrated. Controlling the release of surfactant is also an efficient way for pendant droplet sensor. Upon exposure to trypsin, a gelatin hydrogel that is encapsulated with Triton X-100 can be broken down in a targeted manner, thus liberating the surfactant and leading to a reduction in droplet surface tension [149]. This pendant droplet-based sensor is highly promising for sensing purposes, as it does not require labelled molecules, synthetic particles, or complex processes, making it a simple and inexpensive approach. In addition, adding a surfactant to the liquid can reduce the surface tension and prevent the formation of coffee-ring patterns, resulting in a homogeneous deposit spot [214,215]. Surfactants can also reduce the interfacial tension between two immiscible liquids, which can improve the adsorption or binding of analytes. To note, care should be taken to select a surfactant that does not interfere with the target analyte or the receptor used in the biosensor.

Another approach is to introduce a viscosity modifier to the liquid, such as a polymer or a nanoparticle. Viscosity modifiers can slow down the radial flow of liquid and promote the uniform distribution of solutes. Polymers such as hyaluronic acid, amylopectin, conductive polymers have been used as viscosity modifiers in interfacial biosensors [216–218], while nanoparticles such as silica and gold nanoparticles have been used to create a dense network that prevents the formation of coffee-ring patterns [219–221]. However, the addition of viscosity modifiers can also affect the diffusion of analytes and the binding kinetics, which can lead to slower response times and reduced sensitivity.

For example, Hu et al. proposed a simple, label-free and equipment-free paper-based flow sensors for detecting hyaluronidase (Fig. 8c) [216]. This method relied on a change in viscosity caused by enzymatic hydrolysis in a responsive polymer solution, leading to a longer water flow distance on the pH indicator paper. The third approach is to regulate the interface interaction by utilizing a multi-phase system, consisting of two or more liquids present at the interface. This can be achieved by manipulating the conformation of amphiphilic molecules (Fig. 8d) [222]. Altering the length or external temperature can tune the droplet wetting state from pinning to sliding. Moreover, adding a third liquid, such as an oil or a polymer, to the system promotes the uniform distribution of solutes [223–225]. The third liquid can act as a barrier that prevents the radial flow of the droplet, resulting in a homogeneous deposited spot. For example, adding an oil phase to an aqueous phase can create a Pickering emulsion [226], in which the oil droplets act as stabilizers that prevent the formation of coffee-ring patterns. However, the addition of a third liquid can also complicate the fabrication process and increase the cost of the biosensor.

Applications of superwettable biosensing

With the progress of superwettability-centered science and technology, numerous sensing interfaces in molecular detection and bio-analytical chemistry have been established. Especially, superwettable

surfaces have been widely used as important sensing interfaces to be interfaced with diverse targets, such as nucleic acids, proteins, small molecules, bacteria, and cells. Typical examples using different sensing mechanism and superwetttable materials are summarized in Table 1.

Nucleic acid analysis

The superwetttable approach has drawn a lot of interest due to its adaptability, straightforward experimental setup, etc. Among all the superwetttable biosensing applications, nucleic acid analysis, such as DNA and miRNA, has received the greatest attention. The popularity of this research hotspot may be derived from an intriguing mix of various aspects, including the promise of the superwetttable approach, the significance of nucleic acid, and the unique features of nucleic acid. Nucleic acids analysis has always been multidisciplinary, including medicine, chemistry, and biology. In recent years, a series of method has been produced, which can be classified into three groups: (1) DNA conformation-regulated interfacial interaction; (2) DNA-modified superwetttable biosensor; (3) DNA biochemistry-regulated liquid phase.

DNA-modified superwetttable biosensor has been a crucial research field in recent years. Generally, superhydrophilic surface can be

prepared from superhydrophobic surface by UV irradiation or oxygen plasma treatment using a mask [227–229]. The size of superhydrophilic region, which has influences on the sensing performance, can be controlled by the design of mask. Several strategies have been reported to immobilize DNA capture probes on the superhydrophilic surface. For example, (3-glycidioxypropyl) trimethoxysilane modified superhydrophobic microwells is capable to react with aminolabelled DNA. This sensitive nucleic acid sensing platform, constructed based on superhydrophilic microwells (0.190–1.483 mm) spotted on superhydrophobic substrates, has a detection limit of 2.3×10^{-16} M [161]. After simple washing with milli-Q water and buffer solution, the DNA modified superwetttable pattern can be used for the construction of sensitive electrochemical and fluorescent biosensors to detect metal ions and miRNA [230,231]. Inspired by the self-cleaning property of titanium dioxide (TiO₂) nanoparticles (98 ± 35 nm), Zhang et al. reported a renewable superwetttable miRNA biochip. Such patterned microchips achieved low LOD down to picomolar level (Fig. 9a) [227]. Moreover, the superwetttable interface guarantees that droplets are immobilized in superhydrophilic microwells, which have the potential to be utilized in disease diagnosis. Target-induced DNA conformational switch is a biophysical process between randomly coiled structures and folded state.

Table 1
Summary of various superwetttable biosensors for detecting different targets.

Mechanism	Materials	Wettability	Target	LOD	Ref.
1	Organogel	Slippery	ATP, miRNA, thrombin, kanamycin, and DNA	2.08 pM	191
1	GOx, GNPs, silica coated MNPs	Superhydrophilic	PSA	3.2 pg/mL	195
3	Organogel	Superhydrophilic	ATP	172 pM	222
2	TiO ₂ nanowires	Superhydrophilic/superhydrophobic micropattern	miRNA	88 pM	227
2	Nanodendritic gold	Superhydrophilic/superhydrophobic micropattern	miRNA	1 pM	229
2	Lauryl methacrylate, hydroxyethyl acrylate	Superhydrophilic/superhydrophobic micropattern	Metal ions and miRNA	0.4 pM	230
2	Polyacrylic acid, hexadecyltrimethoxysilane	Superhydrophilic/superhydrophobic micropattern	Fe ³⁺ , Cu ²⁺	0.045 μM / 0.030 μM	231
1	Organogel	Slippery	Thrombin, miRNA	0.8 nM / 1pM	234
2	3-glycidioxypropyl trimethoxysilane epoxysilane	Slippery micropattern	<i>E. coli</i>	250 CFU/mL	235
2	PMMA	Slippery micropattern	IL-6	0.5 pg/mL	236
2	COP	Micropattern	<i>E. coli</i>	103 CFU/mL	237
2	α-cellulose	Superhydrophilic/superhydrophobic micropattern	Hg ²⁺	100 pM	240
2	Self-assembled monolayers modified gold electrode	Hydrophobic	DOX	100 nM	241
2	Porous support PVDF film	Superhydrophilic/superhydrophobic micropattern	Organic dyes / miRNA	24.1–59.6 fM / 308.5 fM	242
1	Nitrocellulose and polycarbonate	Superhydrophobic	human IgG model antigen	73.6 pg/mL	246
3	Polystyrene beads	Superhydrophobic	IL6 and PDGF-2 human protein	8.0 μg/L	247
2	Silicon pillar	Superhydrophobic	IgG immunoglobulins	8.0 μg/L	249
2	ITO/Ti/Au/nAu	Superhydrophilic/superhydrophobic micropattern	miRNA-141 and PSA	0.8 nM and 1.0 pM	251
1	Sticky superhydrophobic substrates	Superhydrophobic	Cell	Occupancy ~81%	259
1	Silicon wafer	Superhydrophilic/superhydrophobic substrate	Cell	Single cell	260
3	Graphene and hydrogenated graphene	Superhydrophilic	Cell	Single cell	261
1	Silicon nanoglass	Superhydrophobic/Superhydrophilic micropattern	Cell	Single cell	262
3	Photoresponsive spiropyran	Superhydrophilic	Cell	Single cell	263
2	ITO nanowire	Superhydrophobic/Superhydrophilic micropattern	Cell	Single cell	264
2	Magnetic beads	Superhydrophilic	H1N1 virus	0.032 HAU	268
1	Isopropyl-β-D-galactopyranoside, Fluorescein-di-β-D-galactopyranoside	Hydrophilic/hydrophobic micropattern	Cell	Single cell	270
2	PDMS and carnauba wax	Superhydrophobic	<i>E. coli</i> and <i>S. aureus</i>	Bactericidal efficiency > 97%	273
2	Chlorin e6	Superhydrophobic	<i>S. aureus</i> , <i>E. coli</i> , and <i>P. aeruginosa</i>	Bactericidal efficiency ≥ 99%	274
2	DPSNs-Au-MBA-aptamer	Superhydrophobic	<i>S. aureus</i>	2.6 CFU/mL	275

Abbreviation: 1: Stimuli-responsive mechanism; 2: Droplet evaporation-enhanced enrichment mechanism; 3: Liquid phase-regulated sensing mechanism; GOx, glucose oxidase; GNPs, gold nanoparticles; MNPs, magnetic nanoparticles; PSA, prostate specific antigen; *E. coli*, *Escherichia coli*; PMMA, poly(methyl methacrylate); IL-6, Interleukin 6; COP, cyclo-olefin polymer; *S. aureus*, *Staphylococcus aureus*; DOX, doxorubicin; PVDF, polyvinylidene fluoride; PDMS, polydimethylsiloxane; ITO, indium tin oxide; DPSNs, dendritic porous silica nanoparticles, MBA, 4-mercaptobenzoic acid, *P. aeruginosa*, *Pseudomonas aeruginosa*.

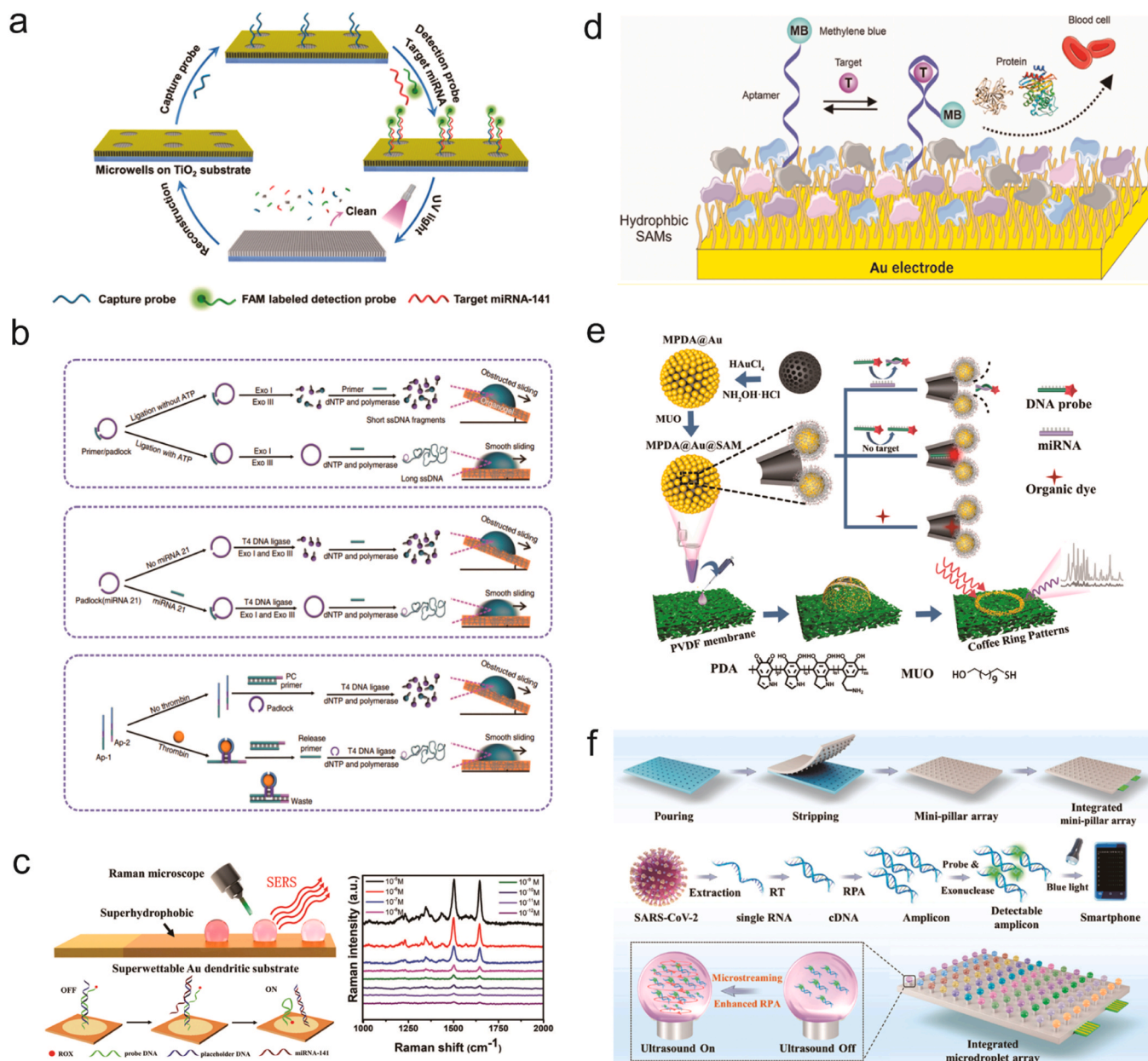


Fig. 9. Superwettable detection of nucleic acid. (a) TiO₂ based renewable superwettable micropattern for fluorescence detection of miRNA. [227] (b) Rolling circle amplification-based biosensor for miRNA detection on SLIPS. [234] (c) Superhydrophobic nanodendritic gold substrates for SERS detection of miRNA. [229] (d) Self-assembled hydrophobic monolayers for constructing anti-fouling electrochemical biosensor. [241] (e) Coffee-ring enrichment for the detection of miRNA. [242] (f) Ultrasonic micropillar-based isothermal amplification for the detection of SARS-CoV-2. [243] Part (a) Reproduced with permission [227]. Copyright 2018, Elsevier Publishing Group. Part (b) Reproduced with permission [234]. Copyright 2020, Nature Publishing Group. Part (c) Reproduced with permission [229]. Copyright 2018, RSC Publishing Group. Part (d) Reproduced with permission [241]. Copyright 2022, ACS Publishing Group. Part (e) Reproduced with permission [242]. Copyright 2022, Elsevier Publishing Group. Part (f) Reproduced with permission [243]. Copyright 2022, ACS Publishing Group.

As is well known, single-stranded DNA (ssDNA) is an amphiphilic molecule with hydrophilic phosphate backbone and a hydrophobic nucleobase. Double-stranded DNA displayed is mainly hydrophilic because hydrophobic bases are paired by hydrogen bonds. Tuning the wettability of ssDNA is theoretically and experimentally achieved by regulating the length [232–234]. Base-base stacking is promoted by long ssDNA, which takes on a shape that reduces the number of hydrophobic regions exposed and available for interaction with the hydrophobic surface. This leads to a feeble hydrophobic interaction at the interface of liquid and solid. In contrast, short ssDNA has relatively strong hydrophobic interaction at liquid/solid interface. The difference of interfacial force is reflected in the difference of interfacial wetting behavior of liquid droplets, which can be used for biosensing. Examples include our

recent work on the target-induced rolling-circle amplification (RCA) for the detection of ATP, miRNA, and thrombin using slippery organogel surface [191,222,234]. The RCA reaction generated long ssDNA, which improved the interfacial hydrophobicity and increased critical sliding angle of droplet (Fig. 9b) [234]. Long ssDNA molecules are more likely to form a cheerfully curled conformation, minimizing nucleobase exposure and achieving weak interfacial adhesion to allow droplets to slide easily. Although it exhibited significant sensing performance under complex conditions, several issues still existed to be solved, such as the accurate control of oil layer thickness and the hydrophobicity interference of enzymes and proteins. Didar and coworkers extended this strategy to SLIPS biosensor [235,236]. DNazymes were incorporated in lubricant-infused surfaces, leading to detection of target associated with

Escherichia coli (*E. coli*) in milk, meat, fruit juice, and food package [235]. The frictionless and nonadhesive superhydrophobic and lubricant-infused regions maintain the ability of the DNA probe to interact with its intended target while preventing any unwanted adhesion to the surface. These superwetttable biosensors can readily be prepared with flexible substrates, such as food packaging and bottles [237], indicating the promise for monitoring pathogen contamination in real-time, without requiring the containers to be opened. Ongoing efforts should focus on mitigating the negative public-health-related impacts of food-borne illnesses the superwetttable surface may generated.

