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Review article

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FLEXIBLE SENSORS FOR FOOD MONITORING. PART II: APPLICATIONS

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*Flexible sensor,
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ABSTRACT

Monitoring and maintaining food quality, safety, and authenticity are the most important concerns in the food industry. The cutting-edge flexible sensors for food monitoring precisely satisfy the needs of acquiring information on multiple parameters in a small space, they provide for the more reasonable layout, get data on the mechanical deformations, and can be conformably attached to arbitrarily curved surfaces. The flexible sensing materials with a large area of specific surface, that ensure high mobility and density of the media, feature dense active sites, outstanding adjustability and high processing capacities, such as two-dimensional carbon nanomaterials, conductive polymers, and nano-hybrid materials; those materials have further improved the sensitivity, stability and selectivity of the flexible sensors' perception. This article attempts to critically review the present state-of-arts developments in relation to the materials, manufacturing techniques and sensing mechanisms of the devices, as well as the applications of the electrically-transduced flexible sensors. Moreover, this article elaborates on the transduction mechanisms of the several typical transducers, with a focus on the physics behind, including the modulation of the doping level, Schottky barrier, and interfacial layer that typically cause changes in conductivity, functionality and permittivity. We also highlight the benefits and the technical challenges along with the appropriate solutions provided by the presented flexible sensors, and we also consider the potential strategies that allow overcoming limitations in power consumption, quantitatively assess the trade-offs in maintaining the quality and marketability, to optimize wireless communication and explore new sensing patterns.

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Обзорная статья

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ГИБКИЕ СЕНСОРЫ ДЛЯ МОНИТОРИНГА ПИЩЕВЫХ ПРОДУКТОВ: ЧАСТЬ II – ПРИМЕНЕНИЕ

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КЛЮЧЕВЫЕ СЛОВА: АННОТАЦИЯ

*гибкий сенсор,
мониторинг
пищевых продуктов,
эластичные по своей
природе, механическое
соответствие,
проводящий электрод,
электрическое
свойства, сенсорный
механизм, механизм
преобразования*

Мониторинг и поддержание качества пищевых продуктов, их безопасность и аутентичность являются наиболее важными проблемами в производстве продуктов питания. Новейшие гибкие сенсоры для мониторинга качества пищевых продуктов идеально удовлетворяют потребность получения информации по многочисленным параметрам, при этом занимая очень малое пространство и обеспечивая возможность наиболее разумного размещения в производственном процессе. Эти сенсоры предоставляют данные о механических деформациях, и могут удобно размещаться на произвольно вогнутых поверхностях. Гибкие чувствительные материалы с большой площадью удельной поверхности, обеспечивающие высокую подвижность и портативность чувствительных элементов, характеризуются высокой плотностью действующих ячеек, превосходной настраиваемостью и отличными технологическими возможностями. В частности, двумерные углеродные наноматериалы, проводящие полимеры, и наномерные гибридные материалы еще больше повысили чувствительность гибких сенсоров, обеспечили стабильность считывания и селективность срабатывания. В данной статье авторы попытались дать критический обзор новейших разработок в сфере применяемых материалов, производственных технологий, разобрать принципы действия сенсорных механизмов этих устройств, а также дать обзор способам применения гибких сенсоров с электрическими преобразователями. Кроме того, в этой статье подробно рассматриваются механизмы преобразования, обуславливающие действие некоторых типовых преобразователей, с упором на физику, лежащую в основе таковых явлений, включая модуляцию уровня легирования барьера Шоттки и межфазного слоя, которые обычно вызывают изменения проводимости, диэлектрической проницаемости и функциональности. Мы также подчеркиваем преимущества и технические проблемы, перечисляем соответствующие решения, обеспечиваемые гибкими сенсорами, а также рассматриваем потенциальные стратегии, позволяющие преодолеть ограничения в энергопотреблении, количественно оценить компромиссы в поддержании качества и конкурентоспособности продукта, оптимизировать беспроводную связь и исследовать новые модели считывания данных путем их сенсорного обнаружения.