Another strategy is to deposit nanomaterials, such as Nobel metal nanoparticles and inorganic nanoparticles [16,238,239], on the superhydrophilic region. Nanomaterials modified surface has high specific surface area, which is favorable for DNA capture probe loading capacity. The roughness is also improved to a certain extent, resulting in the confined molecular recognition and improving the probability of molecular collision. By labeling fluorescent and SERS tags on the DNA terminal, the target-induced conformation switch can be employed for highly sensitive and reliable detection of miRNA [240]. The SERS platform constructed by incorporating plasma silver nanoparticles in the hydrophilic region ($\sim 400 \mu\text{m}$) had a detection limit of 100 pM for Hg^{2+} . A superhydrophobic nanodendritic gold substrate can be produced by merging a superwetttable interface with a nanodendritic gold structure, allowing for SERS detection with high sensitivity down to 10^{-12} M (Fig. 9c) [229]. The nanodendritic gold substrate enhances Raman signal by providing numerous hotspots. Furthermore, self-assembled monolayers with hydrophobic properties can act as a molecular barrier to prevent or reduce the occurrence of unspecific adsorption of bodily fluids such as sweat, urine, and blood on traditional electrode surfaces (Fig. 9d) [241]. Apart from coffee-ring-free surface, uniform coffee-ring also provides a sensitive method for biosensing. Zhang and colleagues placed AuNPs on the external layer of mesoporous polydopamine nanoparticles and modified them using a self-assembled monolayer of 11-Mercapto-1-undecanol in order to selectively enrich analytes in the mesopores (Fig. 9e) [242]. Afterwards, the nanoparticles were organized into a uniform coffee ring pattern with a width of 500 μm and an inner diameter of approximately 4 mm on a porous host made of polyvinylidene fluoride film with a pore size of 0.45 μm by rapidly packing the particles facilitated by the infiltration of water in the membrane pores. Ultimately, this biosensor realized highly sensitive analysis of miRNA with a limit of detection of 308.5 fM. Accurate control the number of DNA capture probe on nanomaterial-modified microwells is an unsolved problem, which might have impact on the reproducibility of test results. To reduce the relative standard deviation of detection, high-throughput sensing array may be a solution.

Biochemical reactions can be completed in liquid phase to produce signal probes in a homogeneous solution, which can then be dropped onto superwetttable surfaces for detection after droplet evaporation. These surfaces, including superhydrophobic, SLIPS, micropillars, and micropatterns, improve sensitivity through droplet-evaporation enhanced enrichment without involving molecular recognition [243, 244]. Xu and colleagues demonstrated an integrated microdroplet array platform consisting of superwetttable micropillar arrays and an ultrasonic unit to enhance recombinase polymerase amplification (RPA) for the simple and high-throughput determination of SARS-CoV-2 (Fig. 9f) [243]. The microdroplets on individual micropillars greatly decrease reagent consumption, while microstreaming driven by ultrasound accelerates amplification by promoting RPA component interaction. Detection time is shortened by 38.8 – 59.3%, and end-point fluorescence intensity increases by nearly 2 times. The ultrasonic superwetttable micropillar has a lower detection limit of 0.42 copy/ μL for SARS-CoV-2 when compared to controls. This makes it suitable for high-throughput nucleic acid analysis during the epidemic and has the potential for detecting other viruses or pathogens.

These superwetttable biosensors can detect various nucleic acids, such as DNA and miRNA, with excellent reliability and stability, even in

complex environments like human serum, urine, and cells. The outstanding sensing performance makes it highly promising for precise disease detection. However, the complex labeling of the biomolecules and usage of large instruments could restrict their application in point-of-care detection. To overcome this limitation, the future approach will concentrate on the incorporation of superwetttable biosensors with portable equipment to facilitate point-of-care testing.

Immunoassay

Superwetttable immunoassay has gained momentum in recent years because of its significant potential in the field of biomedical applications. Similar to the DNA immobilization process, antibodies contain amino groups, which can be fixed on the solid surface using the same covalent bonding. Taking advantage of the sensitive analytical mechanisms, the immunosensing system can transform different signal responses, such as visual, electrochemical, fluorescent, and SERS, to investigate various immunoreactions based on the specific immunorecognition events. In such immunoassay system, superwetttable biosensors modified with superhydrophobic or lubricant-infused regions demonstrated excellent anti-biofouling features. Compared with the traditional interfacial blocking reagents, such as bovine serum albumin (BSA), it displayed better interfering proteins repelling performance.

At present, the established superwetttable immunoassay is generally performed by an enzymatic reaction. Enzyme-based immunoassays, such as the commonly used sandwich and competitive formats, have been created because they offer a viable way to improve the signal. The typical choices for enzyme labels include horseradish peroxidase (HRP), β -galactosidase (β -Gal), alkaline phosphatase (ALP), and glucose oxidase (GOx). For example, GOx is typically used in sandwich-type immunoassays for PSA detection using the pH-responsive superwetttable surface [195]. Primary antibody against PSA was used to modify magnetic beads. After immuno-binding, a secondary antibody labeled with GOx was loaded onto gold nanoparticles. The glucose is then added, which leads to the formation of gluconic acid, causing the local pH value to drop. As a result, the pH-responsive superwetttable chip undergoes a wettability switch from hydrophobic to hydrophilic. By observing the change in CA, prostate cancer can be visually detected in clinical blood samples (Fig. 10a) [195]. Gold-enhanced and magnetic electrochemical immunoassays were developed for multianalyte detection using an aqueous solution diffusion-localized platform, which minimizing the coffee-ring effect and improved the sensitivity [245,246]. By preparing various superwetttable patterns, enzyme-linked immunosorbent assays (ELISA) were conducted to sensitive detect human immunoglobulin (IgG), interleukin 6 (IL-6), PDGF-2, and H1N1 viruses [247–250]. Superwetttable micropillars were created by incorporating gold nanoparticles (60 ± 35 nm) functionalized with antibodies, which precise delivery of analytes from the sample to the active sites of the device, allowing for the specific detection of human IgG in urine with impressive sensitivity and precision (Fig. 10b) [249]. The detection of IgG in actual urine samples was successfully performed with high sensitivity. The detection limit of 8.0 $\mu\text{g/L}$ was better than that achieved by traditional nephelometric techniques. Furthermore, SLIPS technology can prevent non-specific adhesion and enable the detection of IL-6 in whole blood or plasma with a detection limit of 0.5 pg/mL (Fig. 10c) [236]. To link the capture antibody to the fluorosilanized surface and prevent non-specific adhesion, an epoxy-based silane is used to functionalize the antibody. This SLIPS biosensor enables high promise in early diagnosis of disease in viral respiratory infections. The modified antibody SLIPS biosensor eliminates non-specific adhesion, allowing for the detection of IL-6 in whole human plasma or blood during coagulation, with a limit of detection of 0.5 pg/mL. By applying patterned bioink infused with a lubricant coating, the capture antibody is further functionalized for covalent linkage to the fluorosilanized surface and to prevent non-specific adhesion. The SLIPS-based biosensor permits early diagnosis of worst-case outcome in viral respiratory infections. Treating with oxygen

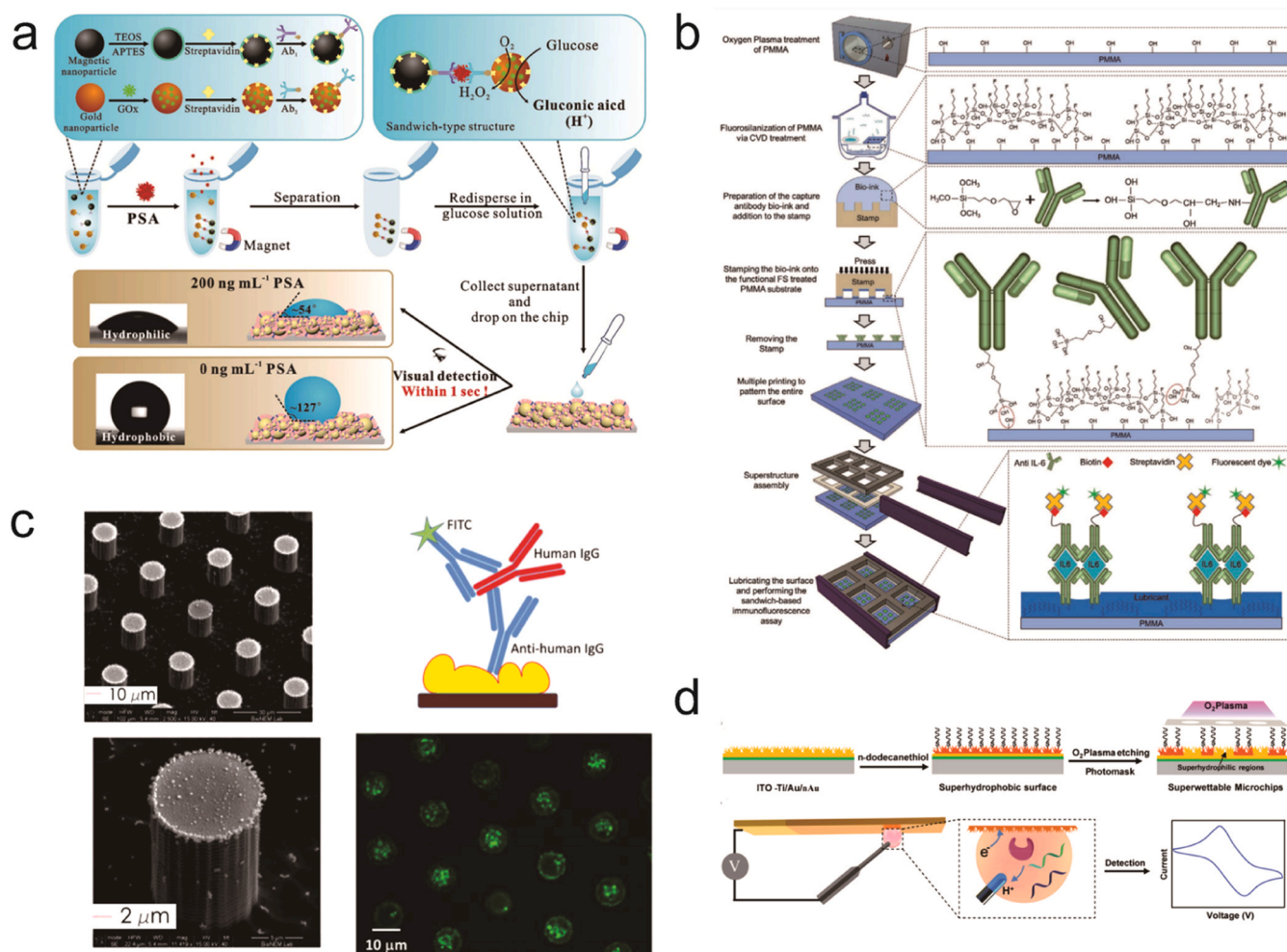


Fig. 10. Superwetable biosensor for immunoassay. (a) Sandwich immunoassay based visual detection of PSA using pH-responsive superwetable biosensor. [195] (b) Fluorescent immunoassay for IgG detection on superwetable micropillar. [249] (c) Lubricant-infused biosensors with antibody micropatterning for the detection of IL-6 in human whole plasma using immunofluorescence. [236] (d) Superwetable biosensor for electrochemical detection of PSA. [251] Part (a) Reproduced with permission [195]. Copyright 2019, Springer Publishing Group. Part (b) Reproduced with permission [249]. Copyright 2019, ACS Publishing Group. Part (c) Reproduced with permission [236]. Copyright 2020, Wiley Publishing Group. Part (d) Reproduced with permission [251]. Copyright 2018, ACS Publishing Group.

plasma, superhydrophilic microwell was fabricated on the superhydrophobic gold surface. Integration of such superwetable biosensor with an electrochemical signal allows for accurate detection of PSA (Fig. 10d), [251] offering a simple and energy-efficient technology with great potential for clinical biosensing applications.

Numerous contemporary studies have demonstrated that superwetable surfaces could be used to control the droplet wetting performance and enrich analytes, resulting in the enhanced sensitivity and elimination of non-specific adsorption. It is important to give additional focus to certain novel tactics. The manufacture of a point-of-care testing sensor using a microfluidic paper-based analytical device that features superwetable immunoassay is anticipated. Integrating the portable smart phone with superwetable immunoassay is promising for the healthcare management and on-site environmental monitoring.

Single-cell trapping

Because of the critical role of cells in biological technology and human wellness, cell-related biosensing has garnered a great deal of interest in recent years. Researches using superwetable surfaces has improved our knowledge of cellular responses to stimulation, cell fate transition, and gene transcription stochasticity [252–254]. The use of physical methods, like stencil patterning, droplet encapsulation, and

passive/active traps, provide high selectivity and throughput for capturing cells in predetermined patterns [255,256]. However, these methods are restricted in their ability to be scaled up and used in cell assays due to damage caused by shear stress, thermal effects, and photodamage. To improve cell viability and functionality, microfluidics is often combined with physical methods [257,258]. Alternatively, physicochemical patterning relies on cell-surface interactions and surface modification strategies which efficiently trap cells onto superwetable surfaces, forming patterns that are cytophobic and cytophilic.

Among these techniques, inkjet-printing stands out as the gold-standard for single-cell patterning applications owing to its high resolution, cost efficiency, negligible cell damage, and nanometer-sized feature sizes. Inkjet-printed sticky superhydrophobic surfaces with protein spots (4 μm) can trap multiple single-cell with high-efficiency (Fig. 11a) [259]. Within 30 min, the subcellular-sized patterning chip has the potential to attain almost 81% single-cell occupancy, making it a useful tool in real-time apoptosis research for individual cells. To meet the demands of the single-cell analysis, Song and colleagues utilized a substrate with a wettability-pattern (100 μm). to separate and manipulate individual cells, forming an array of femtoliter droplets (Fig. 11b) [260]. These droplet arrays containing single cells could be utilized for high-throughput functional bioanalysis. Non-destructive hydrogen plasma can be used to systematically control the wettability of graphene

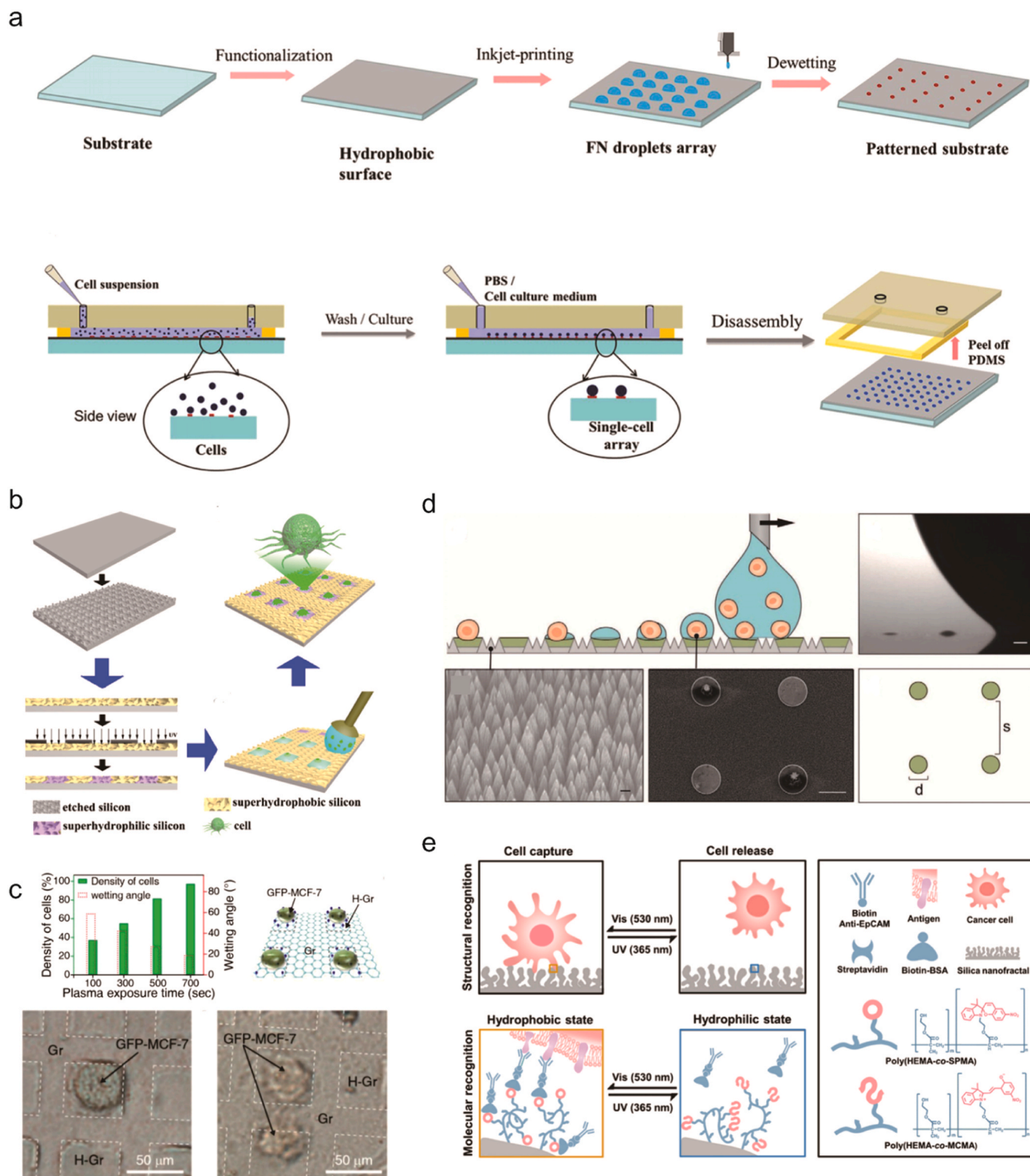


Fig. 11. Superwetttable biosensor for single cell trapping. (a) Inkjet-printing micropatterned surface for single-cell trapping on sticky superhydrophobic surface.[259] (b) Wettability-pattern for single cell isolation.[260] (c) Wettaable graphene surface for cancer cell isolation.[261] (d) Superwetttable microarrays for high-efficient single-cell trapping.[262] (e) Cell release amplified by photoresponsive molecule on a superwetttable surface.[266] Part (a) Reproduced with permission [259]. Copyright 2018, ACS Publishing Group. Part (b) Reproduced with permission [260]. Copyright 2015, ACS Publishing Group. Part (c) Reproduced with permission [261]. Copyright 2020, ACS Publishing Group. Part (d) Reproduced with permission [262]. Copyright 2021, Wiley Publishing Group. Part (e) Reproduced with permission [266]. Copyright 2019, ACS Publishing Group.

from hydrophobic to superhydrophilic and pattern it into microscale sections (Fig. 11c) [261]. Utilizing the significant difference in wettability across various areas of graphene, a solitary cancer cell was extracted and utilized for assays through the creation of a single cell array. The size of the micropattern has great influence on the single cell trapping. Paavo et al. concluded that superhydrophilic sites, which were twice as large as the cells, were the most effective for acquiring a single-cell trap (Fig. 11d), resulting in up to 30% single-cell trapping success [260–262]. Jokinen and co-workers demonstrate the use of

superhydrophobic–superhydrophilic microarrays to capture individual immune cells ($\approx 10 - 20 \mu\text{m}$) by utilizing superhydrophilic spots that have diameters smaller than $30 \mu\text{m}$. As a result, the trapping of single cells is based on size selectivity, rather than Poisson dilution. These superwetable micropatterns offer a flexible and promising strategy for efficient analysis and screening of cells.

The use of cell manipulation in various biological scenarios, such as cell-matrix interaction, cell-based diagnosis, and tissue engineering, has raised significant concerns. However, capturing and releasing cells with

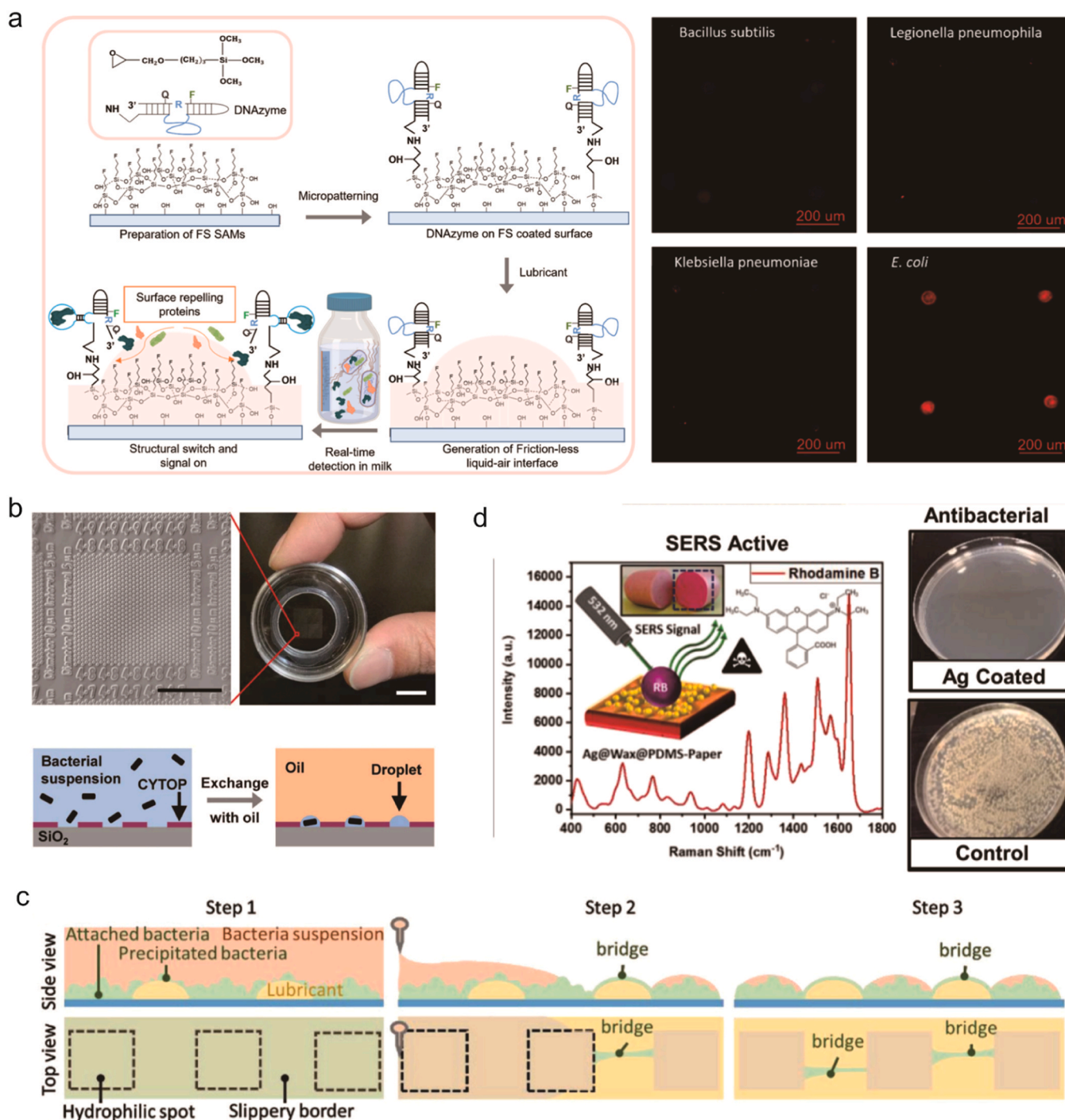


Fig. 12. Superwetable biosensor for bacteria-related study. (a) DNAzyme-based lubricant-infused surfaces for *E. coli* detection. [235] (b) Drug efflux assay at the single-bacteria level by a femtoliter droplet array. [270] (c) Geometric bridges for bacteria assay using patterned SLIPS. [271] (d) Superhydrophobic surfaces with high SERS and antibacterial activity. [273] Part (a) Reproduced with permission [235]. Copyright 2022, ACS Publishing Group. Part (b) Reproduced with permission [270]. Copyright 2012, RSC Publishing Group. Part (c) Reproduced with permission [271]. Copyright 2020, Wiley Publishing Group. Part (d) Reproduced with permission [273]. Copyright 2022, Elsevier Publishing Group.

high efficiency is still a significant challenge. Wang' group has presented a series strategy to address this issue by tuning the chemistry and topological structure of superwetable surface [263–265]. A non-photosensitive molecule is present on the nanostructured surface coated with photoresponsive spiropyran, as shown in Fig. 11e, which enables the efficient discharge of cancer cells [261,266]. This method achieved high efficiency for cell trapping and releasing because the protrusions of cancer cells can interact with the nanofractal surfaces, while the non-photosensitive and hydrophilic molecule can increase molecular recognition by reducing steric hindrance and preventing nonspecific cell adhesion. Therefore, this approach could offer a new path to creating advanced smart materials that can support high-quality biological analysis and clinical diagnosis. The process of single-cell patterning remains challenging, especially in comparison to bulk-cell culture, due to the requirement for cells of micrometer scale. Most methods necessitate expensive fabrication equipment and large amounts of materials, making it difficult to realize precise and high-throughput single-cell patterning. Fortunately, the advancements in micro/nano-fabrication technology present exciting opportunities to overcome these obstacles [267,268]. Future researchers, such as increased throughput, precision, and multiplexing through automation and serial processing, as well as the versatility of single-cell biosensors and enhancements in sub-micron to nanometer scale structures, are necessary for cell-related investigations. Additionally, further studies are required to establish single-cell patterning as a universal and accessible personalized strategy for drug discovery, clinical diagnosis, and disease treatment. Despite the challenges, the method of single-cell patterning has vast potential to be utilized as a potent tool for discovering biological information.

Bacteria-related study

The significance of pathogenic bacteria has recently been highlighted because of their severe impacts on public health, food safety, and the human living environment. Studying the pathogenic bacteria pose several challenges due to their small size and large population. Fortunately, wettability patterned microchips offer a promising solution for the study of microbiology as they have similar dimensions and high-throughput capabilities. The increase in multi-drug resistance among bacteria species in clinical settings is a pressing concern for managing infections in the public. When tested in a complex background matrix, such as milk and meat, DNAzyme-based lubricant-infused surfaces (LISzyme) are demonstrated to be more efficient in detecting bacteria and preventing nonspecific binding in comparison to other commonly used "blocking" approaches [235,269]. The lubricant infusion on sensing surfaces leads to a 4-fold improvement in the signal-to-noise ratio detected by the DNAzyme, as compared to untreated surfaces (Fig. 12a) [235]. LISzyme (30–40 nm) significantly improves the biosensing performance for *E. coli*, resulting in a low limit of detection down to 250 CFU/mL in milk within 60 min. Wettability-patterned biosensor (0.8 × 1.8 cm) is an ideal candidate to directly evaluate drug efflux activity in single *E. coli* encapsulated and separated in a femtoliter droplet array with a fluorogenic substance (Fig. 12b) [270]. This femtoliter microdroplet array, based on water-in-oil acquisition, has the advantage of being accessible to individual droplets, and the efflux-active single cell in the droplets can be easily collected under an optical microscope. Infusing with water in hydrophilic porous polymer domains, SLIPS-based micropatterned biochips are suitable for the preparation of arrays of biofilm microclusters with specific 2D geometries (Fig. 12c) [271]. It demonstrated significant performance for the study of the formation of biofilm microclusters of *Pseudomonas aeruginosa* (*P. aeruginosa*) using various bacterial strains. A unique structure of "bacterial bridges" can be fabricated by utilizing a patterned lubricant-infused surface to connect biofilms of *P. aeruginosa* [271,272]. Modifying with Nobel metal nanoparticles, such as Au and Ag, multi-functional superhydrophobic surfaces can be designed with SERS

and antibacterial activity [273,274]. Onses et al. utilized antimicrobial superhydrophobic surfaces prepared from polydimethylsiloxane (PDMS) and Brazilian carnauba wax, the antibacterial activity against *E. coli* and *Staphylococcus aureus* (*S. aureus*) achieved to 99.37% and 93.48%, respectively, after silver plating of the film (Ag thicknesses: 25 nm, 50 nm, 100 nm, and 200 nm) (Fig. 12d) [273]. The superwetable surfaces display an excellent antibacterial property while few of them achieved quantitatively detection. Integrating sensing ability into antibacterial surfaces is an efficient strategy to recognize bacteria in the environment, which can facilitate public health authorities in handling infectious disease epidemics. Xu and co-workers proposed a multifunctional patterned liquid-infused surface, which was fabricated using porous nanocoatings [275]. Combined with the dendritic porous silica nanoparticles (259.6 ± 16.4 nm) -Au-mercaptopbenzoic acid-aptamer SERS tag, the surface managed to repel bacteria completely in the hydrophobic region, in addition to allowing for highly sensitive and reliable SERS detection of *S. aureus* in a low sample volume of 1 μL. The modification of the superhydrophilic microwell with antibodies resulted in a low LOD of 2.6 CFU/mL. These findings give significant information on the design and preparation of superwetable biosensors with integrated sensitive characteristics, antibacterial, and stability features.

Current superwetable surfaces are mainly focus on the antibacterial investigations. Future research directions are suggested to pay more attention for quantitative detection of bacteria using different sensing method. The superwetable-patterned surfaces extend the impact microchip and provide diverse applications into the fields of geoscience, environment, and biochemistry.

Wearable electronics

Wettability is an essential factor to consider when designing wearable electronics due to the frequent interaction of wearable devices with human fluids, such as sweat. Flexible sensors that capable of healthcare monitoring and real-time communication with mobile phones are envisaged in particular. Substantial work has been conducted into constructing flexible devices while also addressing the incorporation of superwettability to accomplish desired biofluid management and usage [276–279].

Generally, superwettability-patterned flexible sensors contain three functional domains, including superhydrophobic region, indicators-modified superhydrophilic layer, and biocompatible adhesive region, for in situ sweat biosensing. Superhydrophobic region using polymer encapsulation and superhydrophobic coating have been explored to make the property, which removes various contaminations including dust, bacterial, and microorganisms. The secretion of perspiration can be enriched to the area that is highly wettable, where it interacts with core biosensing elements to be analyzed through a colorimetric indicator. The combination of colorimetry and portable smart phones provides the superwetted strips with diverse biosensing capabilities, which make them a promising candidate for personal healthcare monitoring. For example, Xu et al. reported a series of wearable superwetted biochips, which patterned superhydrophilic colorimetric testing zone on a superhydrophobic substrate [206,280–285]. This flexible skin-mounted strip combines superhydrophobic-superhydrophilic microarrays and nanodendritic colorimetric biosensors. It enables simple and reliable detection of metabolic ions and molecule in sweat, such as glucose, calcium, chloride, and pH (Fig. 13a) [206]. Efficient sweat collection with reduced evaporation or contamination is crucial for obtaining reliable and accurate results in sweat analysis. Even though different epidermal microfluidic sensors and materials have been utilized for sweat collecting, surface wettability plays a vital role in the enhancement of sensing performance by effectively controlling the capillary force. Recently, Janus textiles are emerged for self-pumping sweat sampling and analysis [126,127,286–288]. The Janus textile can provide exceptional comfort by following a contact-pumping model, as it can transport sweat from the skin's hydrophobic side to the hydrophilic side

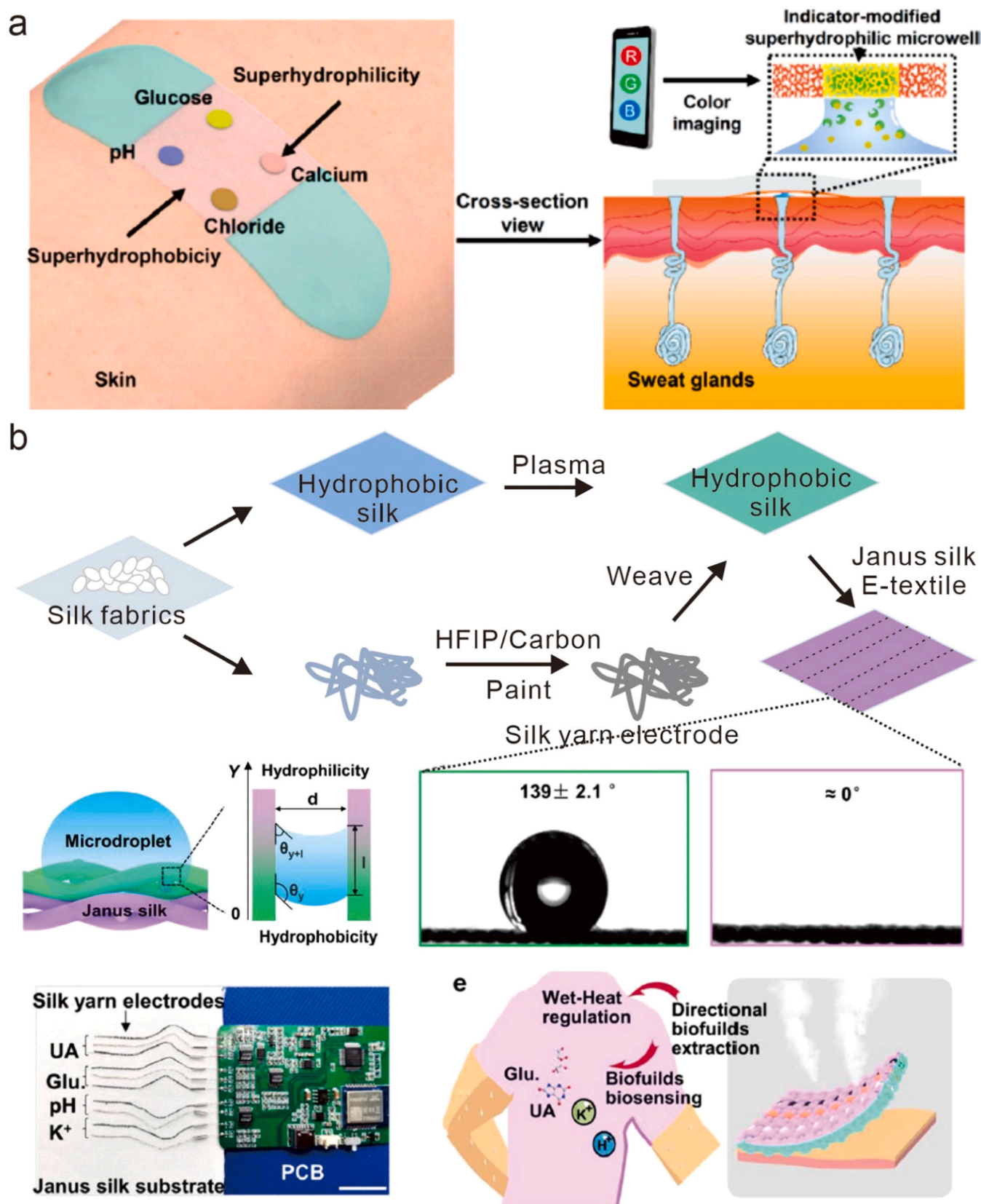


Fig. 13. Superwettable biosensor for wearable electronic. (a) Superwettable and flexible band serves as an effective biosensor for sampling and sensing sweat.[206] (b) Janus silk based electronic textiles provide high-efficiency biofluid sensing performance with wet-thermal comfort.[126] Part (a) Reproduced with permission [206]. Copyright 2019, ACS Publishing Group. Part (b) Reproduced with permission [126]. Copyright 2021, ACS Publishing Group.