1. Flexible sensors for food monitoring

The wide range of flexible sensing techniques, shapes and forms that have been applied to food monitoring will be considered here. In this section, we will systematically discuss three types of electrically-transduced analytical flexible sensors. These sensors include physical sensors, which are used for monitoring food texture, fruit ripeness, and temperature/

humidity variations in the warehouse environment during their storage; chemical sensors, which are used for monitoring pH, heavy metals content, marker gases of food spoilage (e. g., H₂S, NH₃, etc.), content of pesticide and residues of antibacterial substances; and biosensors which are used for monitoring bacteria, organophosphates, biogenic amines and pesticide residues. Flexible sensors based on simple colorimetric, fluores-

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cent, and paper chromatography surface enhancement of Raman scattering (SERS) have also been widely used in food quality, safety, and authenticity monitoring due to their simplicity and cost-effectiveness. However, they are not strictly considered as electrically-transduced sensors as defined previously. Therefore, we will not consider these sensors here.

1.1. Flexible physical sensors

1.1.1. Temperature

Temperature is one of the important factors that affect the quality and safety of food. Temperature fluctuations at any point within the chain of the cold storage and distribution, like handling, loading, unloading, temperature cycles in the walk-in coolers, at the storage displays, and during its transportation home provide a strong impact on the shelf life of refrigerated food products [1,2,3,4,5]. Temperature plays a significant role in determining the rate of microbial activity development, chemical changes, and loss of nutritional value. Specifically, during refrigeration there is a potential risk for the staple food products, as the temperature between 4 and 12 °C is optimal for the growth of psychotropic bacteria and thermo-tolerant fungi. To ensure food safety and quality it is crucial to monitor and regulate temperature at every step of food production, storage, transport and distribution [6,7].

Escobedo et al. [8] designed a flexible printed temperature sensing cell. This sensor uses a commercially available PVC board as its flexible substrate; its conductive electrodes are made of silver conductive paste applied by stencil printing method. The temperature sensing material is a conductive polymer PEDOT: PSS. The sensing mechanism can be described as follows: change in environmental temperature affects as a physical perturbation on the sensing material, the sensing material acts as a resistor, and the mobility of the internal charge carriers in PEDOT: PSS polymer increases along with the increase of temperature, thus leading to an increase in its conductivity. The conductivity increase is transduced by the transducer into a resistance decrease, which in its turn raises circuit current and makes the LED indicator flash brighter. Being combined with another strain sensor integrated on the substrate card, the originally lit LED will turn off when the food packaging bag shows signs of Blown Pack Spoilage (BPS) due to food spoilage, which process proves that the food is no longer suitable for consumption. The temperature sensor based on PEDOT: PSS showed 70% change of resistance for a temperature variation of approximately 60 °C. Although the resistance-based sensing mechanism lacks a quantitative correlation between the sensor response and the precise level of spoilage, its threshold detection is quite sufficient for the intended application in the field of BPS detection.

1.1.2. Humidity

The level of humidity plays a crucial role in various sectors of the food industry, ranging from dairy and meat to dehydrated products [9]. Humidity is a key indicator to monitor when testing food quality, as increased moisture provides a favorable environment for the growth of bacteria and fungi, thus becoming a safety issue for product consumption [10]. In addition to microbial growth, humidity can also cause spoilage of dehydrated products, result in product softening and moisture absorption, thus shortening the shelf life [11]. Therefore, monitoring and maintaining the due humidity levels in food packaging and storage rooms are crucial for preserving the quality and texture of food [12,13].