Fig. 14. Challenges and prospects of superwetable biosensor.

of the embedded electrode surface in a thorough and unidirectional manner (Fig. 13b) [126]. The superwetable flexible sensors have several merits, such as affordability, scalability in production, and uncomplicated design, indicating great potential in disease diagnosis and healthcare management.

The degradation of hydrophobicity transpires in environments that have water vapor or high humidity because small water molecules nucleate and condense on the micro-/nanostructured surface. Long-term use of the wearable device depends on its durability, but micro/nanostructured superwetable surfaces are vulnerable to extrusions and abrasions, making it crucial to develop advanced design methods. One potential solution is to implement a microstructured surface frame as an "armor" to prevent the removal of micro-/nanostructures and improve reliability.

Conclusions and prospects

This review summarizes recent progresses in biosensors with special wettability, ranging from the fundamental theory, classification, sensing mechanisms, to their application potentials for sensitive detection of nucleic acid analysis, immunoassay, single-cell trapping, bacteria-related study, and wearable biosensing. With a better knowledge of the processes of wetting phenomena and improvements in biosensing technology, many wettability materials and biosensing methodologies with high sensitivity and selectivity are continually emerging. Nonetheless, some major issues need to be solved in future advancements. From basic to applied research, we address a wide range of exceptional prospects.

Single nanoparticle wettability and biosensing

Nanoparticles, featuring intricate geometries and surface chemistries, have immense potential for tuning the assembly of biological aggregates at sensing interfaces, leading to changes in detection signals. To accurately control the configuration and interparticle interactions, which determine the microstructures and signal transduction ability of particle-laden fluid interfaces, it is essential to comprehend the wetting behavior of individual nanoparticles. Understanding nanoscale wetting is a challenging issue, and the problem persists for single nanoparticles.

The wetting performance plays a vital role in understanding the structural and thermodynamic properties of nanoparticles on sensing interfaces. Therefore, developing advanced technology is crucial to characterize the wettability of single nanoparticle properly. In the context of single nanoparticle biosensing, it is necessary to engineer the structure and chemistry of nanoparticle, amplify microscopic wetting behavior into measurable signals, and establish quantitative relationships between the target concentration and nanoparticle wetting behavior at the single nanoparticle level.

Single molecule detection and imaging

Single molecule sensing has attracted the attention of chemistry, biology, and biophysics. Single molecule detection can potentially contribute to the development of next-generation bioanalytical and diagnostic technologies. Currently, it remains challenging to detect single molecule by using the superwetting signal such as contact angle, sliding angle, etc. Because the macroscopic wetting behavior is the collection of a large number of microscopic molecular behaviors. One promising strategy is to develop novel signal transduction strategy converting wetting performance into high spatiotemporal resolution signal, such as fluorescence and surface-enhanced Raman scattering. In addition, the superhydrophobic or superhydrophilic substrate requires high roughness, resulting in the low transparency and high background for single molecule imaging. Thus, improving the efficiency and accuracy for recognizing and imaging of single molecule will be of great importance for future applications.

Simultaneous detection of multiple targets

Despite much exciting and significant development, it is still challenging to identify many targets simultaneously using wetting performance as an output signal. Due to a lack of high-throughput and multifunctional capabilities, the superwetable biosensor can only detect one target by reading contact angle, sliding angle, droplet displacement, and so on. Yet, in some cases, such as disease diagnosis and pollution detection, distinct targets have to be detected at the same time. Although combining the functionalized superwetable biosensor with various signal output approaches (fluorescence, electrochemical, SERS, and colorimetry) has resulted in the detection of multiple targets, simultaneous detection of multiple targets by reading wetting behavior is an urgent problem that needs to be solved at the present.

Multi-step processes in biosensing

Integrating multiple processes in droplet arrays on superwetable surfaces, such as wettability-patterned surfaces and micropillar, poses a challenge. Functional bioassays including nucleic acid analysis, immunoassays, and cell-based assays typically require multiple steps. However, most bioassays performed on superwetable biosensor are usually restricted to one or two-step assays. If multi-step bioassays can be arranged on superwetable surfaces, most functional bioassays can be easily realized with high-throughput analysis, benefiting the application in biomedical industry. One alternative strategy is combining microfluidic devices with superwetable surfaces, constructing virtue units and walls which can be solid microstructure, hydrogel, detectors, droplet, or electronics. However, to meet industrial-level needs for multistep and complicated operations, most superwetable biosensors require external manipulation. Microfluidic channels provide for dynamic control of liquid and sample introduction, mixing, and initial reaction. These two technologies complement each other and have the potential to contribute to future automatic and industrial-level applications.

Industrialization and marketization

Regarding the biosensor with specific wettability, another concern lies in their industrialization and marketization. Most of these biosensors are still in the proof-of-concept stage, and have a long way to go before being applicable in practical scenarios. Therefore, it is crucial to improve the durability, intelligence, biosafety, flexibility, and controllability of these biosensors to make them commercially viable. To speed up this process, collaborative efforts from entrepreneurs, scientists, and medical professionals must be considered.

Although these unresolved essential difficulties need be addressed, their remarkable performance in practical applications has motivated the researchers to concentrate greater efforts to accelerating the commercialization of such superwettable biosensors. Future advances in development of novel interfacial sensing mechanism, multifunctional sensing substrate, and diversified sensing models are anticipated to revolutionize various research areas in the future, including on-site environmental monitoring and disease diagnosis. We believe that this review will motivate scientists from various fields to engage on resolving these existing challenges and extensively adopting superwettable biosensors as powerful bio-detection platforms.

CRedit authorship contribution statement

Zhong Feng Gao, Fan Xia, and Qin Wei conceptualized the paper. Zhong Feng Gao, Hai Zhu, Yanlei Li, and Hongmin Ma prepared the original draft of the manuscript. Xiaochen Yang, Xiang Ren, and Dan Wu created the specific visualization. Qin Wei, Fan Xia, and Huangxian Ju reviewed and edited the work.

Declaration of Competing Interest

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

Data Availability

No data was used for the research described in the article.

Acknowledgements

This work was supported by the National Key R&D Program of China (2021YFA1200400 and 2021YFA1200403), the National Natural Science Foundation of China (22176080, 22090050, and 22274062), the Natural Science Foundation of Shandong Province (ZR2023YQ015), and the Special Foundation for Taishan Scholar Professorship of Shandong Province (ts20130937).

References

- [1] D. Ye, X. Zuo, C. Fan, DNA nanotechnology-enabled interfacial engineering for biosensor development, *Annu. Rev. Anal. Chem.* 11 (2018) 171–195.
- [2] F. Yang, X. Zuo, C. Fan, X.E. Zhang, Biomacromolecular nanostructures-based interfacial engineering: from precise assembly to precision biosensing, *Natl. Sci. Rev.* 5 (2018) 740–755.
- [3] J. Wu, H. Liu, W. Chen, B. Ma, H. Ju, Device integration of electrochemical biosensors, *Nat. Rev. Bioeng.* 1 (2023) 346–360.
- [4] W. Wang, S. Yu, S. Huang, S. Bi, H. Han, J.R. Zhang, Y. Lu, J.J. Zhu, Bioapplications of DNA nanotechnology at the solid–liquid interface, *Chem. Soc. Rev.* 48 (2019) 4892–4920.
- [5] H. Altug, S.H. Oh, S.A. Maier, J. Homola, Advances and applications of nanophotonic biosensors, *Nat. Nanotechnol.* 17 (2022) 5–16.
- [6] C. Zong, M. Xu, L.J. Xu, T. Wei, X. Ma, X.S. Zheng, R. Hu, B. Ren, Surface-enhanced Raman spectroscopy for bioanalysis: reliability and challenges, *Chem. Rev.* 118 (2018) 4946–4980.
- [7] O. Herud-Sikimic, A.C. Stiel, M. Kolb, S. Shanmugaratnam, K.W. Berendzen, C. Feldhaus, B. Hocker, G. Jurgens, A biosensor for the direct visualization of auxin, *Nature* 592 (2021) 768–772.
- [8] G. Angelovski, B.J. Tickner, G. Wang, Opportunities and challenges with hyperpolarized bioresponsive probes for functional imaging using magnetic resonance, *Nat. Chem.* 15 (2023) 755–763.
- [9] A. Quijano-Rubio, H.W. Yeh, J. Park, H. Lee, R.A. Langan, S.E. Boyken, M. J. Lajoie, L. Cao, C.M. Chow, M.C. Miranda, J. Wi, H.J. Hong, L. Stewart, B. Oh, D. Baker, De novo design of modular and tunable protein biosensors, *Nature* 591 (2021) 482–487.
- [10] J.R. Sempionatto, J.A. Lasalde-Ramírez, K. Mahato, J. Wang, W. Gao, Wearable chemical sensors for biomarker discovery in the omics era, *Nat. Rev. Chem.* 6 (2022) 899–915.
- [11] H. Teymourian, A. Barfidokht, J. Wang, Electrochemical glucose sensors in diabetes management: an updated review (2010–2020), *Chem. Soc. Rev.* 49 (2020) 7671–7709.
- [12] X. Su, X. Liu, Y. Xie, M. Chen, H. Zhong, M. Li, Quantitative label-free SERS detection of trace fentanyl in biofluids with a freestanding hydrophobic plasmonic paper biosensor, *Anal. Chem.* 95 (2023) 3821–3829.
- [13] H. Yang, P. Wang, F. Geng, Q. Wu, F. Song, C. Ding, Copolymerization of zwitterionic sulfobetaine and hydrophobic acrylamide based antifouling electrochemical biosensors for detection of CA125 in clinical serum samples, *Sens. Actuators-B* 387 (2023), 133820.
- [14] J. Zhu, B. Yang, L. Peng, J. Wu, H. Hao, S. Lou, Target-triggered double fluorescent biosensors for rapid and sensitive detection of long-chain perfluorinated compounds using DNA probe and lysozyme fiber, *Sci. Total. Environ.* 860 (2023), 160496.
- [15] A. Shome, A. Das, A. Borbora, M. Dhar, U. Manna, Role of chemistry in bio-inspired liquid wettability, *Chem. Soc. Rev.* 51 (2022) 5452–5497.
- [16] Y. Dong, J. Li, C. Janiak, X.Y. Yang, Interfacial design for detection of a few molecules, *Chem. Soc. Rev.* 52 (2023) 779–794.
- [17] P. Sinha Mahapatra, R. Ganguly, A. Ghosh, S. Chatterjee, S. Lowrey, A. D. Sommers, C.M. Megaridis, Patterning wettability for open-surface fluidic manipulation: fundamentals and applications, *Chem. Rev.* 122 (2022) 16752–16801.
- [18] S. Miao, X. Cao, M. Lu, X. Liu, Tailoring micro/nano-materials with special wettability for biomedical devices, *Biomed. Technol.* 2 (2023) 15–30.
- [19] L. Wang, H. Li, X. Wang, X. Yang, C. Tian, D. Sun, L. Liu, J. Li, Modification of low-energy surfaces using bicyclic peptides discovered by phage display, *J. Am. Chem. Soc.* 145 (2023) 17613–17620.
- [20] F. Chen, Y. Wang, Y. Tian, D. Zhang, J. Song, C.R. Crick, C.J. Carmalt, I.P. Parkin, Y. Lu, Robust and durable liquid-repellent surfaces, *Chem. Soc. Rev.* 51 (2022) 8476–8583.
- [21] H. Zhu, Y. Huang, F. Xia, Environmentally friendly superhydrophobic osmanthus flowers for oil spill cleanup, *Appl. Mater. Today* 19 (2020), 100607.
- [22] K.B. Ramadi, J.C. McRae, G. Selsing, A. Su, R. Fernandes, M. Hickling, B. Rios, S. Babae, S. Min, D. Gwynne, N.Z. Jia, A. Aragon, K. Ishida, J. Kuosmanen, J. Jenkins, A. Hayward, K. Kamrin, G. Traverso, Bioinspired, ingestible electrochemical capsules for hunger-regulating hormone modulation, *Sci. Robot.* 8 (2023), eade9676.
- [23] T. Li, S. Yu, B. Sun, Y. Li, X. Wang, Y. Pan, C. Song, Y. Ren, Zhanxiang Zhang, K.T. V. Grattan, Z. Wu, J. Zhao, Bioinspired claw-engaged and biolubricated swimming microrobots creating active retention in blood vessels, *Sci. Adv.* 9 (2023), eadg4501.
- [24] X. Zhang, G. Chen, H. Zhang, L. Shang, Y. Zhao, Bioinspired oral delivery devices, *Nat. Rev. Bioeng.* 1 (2023) 208–225.
- [25] Y. Tan, J. Yang, Y. Li, X. Li, Q. Wu, Y. Fan, F. Yu, J. Cui, L. Chen, D. Wang, X. Deng, Liquid-pressure-guided superhydrophobic surfaces with adaptive adhesion and stability, *Adv. Mater.* 34 (2022) 2202167.
- [26] J. Ju, H. Bai, Y. Zheng, T. Zhao, R. Fang, L. Jiang, A multi-structural and multi-functional integrated fog collection system in cactus, *Nat. Commun.* 3 (2012) 1247.
- [27] W. Barthlott, C. Neinhuis, Purity of the sacred lotus, or escape from contamination in biological surfaces, *Planta* 202 (1997) 1–8.
- [28] L. Feng, S. Li, Y. Li, H. Li, L. Zhang, J. Zhai, Y. Song, B. Liu, L. Jiang, D. Zhu, Super-hydrophobic surfaces: from natural to artificial, *Adv. Mater.* 14 (2002) 1857–1860.
- [29] X. Xie, W. Feng, Reactive superhydrophobic surfaces for interlayer electrical connectivity in three-dimensional electronics, *Angew. Chem. Int. Ed.* (2023), e202302837.
- [30] X. Zhang, S. Ben, Z. Zhao, Y. Ning, Q. Li, Z. Long, C. Yu, K. Liu, L. Jiang, Lossless and directional transport of droplets on multi-bioinspired superwetting V-shape rails, *Adv. Funct. Mater.* 33 (2023) 2212217.
- [31] A. Shome, A. Das, A. Borbora, M. Dhar, U. Manna, Role of chemistry in bio-inspired liquid wettability, *Chem. Soc. Rev.* 51 (2022) 5452–5497.
- [32] L. Feng, Y. Zhang, J. Xi, Y. Zhu, N. Wang, F. Xia, L. Jiang, Petal effect: a superhydrophobic state with high adhesive force, *Langmuir* 24 (2008) 4114–4119.
- [33] J. Nickerl, R. Helbig, H.J. Schulz, C. Werner, C. Neinhuis, Diversity and potential correlations to the function of Collembola cuticle structures, *Zoomorphology* 132 (2013) 183–195.
- [34] A.K. Epstein, B. Pokroy, A. Seminara, J. Aizenberg, Bacterial biofilm shows persistent resistance to liquid wetting and gas penetration, *Proc. Natl. Acad. Sci. U. S. A.* 108 (2011) 995–1000.
- [35] R. Rakitov, S.N. Gorb, Brochosomal coats turn leafhopper (Insecta, Hemiptera, Cicadellidae) integument to superhydrophobic state, *P. Roy. Soc. B-Biol. Sci.* 280 (2013) 20122391.
- [36] R. Helbig, J. Nickerl, C. Neinhuis, C. Werner, Smart skin patterns protect springtails, *Plos One* 6 (2011) 25105.

- [37] R. Hensel, R. Helbig, S. Aland, H.G. Braun, A. Voigt, C. Neinhuis, C. Werner, Wetting resistance at its topographical limit: the benefit of mushroom and serif T structures, *Langmuir* 29 (2013) 1100–1112.
- [38] A.R. Parker, C.R. Lawrence, Water capture by a desert beetle, *Nature* 414 (2001) 33–34.
- [39] D. Wang, Q. Sun, M.J. Hokkanen, C. Zhang, F.Y. Lin, Q. Liu, S.P. Zhu, T. Zhou, Q. Chang, B. He, Q. Zhou, L. Chen, Z. Wang, R.H.A. Ras, X. Deng, Design of robust superhydrophobic surfaces, *Nature* 582 (2020) 55–59.
- [40] X. Deng, L. Mammen, H.J. Butt, D. Vollmer, Candle soot as a template for a transparent robust superamphiphobic coating, *Science* 335 (2012) 67–70.
- [41] H. Zhu, Y. Huang, X. Lou, F. Xia, Bioinspired superwetting surfaces for biosensing, *View* 2 (2021) 20200053.
- [42] J. Choi, S. Lee, K. Ohkawa, D.S. Hwang, Counterplotting the mechanosensing-based fouling mechanism of mussels against fouling, *ACS Nano* 15 (2021) 18566–18579.
- [43] L. Yang, Y. Feng, Z. He, X. Jiang, X. Luo, H. Dai, L. Jiang, Fast processing nylon mesh by surface diffuse atmospheric plasma for large-area oil/water separation, *Nano Res.* (2023), <https://doi.org/10.1007/s12274-023-5677-z>.
- [44] X. Yan, B. Ji, L. Feng, X. Wang, D. Yang, K.F. Rabbi, Q. Peng, M.J. Hoque, P. Jin, E. Bello, S. Sett, M. Aiieyne, D.M. Crokek, N. Miljkovic, Particulate-droplet coalescence and self-transport on superhydrophobic surfaces, *ACS Nano* 16 (2022) 12910–12921.
- [45] H.H. Vu, N.T. Nguyen, N. Kashaninejad, Re-entrant microstructures for robust liquid repellent surfaces, *Adv. Mater. Technol.* 8 (2023) 2201836.
- [46] J. Guo, W. Huang, Z. Guo, W. Liu, Design of a venation-like patterned surface with hybrid wettability for highly efficient fog harvesting, *Nano Lett.* 22 (2022) 3104–3111.
- [47] I. Haechler, N. Ferru, G. Schnoering, E. Mitridis, T.M. Schutzius, D. Poulikakos, Transparent sunlight-activated antifogging metamaterials, *Nat. Nanotechnol.* 18 (2023) 137–144.
- [48] Y. Jin, X. Liu, W. Xu, P. Sun, S. Huang, S. Yang, X. Yang, Q. Wang, R.H.W. Lam, R. Li, Z. Wang, Charge-powered electrostatics for versatile droplet manipulation, *ACS Nano* 17 (2023) 10713–10720.
- [49] J. Li, Y. Fu, J. Zhou, K. Ya, X. Ma, S. Gao, Z. Wang, J. Dai, D. Lei, X. Yu, Ultrathin, soft, radiative cooling interfaces for advanced thermal management in skin electronics, *Sci. Adv.* 9 (2023), eadg1837.
- [50] G. Liu, W.S.Y. Wong, M. Kraft, J.W. Ager, D. Vollmer, R. Xu, Wetting-regulated gas-involving (photo) electrocatalysis: biomimetics in energy conversion, *Chem. Soc. Rev.* 50 (2021) 10674–10699.
- [51] S. Zhou, L. Jiang, Z. Dong, Wetting-regulated gas-involving (photo) electrocatalysis: biomimetics in energy conversion, *Chem. Rev.* 123 (2023) 2276–2310.
- [52] S.H. Li, J.Y. Huang, Z. Chen, G.Q. Chen, Y.K. Lai, A review on special wettability textiles: theoretical models, fabrication technologies and multifunctional applications, *J. Mater. Chem. A* 5 (2017) 31–55.
- [53] H.Z. Li, A. Li, Z.P. Zhao, M.Z. Li, Y.L. Song, Heterogeneous wettability surfaces: principle, construction, and applications, *Small. Struct.* 1 (2020) 2000028.
- [54] L. Sun, J. Guo, H. Chen, D. Zhang, L. Shang, B. Zhang, Y. Zhao, Tailoring materials with specific wettability in biomedical engineering, *Adv. Sci.* 8 (2021) 2100126.
- [55] Y.W. Li, B.F. Liu, X.C. Zhang, Microfluidic chemostatic bioreactor for high-throughput screening and sustainable co-harvesting of biomass and biodiesel in microalgae, *Mater. Today* 51 (2021) 273–293.
- [56] Z.Q. Dong, P.A. Levkin, 3D Microprinting of super-repellent microstructures: recent developments, challenges, and opportunities, *Adv. Funct. Mater.* (2023), 2213916.
- [57] T. Xu, L.P. Xu, X. Zhang, S. Wang, Bioinspired superwetable micropatterns for biosensing, *Chem. Soc. Rev.* 48 (2019) 3153–3165.
- [58] Y.L. Wang, F. Liu, Y.M. Yang, L.P. Xu, Droplet evaporation-induced analyte concentration toward sensitive biosensing, *Mater. Chem. Front.* 5 (2021) 5639–5652.
- [59] Q. Zhu, Y. Yang, H. Gao, L.P. Xu, S. Wang, Bioinspired superwetable electrodes towards electrochemical biosensing, *Chem. Sci.* 13 (2022) 5069–5084.
- [60] A. Tuteja, W. Choi, M. Ma, J.M. Mabry, S.A. Mazzella, G.C. Rutledge, G. H. McKinley, R.E. Cohen, Designing superoleophobic surfaces, *Science* 318 (2007) 1618–1622.
- [61] J.M. Berg, L.T. Eriksson, P.M. Claesson, K.G.N. Borve, Three-component Langmuir-Blodgett films with a controllable degree of polarity, *Langmuir* 10 (1994) 1225–1234.
- [62] E. Kim, D. Kim, K. Kwak, Y. Nagata, M. Bonn, M. Cho, Wettability of graphene, water contact angle, and interfacial water structure, *Chem* 8 (2022) 1187–1200.
- [63] T. Young, III, An essay on the cohesion of fluids, *Philos. Trans. R. Soc. Lond.* 95 (1805) 65–87.
- [64] B. Su, Y. Tian, L. Jiang, Bioinspired interfaces with superwettability: from materials to chemistry, *J. Am. Chem. Soc.* 138 (2016) 1727–1748.
- [65] S. Zhou, L. Jiang, Z. Dong, Overflow control for sustainable development by superwetting surface with biomimetic structure, *Chem. Rev.* 123 (2023) 2276–2310.
- [66] J. Feng, Y. Qiu, L. Jiang, Y. Wu, Long-range-ordered assembly of micro-/nanostripes at superwetting interfaces, *Adv. Mater.* 34 (2022) 2106857.
- [67] M.A. Lemp, F.J. Holly, S. Iwata, C.H. Dohman, The precorneal tear film: I. Factors in spreading and maintaining a continuous tear film over the corneal surface, *Arch. Ophthalmol. - Chica.* 83 (1970) 89–94.
- [68] M. Liu, S. Wang, Z. Wei, Y. Song, L. Jiang, Bioinspired design of a superoleophobic and low adhesive water/solid interface, *Adv. Mater.* 21 (2009) 665–669.
- [69] J. Yong, F. Chen, Y. Fang, J. Huo, Q. Yang, J. Zhang, H. Bian, X. Hou, Bioinspired design of underwater superaerophobic and superaerophilic surfaces by femtosecond laser ablation for anti- or capturing bubbles, *ACS Appl. Mater. Interfaces* 9 (2017) 39863–39871.
- [70] G.D. Bixler, B. Bhushan, Bioinspired rice leaf and butterfly wing surface structures combining shark skin and lotus effects, *Soft. Matter* 8 (2012) 11271–11284.
- [71] D.M. Drotlef, L. Stepien, M. Kappl, W.J.P. Barnes, H.J. Butt, A. del Campo, Insights into the adhesive mechanisms of tree frogs using artificial mimics, *Adv. Funct. Mater.* 23 (2013) 1137–1146.
- [72] G. Wang, R. Han, Q. Li, Y. Han, X. Luo, Electrochemical biosensors capable of detecting biomarkers in human serum with unique long-term antifouling abilities based on designed multifunctional peptides, *Anal. Chem.* 92 (2020) 7186–7193.
- [73] S. Gao, J. Chen, Y. Zheng, A. Wang, D. Dong, Y. Zhu, Y. Zhang, W. Fang, J. Jin, Gradient adhesive hydrogel decorated superhydrophilic membranes for ultra-stable oil/water separation, *Adv. Funct. Mater.* 32 (2022) 2205990.
- [74] Q. Hao, Q. Xu, S. Niu, C. Ding, X. Luo, Anti-fouling magnetic beads combined with signal amplification strategies for ultra-sensitive and selective electrochemiluminescence detection of microRNAs in complex biological media, *Anal. Chem.* 93 (2021) 10679–10687.
- [75] G. Sun, P. Wang, Y. Jiang, H. Sun, T. Liu, G. Li, W. Yu, C. Meng, S. Guo, Bioinspired flexible, breathable, waterproof and self-cleaning iontronic tactile sensors for special underwater sensing applications, *Nano Energy* 110 (2023), 108367.
- [76] F. Tang, S. Li, H.Y. Yu, C. Wang, Y. Li, Z. Li, J. Yao, J. Tang, J. Zhu, Antioil Ag₃PO₄ nanoparticle/polydopamine/Al₂O₃ sandwich structure for complex wastewater treatment: dynamic catalysis under natural light, *ACS Sustain. Chem. Eng.* 8 (2020) 17458–17465.
- [77] Q. Zhang, N. Zhang, K. Li, X. Zhang, Y. Du, D. Tian, L. Jiang, Dynamically adjustable wet ridge for directional liquid movement and controllable coating distribution, *Chem. Eng. J.* 469 (2023), 143998.
- [78] A. Cassie, S. Baxter, Wettability of porous surfaces, *Trans. Faraday Soc.* 40 (1944) 546–551.
- [79] R.N. Wenzel, Resistance of solid surfaces to wetting by water, *Ind. Eng. Chem.* 28 (1936) 988–994.
- [80] A. Lafuma, D. Quere, Superhydrophobic states, *Nat. Mater.* 2 (2003) 457–460.
- [81] H. Zhao, W.C. Gao, Q. Li, M.R. Khan, G.H. Hu, Y. Liu, W. Wu, C.X. Huang, R.K. Y. Li, Recent advances in superhydrophobic polyurethane: preparations and applications, *Adv. Colloid Interfac.* 303 (2022), 102644.
- [82] M. Jin, X. Feng, L. Feng, T. Sun, J. Zhai, T. Li, L. Jiang, Superhydrophobic aligned polystyrene nanotube films with high adhesive force, *Adv. Mater.* 17 (2005) 1977–1981.
- [83] L. Gao, T.J. McCarthy, The “lotus effect” explained: two reasons why two length scales of topography are important, *Langmuir* 22 (2006) 2966–2967.
- [84] S. Pan, J.J. Richardson, A.J. Christofferson, Q.A. Besford, T. Zheng, B.J. Wood, X. Duan, M.J. J. fornerod, C.F.M. Conville, I. Yarovsky, S. Guldin, L. Jiang, F. Caruso, Fluorinated metal–organic coatings with selective wettability, *J. Am. Chem. Soc.* 143 (2021) 9972–9981.
- [85] P. Vineeth, A. Peethan, S.D. George, Special wettability for sensing: Drawing inspiration from nature, *Chem. Eng. J.* 459 (2023), 141615.
- [86] B. Li, W. Liang, B. Zhang, J. Zhang, Self-assembly fundamentals in the reconstruction of lignocellulosic materials: a review, *Colloids Surf. A* 672 (2023), 131759.
- [87] N. Abu Jarad, H. Imran, S.M. Imani, T.F. Didar, L. Soleymani, *Adv. Mater. Technol.* 7 (2022), 2101702.
- [88] X. Zhou, J. Liu, W. Liu, W. Steffen, H.J. Butt, Fabrication of superamphiphobic surfaces via spray coating: a review, *Adv. Mater.* 34 (2022) 2107901.
- [89] H.H. Vu, N.T. Nguyen, N. Kashaninejad, Re-entrant microstructures for robust liquid repellent surfaces, *Adv. Mater. Technol.* 8 (2023) 2201836.
- [90] T.S. Wong, S.H. Kang, S.K. Tang, E.J. Smythe, B.D. Hatton, A. Grinthal, J. Aizenberg, Bioinspired self-repairing slippery surfaces with pressure-stable omniphobicity, *Nature* 477 (2011) 443–447.
- [91] X.D. Lou, Y. Huang, X. Yang, H. Zhu, L.P. Heng, F. Xia, External stimuli responsive liquid-infused surfaces switching between slippery and nonslippery states: fabrications and applications, *Adv. Funct. Mater.* 30 (2020), 1901130.
- [92] W. Pan, Q. Wang, J. Ma, W. Xu, J. Sun, X. Lin, J. Song, Solid-like slippery coating with highly comprehensive performance, *Adv. Funct. Mater.* (2023), 2302311.
- [93] J.K. Hong, K. Mathur, A.M. Ruhoff, B. Akhavan, A. Waterhouse, C. Neto, Design optimization of perfluorinated liquid-infused surfaces for blood-contacting applications, *Adv. Mater. Interfaces* 9 (2022) 2102214.
- [94] L. Jinze, L. Yuhao, Z.F. Gao, Polydopamine-based colorimetric superwetable biosensor for highly sensitive detection of hydrogen peroxide and glucose, *J. Anal. Test.* 7 (2023) 118–127.
- [95] J. Huo, X. Bai, J. Yong, Y. Fang, Q. Yang, X. Hou, F. Chen, How to adjust bubble’s adhesion on solid in aqueous media: Femtosecond laser-ablated patterned shapememory polymer surfaces to achieve bubble multi-manipulation, *Chem. Eng. J.* 414 (2021), 128694.
- [96] N. Gao, L. Wang, Y. Zhang, F. Liang, Y. Fan, Modified ceramic membrane with pH/ethanol induced switchable superwettability for antifouling separation of oil-in-acidic water emulsions, *Sep. Purif. Technol.* 293 (2022), 121022.
- [97] Z. Wang, Y. Dai, C. Fang, L. Chen, Q. Lu, Y. Li, L. Cai, B. Liu, Y.F. Zhang, Y. Li, W. Li, A bio-inspired green method to fabricate pH-responsive sponge with switchable surface wettability for multitasking and effective oil-water separation, *Appl. Surf. Sci.* 602 (2022), 154192.
- [98] Z. Zhang, G. Dai, Y. Liu, W. Fan, K. Yang, Z. Li, A reusable, biomass-derived, and pH-responsive collagen fiber based oil absorbent material for effective separation of oil-in-water emulsions, *Colloids Surf. A* 633 (2022), 127906.

- [99] J. Liu, Z. Sheng, M. Zhang, J. Li, Y. Zhang, X. Xu, S. Yu, M. Cao, X. Hou, Non-Newtonian fluid gating membranes with acoustically responsive and self-protective gas transport control, *Mater. Horiz.* 10 (2023) 899–907.
- [100] W. Li, Y. Zhan, A. Amirfazli, A.R. Siddiqui, S. Yu, Recent progress in stimulus-responsive superhydrophobic surfaces, *Prog. Org. Coat.* 168 (2022), 106877.
- [101] L. Chu, W. Li, Y. Zhan, A. Amirfazli, Magnetically responsive superhydrophobic surfaces with droplet manipulation capability, *Adv. Eng. Mater.* 25 (2023) 2201352.
- [102] Y. Zhan, S. Yu, A. Amirfazli, A.R. Siddiqui, W. Li, Magnetically responsive superhydrophobic surfaces for microdroplet manipulation, *Adv. Mater. Interfaces* 9 (2022) 2102010.
- [103] C. Wei, Y. Zong, Y. Jiang, Bioinspired wire-on-pillar magneto-responsive superhydrophobic arrays, *ACS Appl. Mater. Inter.* 15 (2023) 24989–24998.
- [104] J. Zheng, B. Yang, H. Wang, L. Zhou, Z. Zhang, Z. Zhou, Temperature-responsive, femtosecond laser-ablated ceramic surfaces with switchable wettability for on-demand droplet transfer, *ACS Appl. Mater. Inter.* 15 (2023) 13740–13752.
- [105] Z.F. Gao, Y.X. Li, L.M. Dong, L.L. Zheng, Y. Shen, F. Xia, Photothermal-induced partial Leidenfrost superhydrophobic surface as ultrasensitive surface-enhanced Raman scattering platform for the detection of neonicotinoid insecticides, *Sens. Actuators-B* 348 (2021), 130728.
- [106] B. Qi, B.J. Fan, B. Xu, M. Zhou, Y.Y. Yu, L. Cui, Q. Wang, P. Wang, Enzymatic construction of temperature-responsive PDMAAPS-decorated textiles for oil-water separation, *Colloids Surf. A* 656 (2023), 130340.
- [107] Y. Yang, C. Li, Z. Gao, X. Qi, L. He, W. Huang, J. Wang, Z. Liu, Photo-responsive Mn-doped TiO₂-based superhydrophobic/underwater superoleophobicity membrane for efficient oil-water separation and photothermal decontamination, *Colloids Surf. A* 670 (2023), 131519.
- [108] J.J. Zhang, C. Nie, W.L. Fu, F.L. Cheng, P. Chen, Z.F. Gao, Y. Wu, Y. Shen, Photoresponsive DNA-modified magnetic bead-assisted rolling circle amplification-driven visual photothermal sensing of *Escherichia coli*, *Anal. Chem.* 94 (2022) 16796–16802.
- [109] S. Zhao, X. Yang, Y. Xu, Z. Weng, L. Liao, X. Wang, A sprayable superhydrophobic dental protectant with photo-responsive anti-bacterial, acid-resistant, and anti-fouling functions, *Nano Res.* 15 (2022) 5245–5255.
- [110] Y. Shao, W. Du, Y. Fan, J. Zhao, L. Ren, Near-infrared light accurately controllable superhydrophobic surface from water sticking to repelling, *Chem. Eng. J.* 427 (2022), 131718.
- [111] H. Zheng, H. Wu, Z. Yi, Y. Song, W. Xu, X. Zhou, S. Wang, Z. Wang, Remote-controlled droplet chains-based electricity generators, *Adv. Energy Mater.* 13 (2023) 2203825.
- [112] C. Chen, H. Yao, S. Guo, Z. Lao, Y. Xu, S. Li, S. Wu, Ultra-robust joule-heated superhydrophobic smart window: dually-switching droplets adhesion and transparency via in situ electric-actuated reconfigurable shape-memory shutters, *Adv. Funct. Mater.* 33 (2023) 2210495.
- [113] B. Xiao, C. Shen, Z. Luo, D. Li, X. Kuang, D. Wang, B. Zi, R. Yan, T. Lv, T. Zhou, J. Zhang, Q. Liu, Cu surface doped TiO₂: Constructing Cu single-atoms active sites and broadening the photo-response range for efficient photocatalytic hydrogen production, *Chem. Eng. J.* 468 (2023), 143650.
- [114] C. Jin, D. Sun, Z. Sun, S. Rao, Z. Wu, C. Cheng, L. Liu, Q. Liu, J. Yang, Interfacial engineering of Ni-phytate and Ti₃C₂T_x MXene-sensitized TiO₂ toward enhanced sterilization efficacy under 808 nm NIR light irradiation, *Appl. Catal. B-Environ.* 330 (2023), 122613.
- [115] Y. Ju, R. Liu, G. Ji, L. Su, J. Qiao, W. Xing, D. Fan, K. Zhao, D.D. Dionysiou, Novel strategy for enhanced visible light-responsive photoactivity of ZnFe₂O₄ with a single-mode microwave combustion process: primary parameters, *Chem. Eng. J.* 440 (2022), 135551.
- [116] J.H. Tzeng, C.H. Weng, C.J. Chang, L.T. Yen, M.D.G. deLuna, J.W. Huang, Y. T. Lin, N-Schorl TiO₂ nanocomposite for visible-light photocatalysis deactivation yeast exemplified by *Candida albicans*, *Chem. Eng. J.* 435 (2022), 134294.
- [117] C. Park, J.H. Hong, B.Y. Kim, S. An, S.S. Yoon, Supersonically sprayed copper oxide titania nanowires for antibacterial activities and water purification, *Appl. Surf. Sci.* 611 (2023), 155513.
- [118] T. Huo, F. Li, K. Jiang, W. Kong, X. Zhao, Y. Pan, Fluorocarbon-based selective-superwetting nanofibrous membranes with ultraviolet-driven switchable wettability for oil–water separation, *ACS Appl. Nano Mater.* 5 (2022) 13018–13026.
- [119] K. Liu, Z. Cai, X. Chi, B. Kang, S. Fu, X. Luo, Z.W. Lin, H. Ai, J. Gao, H. Lin, Photoinduced superhydrophilicity of Gd-Doped TiO₂ ellipsoidal nanoparticles boosts T1 contrast enhancement for magnetic resonance imaging, *Nano Lett.* 22 (2022) 3219–3227.
- [120] Q.F. Cheng, M.Z. Li, Y.M. Zheng, B. Su, S.T. Wang, L. Jiang, Janus interface materials: superhydrophobic air/solid interface and superoleophobic water/solid interface inspired by a lotus leaf, *Soft. Matter* 7 (2011) 5948–5951.
- [121] Y.Y. Zhao, C.M. Yu, H. Lan, M.Y. Cao, L. Jiang, Improved interfacial floatability of superhydrophobic/superhydrophilic Janus sheet inspired by lotus leaf, *Adv. Funct. Mater.* 27 (2017) 1701466.
- [122] D.K. Li, Y.F. Fan, G.C. Han, Z.G. Guo, Multibioinspired Janus membranes with superwettability performance for unidirectional transportation and fog collection, *Chem. Eng. J.* 404 (2021), 126515.
- [123] T. Huang, L. Zhang, J. Lao, K. Luo, X. Liu, K. Sui, J. Gao, L. Jiang, Reliable and low temperature actuation of water and oil slugs in Janus photothermal slippery tube, *ACS Appl. Mater. Interfaces* 14 (2022) 17968–17974.
- [124] L. Zhao, Y. Li, M. Yu, Y. Peng, F. Ran, Electrolyte-wettability issues and challenges of electrode materials in electrochemical energy storage, energy conversion, and beyond, *Adv. Sci.* 10 (2023) 2300283.
- [125] C. Liu, Y. Peng, C. Huang, Y. Ning, J. Shang, Y. Li, Bioinspired superhydrophobic/superhydrophilic Janus copper foam for on-demand oil/water separation, *ACS Appl. Mater. Inter.* 14 (2022) 11981–11988.
- [126] X. He, C. Fan, T. Xu, X. Zhang, Biospired Janus silk E-textiles with wet–thermal comfort for highly efficient biofluid monitoring, *Nano Lett.* 21 (2021) 8880–8887.
- [127] M. Wang, H. Zhou, H. Du, L. Chen, G. Zhao, H. Liu, X. Jin, W. Chen, A. Ma, A cyclic freezing-thawing approach to layered Janus hydrogel tapes with single-sided adhesiveness for wearable strain sensors, *Chem. Eng. J.* 446 (2022), 137163.
- [128] J. Sazcek, X. Yao, V. Zivkovic, M. Mamlouk, D. Wang, S.S. Pramana, S. Wang, Long-lived liquid marbles for green applications, *Adv. Funct. Mater.* 31 (2021) 2011198.
- [129] K.D. Kersey, G.A. Lee, J.H. Xu, M.K. Kidder, A.A. Park, Y.L. Joo, Encapsulation of nanoparticle organic hybrid materials within electrospun hydrophobic polymer/ceramic fibers for enhanced CO₂ capture, *Adv. Funct. Mater.* (2023), 2301649.
- [130] F. Ahmad, M.M. Salem-Bekhit, F. Khan, S. Alshehri, A. Khan, M.M. Ghoneim, H. F. Wu, E.I. Taha, I. Elbagory, Unique properties of surface-functionalized nanoparticles for bio-application: functionalization mechanisms and importance in application, *Nanomaterials* 12 (2022) 1333.
- [131] J.C. Liu, L. Luo, H. Xiao, J. Zhu, Y. He, J. Li, Metal affinity of support dictates sintering of gold catalysts, *J. Am. Chem. Soc.* 144 (2022) 20601–20609.
- [132] M. Tenjimbayashi, S. Samitsu, Y. Watanabe, Y. Nakamura, M. Naito, Liquid marble patchwork on super-repellent surface, *Adv. Funct. Mater.* 31 (2021) 2010957.
- [133] Y. Zhang, H. Cui, B.P. Binks, H.C. Shum, Liquid marbles under electric fields: new capabilities for non-wetting droplet manipulation and beyond, *Langmuir* 38 (2022) 9721–9740.
- [134] J. Zhang, Y. Gu, J. Jiang, R. Zheng, pH-responsive liquid marbles based on dihydroxystearic acid, *Langmuir* 38 (2022) 5702–5707.
- [135] Y. Tsumura, K. Oyama, A.L. Fameau, M. Seike, A. Ohtaka, T. Hirai, Y. Nakamura, S. Fujii, Photo/thermo dual stimulus-responsive liquid marbles stabilized with polypyrrole-coated stearic acid particles, *ACS Appl. Mater. Inter.* 14 (2022) 41618–41628.
- [136] Z. Zhao, X. Yao, W. Zhao, B. Shi, S. Sridhar, Y. Pu, S. Pramama, D. Wang, S. Wang, Highly transparent liquid marble in liquid (HT-LML) as 3D miniaturized reactor for real-time bio-/chemical assays, *Chem. Eng. J.* 443 (2022), 136417.
- [137] Y. Sun, Y. Zheng, C. Liu, Y. Zhang, S. Wen, L. Song, M. Zhao, Liquid marbles, floating droplets: preparations, properties, operations and applications, *RSC Adv.* 12 (2022) 15296–15315.
- [138] D. Dedovets, Q. Li, L. Leclercq, V. Nardello-Rataj, J. Leng, S. Zhao, P.M. Perattus, Multiphase microreactors based on liquid–liquid and gas–liquid dispersions stabilized by colloidal catalytic particles, *Angew. Chem. Int. Ed.* 61 (2022), e202107537.
- [139] T. Arbatan, L.Z. Li, J.F. Tian, W. Shen, Liquid marbles as micro-bioreactors for rapid blood typing, *Adv. Healthc. Mater.* 1 (2012) 80–83.
- [140] N. Phillips, R. Mayne, A. Adamatzky, *Chlorella* sensors in liquid marbles and droplets, *Sens. Bio-Sens. Res.* 36 (2022), 100491.
- [141] W. Zhang, N. Srichan, A.F. Chirimes, M. Taylor, K.J. Berean, J.Z. Ou, T. Daenke, A.P. O'Mullane, G. Bryant, K. Kalantar-zadeh, Sonication synthesis of micro-sized silver nanoparticle/oleic acid liquid marbles: a novel SERS sensing platform, *Sens. Actuators-B* 223 (2016) 52–58.
- [142] J.H. Bahng, B. Yeom, Y. Wang, S.O. Tung, J.D. Hoff, N. Kotov, Anomalous dispersions of 'hedgehog' particles, *Nature* 517 (2015) 596–599.
- [143] Z. Zhu, Z. Guan, S. Jia, Z. Lei, S. Lin, H. Zhang, Y. Ma, Z.Q. Tian, C.J. Yang, Au@Pt nanoparticle encapsulated target-responsive hydrogel with volumetric bar-chart chip readout for quantitative point-of-care testing, *Angew. Chem. Int. Ed.* 53 (2014) 12503–12507.
- [144] R. Liu, Y. Huang, Y. Ma, S. Jia, M. Gao, J. Li, H. Zhang, D. Xu, M. Wu, Y. Chen, Z. Zhu, C. Yang, Design and synthesis of target-responsive aptamer-cross-linked hydrogel for visual quantitative detection of ochratoxin A, *ACS Appl. Mater. Inter.* 7 (2015) 6982–6990.
- [145] X. Wei, T. Tian, S. Jia, Z. Zhu, Y. Ma, J. Sun, Z. Lin, C.J. Yang, Target-responsive DNA hydrogel mediated "stop-flow" microfluidic paper-based analytic device for rapid, portable and visual detection of multiple targets, *Anal. Chem.* 87 (2015) 4275–4282.
- [146] X. Wei, T. Tian, S. Jia, Z. Zhu, Y. Ma, J. Sun, Z. Lin, C.J. Yang, Microfluidic distance readout sweet hydrogel integrated paper-based analytical device (μ DiSH-PAD) for visual quantitative point-of-care testing, *Anal. Chem.* 88 (2016) 2345–2352.
- [147] Y. Li, Y. Ma, X. Jiao, T. Li, Z. Lv, C.J. Yang, X. Zhang, Y. Wen, Control of capillary behavior through target-responsive hydrogel permeability alteration for sensitive visual quantitative detection, *Nat. Commun.* 10 (2019) 1036.
- [148] Y. Pan, C. Zhu, W.B. Zeng, P. Fu, C. Chen, B.M. Xu, Z.F. Gao, Visual detection of adenosine triphosphate by Taylor rising: a simple point-of-care testing method based on rolling circle amplification, *Chembiochem* 22 (2021) 3431–3436.
- [149] J.Z. Li, L.M. Dong, L.L. Zheng, W.L. Fu, J.J. Zhang, L. Zhang, Q. Hu, P. Chen, Z. F. Gao, F. Xia, Molecular visual sensing, Boolean logic computing, and data security using a droplet-based superwetting paradigm, *ACS Appl. Mater. Inter.* 14 (2022) 40447–40459.
- [150] Z.F. Gao, L.L. Zheng, L.M. Dong, J.Z. Li, Y. Shen, P. Chen, F. Xia, Label-free resonance Rayleigh scattering amplification for lipopolysaccharide detection and logical circuit by CRISPR/Cas12a-driven guanine nanowire assisted non-cross-linking hybridization chain reaction, *Anal. Chem.* 94 (2022) 6371–6379.
- [151] L.L. Zheng, J.Z. Li, M. Wen, D. Xi, Y. Zhu, Q. Wei, X.B. Zhang, G. Ke, F. Xia, Z. F. Gao, Enthalpy and entropy synergistic regulation-based programmable DNA motifs for biosensing and information encryption, *Sci. Adv.* 9 (2023), ead5868.

- [152] Z. Zhang, L.P. Wen, L. Jiang, Nanofluidics for osmotic energy conversion, *Nat. Rev. Mater.* 6 (2021) 622–639.
- [153] S. Xu, W.C. Li, C. Wang, S. Wang, H. Ma, G.P. Hao, A.H. Lu, Targeted synthesis of anti-hydrolysis 2D-ZIF laminates with super-hydrophobic transport channels via in situ phase transition strategy, *Adv. Funct. Mater.* 32 (2022) 2112947.
- [154] S.P. Zheng, L.B. Huang, Z. Sun, M. Barboiu, Self-assembled artificial ion-channels toward natural selection of functions, *Angew. Chem. Int. Ed.* 60 (2021) 566–597.
- [155] Y.L. Ying, Z.L. Hu, S. Zhang, Y. Qing, A. Fragasso, G. Maglia, A. Meller, H. Bayley, C. Dekker, Y.T. Long, Nanopore-based technologies beyond DNA sequencing, *Nat. Nanotechnol.* 17 (2022) 1136–1146.
- [156] D.N. Philpott, K. Chen, R.S. Atwal, D. Li, J. Christie, E.H. Sargent, S.O. Kelley, Ultrahigh-throughput immunomagnetic cell sorting platform, *Lab. Chip* 22 (2022) 4822–4830.
- [157] D. Zhou, S. Cai, H. Sun, G. Zhong, H. Zhang, D. Sun, F. Su, M. Deng, Y. Tian, Diatom frustules based dissolved oxygen sensor with superhydrophobic surface, *Sens. Actuators-B* 371 (2022), 132549.
- [158] P. Fang, J. Li, F. Jiang, J. Meng, H. Pan, Superwetable dendritic gold nanostructured electrode arrays for electrochemical enzyme-linked immunosorbent assay (ELISA), *Int. J. Electrochem. Sc.* 17 (2022), 220828.
- [159] M. Yang, C. Wang, Y. Wei, C. Liu, F. Lei, X. Zhao, Z. Li, C. Zhang, J. Yu, Construct high-precise SERS sensor by hierarchical superhydrophobic Si/Cu(OH)₂ platform for ultratrace detection of food contaminants, *Sens. Actuators-B* 352 (2022), 131056.
- [160] X. Luo, Z. Tian, C.H. Chen, G.C. Jiang, X.Y. Hu, L.Z. Wang, R. Peng, H.J. Zhang, M.L. Zhong, Laser-textured high-throughput hydrophobic/superhydrophobic SERS platform for fish drugs residue detection, *Opt. Laser Technol.* 152 (2022), 108075.
- [161] L.P. Xu, Y. Chen, G. Yang, W. Shi, B. Dai, G. Li, Y. Cao, Y. Wen, X. Zhang, S. Wang, Ultratrace DNA detection based on the condensing-enrichment effect of superwetable microchips, *Adv. Mater.* 27 (2015) 6878–6884.
- [162] A. Borbora, M. Dhar, A. Shome, N. Barman, S. Roy, U. Manna, Chemically customizing mechanical properties and optical transparency in underwater superoleophobic coating, *Adv. Funct. Mater.* (2023), 2302569.
- [163] D. Yu, J. Huang, Z. Zhang, J. Weng, X. Xu, G. Zhang, J. Zhang, X. Xu, M. Johnson, J. Lyu, H. Yang, W. Wang, Simultaneous realization of superoleophobicity and strong substrate adhesion in water via a unique segment orientation mechanism, *Adv. Mater.* 34 (2022) 2106908.
- [164] J.K. Gao, M.M. Cai, Z.G. Nie, J.W. Zhang, Y. Chen, Simultaneous realization of superoleophobicity and strong substrate adhesion in water via a unique segment orientation mechanism, *Sep. Purif. Technol.* 275 (2021), 119174.
- [165] F. Li, J. Wang, Z. Wang, D. Ji, S. Wang, P. Wei, W. Cao, Bio-inspired eco-friendly superhydrophilic/underwater superoleophobic cotton for oil-water separation and removal of heavy metals, *Biomimetics* 7 (2022) 177.
- [166] L. Cao, L. Wu, C. Li, Y. Tu, H. Wu, B. Shen, J. Meng, X.Q. Hao, B. Yan, F. Li, F. Xia, Y. Huang, Underwater superoleophobic-oleophilic chips for femtomolar aflatoxins identification, *Chin. J. Chem.* 40 (2022) 1464–1470.
- [167] Z. Tong, L. Song, S. Chen, J. Hu, Q. Liu, Y. Ren, X. Zhan, Q. Zhang, Hagfish-inspired smart SLIPS marine antifouling coating based on supramolecular: lubrication modes responsively switching and self-healing properties, *Adv. Funct. Mater.* 32 (2022) 2201290.
- [168] L. Xia, S. Zhang, Z. Guo, Multifunction of biomimetic liquid infused systems derived from SLIPS theory: a review, *Adv. Mater. Inter.* 10 (2023), 2202212.
- [169] S. Wang, Y. Zhang, Y. Han, Y. Hou, Y. Fan, X. Hou, Design of porous membranes by liquid gating technology, *Acc. Mater. Res.* 2 (2021) 407–419.
- [170] J. Zhang, B. Chen, X. Chen, X. Hou, Liquid-based adaptive structural materials, *Adv. Mater.* 33 (2021), e2005664.
- [171] S. Yu, Y. Jing, Y. Fan, L. Xiong, H. Wang, J. Lei, Y. Zhang, J. Liu, S. Wang, X. Chen, H. Sun, X. Hou, Ultrahigh efficient emulsification with drag-reducing liquid gating interfacial behavior, *P. Natl. Acad. Sci. U. S. A.* 119 (2022), e2206462119.
- [172] Y. Zhang, X. Hou, Liquid-based materials, *Nat. Sci. Open.* 1 (2022) 20220035.
- [173] R.M. Shah, A. Cihanoglu, J. Hardcastle, C. Howell, J.D. Schiffman, Liquid-infused membranes exhibit stable flux and fouling resistance, *ACS Appl. Mater. Inter.* 14 (2022) 6148–6156.
- [174] H. Wang, Y. Fan, Y. Hou, B. Chen, J. Lei, S. Yu, X. Chen, X. Hou, Host-guest liquid gating mechanism with specific recognition interface behavior for universal quantitative chemical detection, *Nat. Commun.* 13 (2022) 1906.
- [175] R. Gulfam, Y. Chen, Recent growth of wettability gradient surfaces: a review, *Research* 2022 (2022) 9873075.
- [176] K. Kumar, A. Legge, D.A. Gregory, A. Nichols, H. Jemse, S.J. Ebbens, X. Zhao, 3D printable self-propelling sensors for the assessment of water quality via surface tension, *JCIS Open.* 5 (2022), 100044.
- [177] F. Huang, Y. Chen, Y. Wang, F. Xia, Tunable superamphiphobic surfaces: a platform for naked-eye ATP detection, *Anal. Bioanal. Chem.* 411 (2019) 4721–4727.
- [178] C. Yang, Q. Zeng, J. Huang, Z. Guo, Droplet manipulation on superhydrophobic surfaces based on external stimulation: a review, *Adv. Colloid Interfac.* 306 (2022), 102724.
- [179] J. Bai, R.T. Gao, N.T. Nguyen, X. Liu, X. Zhang, L. Wang, Heterogeneous doping via charge carrier transport improves photoelectrochemical H₂O oxidative H₂O₂ synthesis, *Chem. Eng. J.* 466 (2023), 142984.
- [180] H. Kim, Y. Kang, B. Lim, K. Kim, J. Yoon, A. Ali, S.R. Torati, C. Kim, Tailoring matter orbitals mediated using a nanoscale topographic interface for versatile colloidal current devices, *Mater. Horiz.* 9 (2022) 2353–2363.
- [181] F.K. Alosaimi, T.T. Tung, V.D. Dao, N.K. Huyen, M.J. Nine, K. Hassan, J. Ma, D. Losic, Graphene-based multifunctional surface and structure gradients engineered by atmospheric plasma, *Appl. Mater. Today* 27 (2022), 101486.
- [182] Y. Chen, K. Li, S. Zhang, L. Qin, S. Deng, L. Ge, L.P. Xu, L. Ma, S. Wang, X. Zhang, Bioinspired superwetable microspine chips with directional droplet transportation for biosensing, *ACS Nano* 14 (2020) 4654–4661.
- [183] Z. Cai, F. Chen, Y. Tian, D. Zhang, Z. Lian, M. Cao, Programmable droplet transport on multi-bioinspired slippery surface with tridirectionally anisotropic wettability, *Chem. Eng. J.* 449 (2022), 137831.
- [184] S. Yadav, A. Majumder, Biomimicked large-area anisotropic grooves from *Dracaena sanderiana* leaf enhances cellular alignment and subsequent differentiation, *Bioinspir. Biomim.* 17 (2022), 056002.
- [185] D. Kwiatkowska, Plant biology: How the humble plant droops its leaves, *Curr. Biol.* 33 (2023) R156–R158.
- [186] M. Tenjimbayashi, K. Manabe, A review on control of droplet motion based on wettability modulation: principles, design strategies, recent progress, and applications, *Sci. Technol. Adv. Mat.* 23 (2022) 473–497.
- [187] S. Cheng, C. Huang, W. Chen, P. Zhang, Directional superspreading of water droplets on grooved hydrogel surfaces for open microfluidic platforms, *Small Methods* (2023), 2300221.
- [188] S.R. Narayanasamy, R. Vasireddi, H.Y.N. Holman, M. Trebbin, A sui generis whipping-instability-based self-sequencing multi-monodisperse 2D spray from an anisotropic microfluidic liquid jet device, *Cell Rep. Phys. Sci.* 4 (2023), 101221.
- [189] R. Feng, F. Song, Y.D. Zhang, X.L. Wang, Y.Z. Wang, A confined-etching strategy for intrinsic anisotropic surface wetting patterning, *Nat. Commun.* 13 (2022) 3078.
- [190] T. Guo, P. Che, L. Heng, L. Fan, L. Jiang, Anisotropic slippery surfaces: electric-driven smart control of a drop's slide, *Adv. Mater.* 28 (2016) 6999–7007.
- [191] X. Yang, J. Wang, Z.F. Gao, W. Zhang, H. Zhu, Y. Song, Q. Wang, M. Liu, L. Jiang, Y. Huang, F. Xia, An orthogonal dual-regulation strategy for sensitive biosensing applications, *Natl. Sci. Rev.* 9 (2022), nwac048.
- [192] Z.Z. Jiao, H. Zhou, X.C. Han, D.D. Han, Y.L. Zhang, Photothermal responsive slippery surfaces based on laser-structured graphene@PVDF composites, *J. Colloid Interf. Sci.* 629 (2023) 582–592.
- [193] C. Yang, Y. Yu, X. Wang, Y. Zu, Y. Zhao, L. Shang, Bioinspired stimuli-responsive spindle-knotted fibers for droplet manipulation, *Chem. Eng. J.* 451 (2023), 138669.
- [194] Z.F. Gao, E.E. Sann, X. Lou, R. Liu, J. Dai, X. Zuo, F. Xia, L. Jiang, Naked-eye point-of-care testing platform based on a pH-responsive superwetting surface: toward the non-invasive detection of glucose, *NPG Asia. Mater.* 10 (2018) 177–189.
- [195] J.B. Gao, E.E. Sann, X.Y. Wang, C. Ye, R. Liu, Z.F. Gao, Visual detection of the prostate specific antigen via a sandwich immunoassay and by using a superwetable chip coated with pH-responsive silica nanoparticles, *Microchim. Acta* 186 (2019) 550.
- [196] M. Zhou, Z. Wang, D. Xia, X. Xie, Y. Chen, Y. Xing, K. Cai, J. Zhang, Hybrid nanoassembly with two-tier host-guest architecture and regioselective enrichment capacity for repetitive SERS detection, *Sens. Actuators-B* 369 (2022), 132359.
- [197] H. Zhang, S. Zhang, G. Li, H. Qu, W. Xu, Q. Song, H. Li, Dynamic spreading of insecticidal pesticide droplets on superhydrophobic plant leaves through host-guest chemistry, *ACS Agric. Sci. Technol.* 3 (2022) 158–164.
- [198] S. Sabuncu, R.J. Ramirez, J.M. Fischer, F. Civitci, A. Yildirim, Ultrafast background-free ultrasound imaging using blinking nanoparticles, *Nano Lett.* 23 (2023) 659–666.
- [199] X. Chen, Q. Ding, C. Bi, J. Ruan, S. Yang, Lossless enrichment of trace analytes in levitating droplets for multiphase and multiplex detection, *Nat. Commun.* 13 (2022) 7807.
- [200] S. Weng, L. Tang, M. Qiu, J. Wang, Y. Wu, R. Zhu, C. Wang, P. Li, W. Sha, D. Liang, Surface-enhanced Raman spectroscopy charged probes under inverted superhydrophobic platform for detection of agricultural chemicals residues in rice combined with lightweight deep learning network, *Anal. Chim. Acta* 1262 (2023), 341264.
- [201] C. Cai, Y. Liu, Z. Zhang, T. Tian, Y. Wang, L. Wang, K. Zhang, B. Liu, Activity-based self-enriched SERS sensor for blood metabolite monitoring, *ACS Appl. Mater. Inter.* 15 (2023) 4895–4902.
- [202] R.D. Deegan, O. Bakajin, T.F. Dupont, G. Huber, S.R. Nagel, T.A. Witten, Capillary flow as the cause of ring stains from dried liquid drops, *Nature* 389 (1997) 827–829.
- [203] L. Scriven, C. Sternling, The marangoni effects, *Nature* 187 (1960) 186–188.
- [204] P.J. Yunker, T. Still, M.A. Lohr, A.G. Yodh, Suppression of the coffee-ring effect by shape-dependent capillary interactions, *Nature* 476 (2011) 308–311.
- [205] M.W. Yang, D.J. Chen, J. Hu, X.Y. Zheng, Z.J. Lin, H.M. Zhu, The application of coffee-ring effect in analytical chemistry, *TrAC-Trend. Anal. Chem.* 157 (2022), 116752.
- [206] X. He, T. Xu, Z. Gu, W. Gao, L.P. Xu, T. Pan, X. Zhang, Flexible and superwetable bands as a platform toward sweat sampling and sensing, *Anal. Chem.* 91 (2019) 4296–4300.
- [207] Q. Ding, J. Wang, X. Chen, H. Liu, Q. Li, Y. Wang, S. Yang, Quantitative and sensitive SERS platform with analyte enrichment and filtration function, *Nano Lett.* 20 (2020) 7304–7312.
- [208] Y. Zhu, Y.X. Zhang, W.W. Liu, Y. Ma, Q. Fang, B. Yao, Printing 2-dimensional droplet array for single-cell reverse transcription quantitative PCR assay with a microfluidic robot, *Sci. Rep.* 5 (2015) 9551.
- [209] L. Shen, J. Zhang, Q. Yang, N.E. Manicke, Z. Ouyang, High throughput paper spray mass spectrometry analysis, *Clin. Chim. Acta* 420 (2013) 28–33.
- [210] Y. Yang, W. Wang, H. Liu, L. Tong, X. Mu, Z. Chen, B. Tang, Sensitive quantification of MicroRNA in blood through multi-amplification toehold-

- mediated dna-strand-displacement paper-spray mass spectrometry (TSD-PS MS), *Angew. Chem.* 134 (2022), e202113051.
- [211] Z. Chen, Q. Shi, W. Wang, Z. Jiang, G.L. Zhang, L. Tong, X. Mu, B. Tang, Fabrication of a "selenium signature" chemical probe-modified paper substrate for simultaneous and efficient determination of biothiols by paper spray mass spectrometry, *Anal. Chem.* 93 (2020) 1749–1756.
- [212] P. Basuri, A. Baidya, T. Pradeep, Sub-Parts-per-Trillion level detection of analytes by superhydrophobic preconcentration paper spray ionization mass spectrometry (SHPPSI MS), *Anal. Chem.* 91 (2019) 7118–7124.
- [213] B. Li, Y. Guo, Y. Jiang, J.M. Lin, Q. Hu, L. Yu, A pendant droplet-based sensor for the detection of acetylcholinesterase and its inhibitors, *Chem. Commun.* 57 (2021) 8909–8912.
- [214] I. Marica, M. Stefan, S. Boca, A. Falamaş, C. Farcău, A simple approach for coffee-ring suppression yielding homogeneous drying patterns of ZnO and TiO₂ nanoparticles, *J. Colloid Inter. Sci.* 635 (2023) 117–127.
- [215] N.S. Howard, A.J. Archer, D.N. Sibley, D.J. Southee, K.G.U. Wijayantha, Surfactant control of coffee ring formation in carbon nanotube suspensions, *Langmuir* 39 (2023) 929–941.
- [216] B. Zhao, L. Qi, W. Tai, M. Zhao, X. Chen, L. Yu, J. Shi, X. Wang, J.M. Lin, Q. Hu, Paper-based flow sensor for the detection of hyaluronidase via an enzyme hydrolysis-induced viscosity change in a polymer solution, *Anal. Chem.* 94 (2022) 4643–4649.
- [217] B. Zhao, M. Khan, Y. Liu, W. Tai, C. Mu, W. Wu, M. Zhao, Y. Ma, L. Yu, J.M. Lin, Q. Hu, Distance-based α -amylase biosensor fabricated with amylopectin-coated mesoporous membrane, *Chin. Chem. Lett.* (2023), 108462.
- [218] R. Salahandish, F. Haghayegh, S. Khetani, M. Hassani, A.S. Nezhad, *ACS Appl. Mater. Inter.* 14 (2022) 28651–28662.
- [219] Y. Sun, Y. Song, H. Sun, Q. Tian, Q. Wang, Y. Liu, S. Zhang, Immuno-affinity potent strip with pre-embedded intermixed PEDOT: PSS conductive polymers and graphene nanosheets for bio-ready electrochemical biosensing of central nervous system injury biomarkers, *ACS Appl. Nano Mater.* 5 (2022) 11080–11090.
- [220] M. Yang, D. Chen, J. Hu, X. Zheng, Z.J. Lin, H. Zhu, The application of coffee-ring effect in analytical chemistry, *TrAC-Trend, Anal. Chem.* 157 (2022), 116752.
- [221] C. Ni, J. Zhao, X. Xia, Z. Wang, X. Zhao, J. Yang, N. Zhang, Y. Yang, H. Zhang, D. Gao, Constructing a ring-like self-aggregation SERS sensor with the coffee ring effect for ultrasensitive detection and photocatalytic degradation of the herbicides paraquat and diquat, *J. Agr. Food Chem.* 70 (2022) 15296–15310.
- [222] J. Wang, Y. Huang, K. You, X. Yang, Y. Song, H. Zhu, F. Xia, L. Jiang, Temperature-driven precise control of biological droplet's adhesion on a slippery surface, *ACS Appl. Mater. Inter.* 11 (2019) 7591–7599.
- [223] M.A. Vratsanos, W. Xue, N.D. Rosenmann, L.D. Zarzar, N.C. Gianneschi, Ouzo effect examined at the nanoscale via direct observation of droplet nucleation and morphology, *ACS Cent. Sci.* 9 (2023) 457–465.
- [224] E. Middha, C. Chen, P.N. Manghnani, S. Wang, S. Zhen, Z. Zhao, B. Liu, Synthesis of uniform polymer encapsulated organic nanocrystals through ouzo nanocrystallization, *Small Methods* 6 (2022) 2100808.
- [225] L. Thayyil Raju, O. Koshkina, H. Tan, A. Riedinger, K. Landfester, D. Lohse, X. Zhang, Particle size determines the shape of supraparticles in self-lubricating ternary droplets, *ACS Nano* 15 (2021) 4256–4267.
- [226] Z. Huang, E. Calicchia, I. Jurewicz, E. Munoz, R. Garriga, G. Portale, B.J. owlin, J. L. Keddie, Two-dimensional triblock peptide assemblies for the stabilization of Pickering emulsions with pH responsiveness, *ACS Appl. Mater. Inter.* 14 (2022) 53228–53240.
- [227] T.T. Wu, T.L. Xu, Y.X. Chen, Y.M. Yang, L.P. Xu, X.J. Zhang, S.T. Wang, Renewable superwetable biochip for miRNA detection, *Sens. Actuators-B* 258 (2018) 715–721.
- [228] Z.F. Gao, R. Liu, J.H. Wang, J. Dai, W.H. Huang, M.J. Liu, S.T. Wang, F. Xia, L. Jiang, Controlling droplet motion on an organogel surface by tuning the chain length of DNA and its biosensing application, *Chem* 4 (2018) 2929–2943.
- [229] Y. Song, T. Xu, L.P. Xu, X. Zhang, Superwetable nanodendritic gold substrates for direct miRNA SERS detection, *Nanoscale* 10 (2018) 20990–20994.
- [230] H. Gao, X. Wan, Y. Yang, J. Lu, Q. Zhu, L.P. Xu, S. Wang, Leaf-inspired patterned organohydrogel surface for ultrawide time-range open biosensing, *Adv. Sci.* 10 (2023) 2207702.
- [231] C. Xu, Q. Liu, S. Chu, P. Li, F. Wang, Y. Si, G. Mao, C. Wu, H. Wang, A microdots array-based fluorometric assay with superwettability profile for simultaneous and separate analysis of iron and copper in red wine, *Anal. Chim. Acta* 1254 (2023), 341045.
- [232] J.H. Monserud, D.K. Schwartz, Effects of molecular size and surface hydrophobicity on oligonucleotide interfacial dynamics, *Biomacromolecules* 13 (2012) 4002–4011.
- [233] J.H. Monserud, D.K. Schwartz, Mechanisms of surface-mediated DNA hybridization, *ACS Nano* 8 (2014) 4488–4499.
- [234] Z.F. Gao, R. Liu, J. Wang, J. Dai, W.H. Huang, M. Liu, S. Wang, F. Xia, S. Zhang, L. Jiang, Manipulating the hydrophobicity of DNA as a universal strategy for visual biosensing, *Nat. Protoc.* 15 (2020) 316–337.
- [235] H. Yousefi, S.E. Samani, S. Khan, A. Prasad, A. Shakeri, Y. Li, C.D.M. Filipe, T. F. Didar, LISzyme biosensors: DNAszymes embedded in an anti-biofouling platform for hands-free real-time detection of bacterial contamination in milk, *ACS Nano* 16 (2022) 29–37.
- [236] A. Shakeri, N.A. Jarad, J. Terryberry, S. Khan, A. Leung, S. Chen, T.F. Didar, Antibody micropatterned lubricant-infused biosensors enable sub-picogram immunofluorescence detection of interleukin 6 in human whole plasma, *Small* 16 (2020), e2003844.
- [237] H. Yousefi, M.M. Ali, H.M. Su, C.D.M. Filipe, T.F. Didar, Sentinel wraps: real-time monitoring of food contamination by printing DNAszyme probes on food packaging, *ACS Nano* 12 (2018) 3287–3294.
- [238] Y. Luo, J. Ju, X. Yao, Advanced solar desalination on superwetting surfaces, *J. Mater. Chem. A* 10 (2022) 19348–19366.
- [239] J. Li, C. Lv, J. Song, X. Zhang, X. Huang, Y. Ma, H. Cao, N. Liu, Superwetting Ag/ α -Fe₂O₃ anchored mesh with enhanced photocatalytic and antibacterial activities for efficient water purification, *Green. Energy Environ.* (2022). Doi: 10.1016/j.gee.2022.05.005.
- [240] A. Peethan, M. Aravind, V.K. Unnikrishnan, S. Chidangil, S.D. George, Facile fabrication of plasmonic wettability contrast paper surface for droplet array-based SERS sensing, *Appl. Surf. Sci.* 571 (2022), 151188.
- [241] Z. Zhang, Y. Wang, Z. Mei, Y. Wang, H. Li, S. Li, F. Xia, Incorporating hydrophobic moieties into self-assembled monolayers to enable electrochemical aptamer-based sensors deployed directly in a complex matrix, *ACS Sens* 7 (2022) 2615–2624.
- [242] M. Zhou, Z. Wang, D. Xia, X. Xie, Y. Chen, Y. Xing, K. Cai, J. Zhang, Hybrid nanoassembly with two-tier host-guest architecture and regioselective enrichment capacity for repetitive SERS detection, *Sens. Actuators-B* 369 (2022), 132359.
- [243] M. Zhou, C. Fan, L. Wang, T. Xu, X. Zhang, Enhanced isothermal amplification for ultrafast sensing of SARS-CoV-2 in microdroplets, *Anal. Chem.* 94 (2022) 4135–4140.
- [244] J. Liu, D. Yan, Y. Zhou, Y. Chen, X. Liu, D. Zhao, J. Liu, J. Sun, Droplet motion on superhydrophobic/superhydrophilic wedge-shaped patterned surfaces with different micro-morphologies, *Colloids Surf. A* 647 (2022), 128999.
- [245] J. Nie, Y. Zhang, H. Wang, S. Wang, G. Shen, Superhydrophobic surface-based magnetic electrochemical immunoassay for detection of *Schistosoma japonicum* antibodies, *Biosens. Bioelectron.* 33 (2012) 23–28.
- [246] Y. Zhang, H. Wang, J. Li, J. Nie, Y. Zhang, G. Shen, R. Yu, Nitrocellulose strip array assembled on superhydrophobic surface: an aqueous solution diffusion-localized platform for multianalyte immunogold staining assays, *Biosens. Bioelectron.* 26 (2011) 3272–3277.
- [247] K. Ellinas, K.V. Pliaka, G. Kanakaris, A. Tserepi, L.G. Alexopoulos, E. Gogolides, Micro-bead immunoassays for the detection of IL6 and PDGF-2 proteins on a microfluidic platform, incorporating superhydrophobic passive valves, *Microelectron. Eng.* 175 (2017) 73–80.
- [248] P. Fang, J.H. Li, F.Y. Jiang, J.F. Mengz, H.C. Pant, Superwetable dendritic gold nanostructured electrode arrays for electrochemical enzyme-linked immunosorbent assay (ELISA), *Int. J. Electrochem. Sci.* 17 (2022), 220828.
- [249] B. Della Ventura, M. Gelzo, E. Battista, A. Alabastrì, A. Schi-rato, G. Castaldo, G. Corso, F. Gentile, R. Velotta, Biosensor for point-of-care analysis of immunoglobulins in urine by metal enhanced fluorescence from gold nanoparticles, *ACS Appl. Mater. Inter.* 11 (2019) 3753–3762.
- [250] T. Wu, K. Yin, H. Zhang, L. Wang, Y. He, J. He, J.A. Duan, C.J. Arnusch, Water-triggered visible and infrared light reversible switch using nanowires-covered micropores superhydrophilic surfaces, *Chem. Eng. J.* 461 (2023), 141894.
- [251] T. Xu, Y. Song, W. Gao, T. Wu, L.-P. Xu, X. Zhan, S. Wang, Superwetable electrochemical biosensor toward detection of cancer biomarkers, *ACS Sens* 3 (2018) 72–78.
- [252] D. Anggraini, N. Ota, Y. Shen, T. Tang, Y. Tanaka, Y. Ho-sokawa, M. Li, Y. Yalikun, Recent advances in microfluidic devices for single-cell cultivation: methods and applications, *Lab Chip* 22 (2022) 1438–1468.
- [253] H. Chen, X. Li, D. Li, Superhydrophilic–superhydrophobic patterned surfaces: from simplified fabrication to emerging applications, *Nanotechnol. Preci. Eng.* 5 (2022), 035002.
- [254] J. Li, C. Zhao, Y. Xu, L. Song, Y. Chen, Y. Xu, Y. Ma, S. Wang, A. Xu, F. He, Remodeling of the osteoimmune microenvironment after biomaterials implantation in murine tibia: single-cell transcriptome analysis, *Bioact. Mater.* 22 (2023) 404–422.
- [255] X.Y. Meng, P. Guo, J. Li, H.K. Huang, Z.Q. Li, H.L. Yan, Z.L. Chu, Y.G. Zhou, A versatile and tunable bio-patterning platform for the construction of various cell array biochips, *Biosens. Bioelectron.* 228 (2023), 115203.
- [256] R. Gayathri, S. Kar, M. Nagai, F.G. Tseng, P.S. Mahapatra, T.S. Santra, Single-cell patterning: a new frontier in bioengineering, *Mater. Today Chem.* 26 (2022), 101021.
- [257] S.T. Gebreyesus, G. Muneer, C.C. Huang, A.A. Siyal, M. Anand, Y.J. Chen, H.L. Tu, Recent advances in microfluidics for single-cell functional proteomics, *Lab Chip* 23 (2023) 1726–1751.
- [258] Z. Jiang, H. Shi, X. Tang, J. Qin, Recent advances in droplet microfluidics for single-cell analysis, *TrAC-Trend, Anal. Chem.* 159 (2023), 116932.
- [259] Y. Sun, W. Song, X. Sun, S. Zhang, Inkjet-printing patterned chip on sticky superhydrophobic surface for high-efficiency single-cell array trapping and real-time observation of cellular apoptosis, *ACS Appl. Mater. Inter.* 10 (2018) 31054–31060.
- [260] H. Li, Q. Yang, G. Li, M. Li, S. Wang, Y. Song, Splitting a droplet for femtoliter liquid patterns and single cell isolation, *ACS Appl. Mater. Inter.* 7 (2015) 9060–9065.
- [261] J. Son, J.Y. Lee, N. Han, J. Cha, J. Choi, J. Kwon, S. Nam, K.H. Yoo, G.H. Lee, J. Hong, Tunable wettability of graphene through nondestructive hydrogenation and wettability-based patterning for bioapplications, *Nano Lett.* 20 (2020) 5625–5631.
- [262] P. Raittinen, P. Elomaa, P. Saavalainen, V. Jokinen, Single cell trapping by superhydrophobic/superhydrophilic microarrays, *Adv. Mater. Inter.* 8 (2021) 2100147.
- [263] Z. Wang, X. Wan, S. Wang, Bioinspired chemical design to control interfacial wet adhesion, *Chem* 9 (2023) 771–783.

- [264] F. Zhang, Y. Jiang, X. Liu, J. Meng, P. Zhang, H. Liu, G. Yang, G. Li, L. Jiang, L. J. Wan, J.S. Hu, S. Wang, Hierarchical nanowire arrays as three-dimensional fractal nanobiointerfaces for high efficient capture of cancer cells, *Nano Lett.* 16 (2016) 766–772.
- [265] B. Wang, S. Zhang, J. Meng, L. Min, J. Luo, Z. Zhu, H. Bao, R. Zang, S. Deng, F. Zhang, L. Ma, S. Wang, Evaporation-induced rGO coatings for highly sensitive and non-invasive diagnosis of prostate cancer in the PSA gray zone, *Adv. Mater.* 33 (2021), e2103999.
- [266] G. Li, H. Wang, Z. Zhu, J.B. Fan, Y. Tian, J. Meng, S. Wang, Photo-irresponsive molecule-amplified cell release on photoresponsive nanostructured surfaces, *ACS Appl. Mater. Inter.* 11 (2019) 29681–29688.
- [267] Y. Liu, N. Zhang, D. Tuo, Y. Zhu, J. Rada, W. Yang, H. Song, A.C. Thompson, R. L. Collins, Q. Gan, Superhydrophobic 3D-assembled metallic nanoparticles for trace chemical enrichment in SERS sensing, *Small* 18 (2022) 2204234.
- [268] P.H. Lu, Y.D. Ma, C.Y. Fu, G.B. Lee, A structure-free digital microfluidic platform for detection of influenza A virus by using magnetic beads and electromagnetic forces, *Lab Chip* 20 (2020) 789–797.
- [269] H. Yousefi, H.M. Su, S.M. Imani, K. Alkhalidi, M.F. CD, T.F. Didar, Intelligent food packaging: a review of smart sensing technologies for monitoring food quality, *ACS Sens* 4 (2019) 808–821.
- [270] R. Iino, K. Hayama, H. Amezawa, S. Sakakihara, S.H. Kim, Y. Matsumono, K. Nishino, A. Yamaguchi, H. Noji, A single-cell drug efflux assay in bacteria by using a directly accessible femtoliter droplet array, *Lab Chip* 12 (2012) 3923–3929.
- [271] W.X. Lei, P. Krolla, T. Schwartz, P.A. Levkin, Controlling geometry and flow through bacterial bridges on patterned lubricant-infused surfaces (pLIS), *Small* 16 (2020) 2004575.
- [272] W. Lei, K. Demir, J. Overhage, M. Grunze, T. Schwartz, P.A. Levkin, Droplet-microarray: miniaturized platform for high-throughput screening of antimicrobial compounds, *Adv. Biosyst.* 4 (2020), e2000073.
- [273] F. Sahin, N. Celik, A. Ceylan, S. Pekdemir, M. Ruzi, M.S. Onses, Antifouling superhydrophobic surfaces with bactericidal and SERS activity, *Chem. Eng. J.* 431 (2022), 133445.
- [274] S. Wang, Z. Liu, L. Wang, J. Xu, R. Mo, Y. Jiang, C. Wen, Z. Zhang, L. Ren, Superhydrophobic mechano-bactericidal surface with photodynamic antibacterial capability, *ACS Appl. Mater. Inter.* 15 (2023) 723–735.
- [275] Y. Wang, X. Du, X. Wang, T. Yan, M. Yuan, Y. Yang, B. Jurado-Sanchez, A. Escarpa, L.P. Xu, Patterned liquid-infused nanocoating integrating a sensitive bacterial sensing ability to an antibacterial surface, *ACS Appl. Mater. Inter.* 14 (2022) 23129–23138.
- [276] H. Sun, Y. Bu, H. Liu, J. Wang, W. Yang, Q. Li, Z. Guo, C. Liu, C. Shen, Superhydrophobic conductive rubber band with synergistic dual conductive layer for wide-range sensitive strain sensor, *Sci. Bull.* 67 (2022) 1669–1678.
- [277] W. Chen, W. Fan, Q. Wang, X. Yu, Y. Luo, W. Wang, R. Lei, Y. Li, A nano-micro structure engendered abrasion resistant, superhydrophobic, wearable triboelectric yarn for self-powered sensing, *Nano Energy* 103 (2022), 107769.
- [278] C. Zhang, Z. Li, H. Li, Q. Yang, H. Wang, C. Shan, J. Zhang, X. Hou, F. Chen, Femtosecond laser-induced supermetalphobicity for design and fabrication of flexible tactile electronic skin sensor, *ACS Appl. Mater. Interfaces* 14 (2022) 38328–38338.
- [279] M. Qu, L. Shen, J. Wang, N. Zhang, Y. Pang, Y. Wu, J. Ge, L. Peng, J. Yang, J. He, Superhydrophobic, humidity-resistant, and flexible triboelectric nanogenerators for biomechanical energy harvesting and wearable self-powered sensing, *ACS Appl. Nano Mater.* 5 (2022) 9840–9851.
- [280] X. He, S. Yang, T. Xu, Y. Song, X. Zhang, Microdroplet-captured tapes for rapid sampling and SERS detection of food contaminants, *Biosens. Bioelectron.* 152 (2020), 112013.
- [281] X. He, T. Xu, W. Gao, L.P. Xu, T. Pan, X. Zhang, Flexible superwetttable tapes for on-site detection of heavy metals, *Anal. Chem.* 90 (2018) 14105–14110.
- [282] X. Jin, G. Li, T. Xu, L. Su, D. Yan, X. Zhang, Fully integrated flexible biosensor for wearable continuous glucose monitoring, *Biosens. Bioelectron.* 196 (2022), 113760.
- [283] L. Wang, T. Xu, C. Fan, X. Zhang, Wearable strain sensor for real-time sweat volume monitoring, *IScience* 24 (2021), 102028.
- [284] J.Y. Xiao, C. Fan, T.L. Xu, L. Su, X.J. Zhang, An electrochemical wearable sensor for levodopa quantification in sweat based on a metal–organic framework/graphene oxide composite with integrated enzymes, *Sens. Actuators-B* 359 (2022), 131586.
- [285] C. Liu, T. Xu, D. Wang, X. Zhang, The role of sampling in wearable sweat sensors, *Talanta* 212 (2020), 120801.
- [286] J. Dong, Y. Peng, L. Pu, K. Chang, L. Li, C. Zhang, P. Ma, Y. Huang, T. Liu, Perspiration-wicking and luminescent on-skin electronics based on ultrastretchable Janus E-textiles, *Nano Lett.* 22 (2022) 7597–7605.
- [287] X. Wang, Y. Liu, H. Cheng, X. Ouyang, Surface wettability for skin-interfaced sensors and devices, *Adv. Funct. Mater.* 32 (2022) 2200260.
- [288] Y. Song, L. wang, T. Xu, G. Zhang, X. Zhang, Emerging open-channel droplets array for biosensing, *Natl. Sci. Rev.* (2023), nwad106.



Zhong Feng Gao is currently an associate professor at the School of Chemistry and Chemical Engineering, University of Jinan. He received his Ph.D. from Southwest University under the supervision of Prof. Nian Bing Li in 2016. He worked as a postdoc at the Huazhong University of Science and Technology (HUST) with Prof. Fan Xia and Prof. Lei Jiang from 2016 to 2018. His current research focuses on the design and engineering of bioinspired materials for bioanalysis, nanosensors, and other biomedical applications.



Hai Zhu received his PHD in China University of Geosciences under the supervision of Prof. Fan Xia in 2021. Now, he is working in the University of Hong Kong as a postdoctor fellow. His research interest mainly focuses on the interfacial materials in environmental and chemical applications.



Qin Wei received her Ph.D. degree from Lanzhou Institute of Chemical Physics, Chinese Academy of Sciences in 2004. She is currently a full professor at University of Jinan and a guest professor at Sungkyunkwan University. Her research interest focuses on the analytical biochemistry, biosensing, and molecular diagnosis.



Fan Xia obtained his Ph.D. degree in 2008 from Institute of Chemistry, Chinese Academy of Sciences under the supervision of Prof. Lei Jiang. Thereafter, he worked as postdoctoral fellow under the supervision of Prof. Alan J. Heeger at University of California, Santa Barbara, USA. Afterwards, he received a professor position in HUST (2012) and China University of Geosciences (2016) successively. Currently, he is the vice president of Fuzhou University. His current research activities include channel materials and their applications in biochemical detection.



Huangxian Ju received his BS, MS and PhD degrees from Nanjing University during 1982–1992. He was a postdoc in Montreal University (Canada) from 1996 to 1997 and a guest professor in three universities of Germany and Ireland in 1999–2000. He is currently a full professor at Nanjing University and a guest professor at University of Jinan. His research interests focus on analytical biochemistry and molecular diagnosis.