Molina et al. [14] created interdigitated electrodes (IDE)-based capacitive humidity sensor using flexible PET foils as substrate. Firstly, silver nanoparticles ink was inkjet-printed onto a PET substrate to patternize the planar IDE. Then, electrodeposition was performed on PET foil in a nickel sulfamate bath at 54 °C and current intensity of 20 mA/cm², resulting in electrodeposition of layers ranging in thickness from several hundred nanometers to 15 µm. Cellulose acetate butyrate (CAB) was used as the sensing material and was inkjet-printed onto the IDE capacitor. Thus, the sensor creation was completed. It should be noted that pretreating PET substrates with oxygen plasma does not improve the adhesion of metal to the substrate; it does demonstrate any beneficial results in enhancing the smoothness of line edges. Additionally, when the printed electrodes come into direct contact with the environment, silver is highly likely to interact with moisture, oxidizing and/or releasing some organic residues that may have been extracted from the ink. Passivation of the inkjet-printed silver electrodes with nickel can improve the stability of electroplated sensors. The sensing mechanism of this capacitive humidity sensor can be summarized as follows: the water vapor, which is the target gas, is absorbed by the polymeric sensing layer. Consequently, the sensing layer swells and increases its thickness, which alters its dielectric constant ϵ_s , and modifies the value of C. When the thickness of the sensing layer is increased (up to the saturation value of $\epsilon_s, 1 \sim 0.5$), it can absorb greater number of molecules in the fixed concentration of ambient gas. This causes larger shifts

of C and leads to higher sensitivity but can also lead to unfavorable factors such as long-term response and hysteresis of the sensor. The sensitivity of this device is 2.36 ± 0.08 fF per 1% R. H. It is worth noting that the technique used to fabricate the sensing layer, electrodeposition on inkjet-printed patterns, is a smart technical solution for the simultaneous production of temperature and gas sensing platforms.

1.1.3. Pressure

The growing competition in both domestic and international fruit markets has created a demand for better techniques to evaluate ripeness of fruit. This is necessary to minimize potential losses for the growers and the packers, as well as to prevent spoilage of fruit supplied to the consumers. While determining the optimal harvest time and precise stage of ripeness are crucial in evaluating the quality of many fruit varieties, there is still a necessity to find the appropriate techniques to monitor the ripeness degree of numerous varieties [15,16]. Flexible tactile sensing technology can serve as useful approach in agriculture to evaluate the inherent quality of fruit and supplement the information obtained from visual inspection [1,17]. However, tactile sensors that rely on a single mechanism typically cannot comprehensively assess fruit quality in both qualitative and quantitative terms, which may not provide sufficient guidance for terms of logistics and shelf life longevity [18]. Thus, highly sensitive flexible pressure sensors which use multiple mechanisms are greatly needed [19].

Xia et al. [18] designed a dual-mechanism pressure sensor (DMPS) that is made up of Ag nanowire/PDMS electrodes and ZnO nanoparticle-based membranes that are able to sense pressure. The sensor features sandwich-like structure and is very flexible because of chemical bonding. It can detect both high-frequency dynamic pressure and static load at the same time by using both piezoelectric and piezocapacitive effects. It provides certain benefits in comparison with the traditional piezoelectric sensors, which cannot continuously detect static pressure. Firstly, the Ag-NW ethanol dispersion was drop-applied onto PDMS flexible substrate to create a flexible AgNW/PDMS electrode through solvent evaporation and annealing. Second, PDMS mixture solution containing Zinc oxide powder was spin-applied onto a glass substrate and allowed to cure before its peeling off from the glass substrate. Third, both sides of the PDMS-ZnO film were plasma-treated with O₂ and amine-modified to prepare for the next step of assembly. Finally, two flexible electrodes and piezoelectric film were assembled into a sandwich structure at 60 °C. Due to the strong covalent bond between the -OH modified PDMS and APTES modified PZO, the multilayered structure layers adhere tightly to each other. The sensing mechanisms of this dual mechanism pressure sensor can be described as follows: (1) In piezoelectric mode, the piezoelectric potential that appears between the electrodes is created by deformation of the crystalline material and internal dipole polarization initiated by mechanical stimulation. (2) In piezocapacitive mode the addition of nanofillers into the polymer creates bulges on the membrane surface with empty spaces between each one. When the membrane is compressed, these spaces are filled by neighboring composites, thus causing the increase of the comprehensive dielectric constant of the dielectric layer. It is worth to underline that: (a) In piezoelectric mode, after optimizing the content of ZnO nanoparticles, this pressure sensor shows the improved electrical output properties, resulting in an open-circuit voltage of approximately 2 volts when subjected to an external force of 20 newtons. And it is basically able to ensure stable functioning under 5,000 load cycles in this mode. (b) In piezocapacitive mode, the sensor's pressure sensitivity varies within 0–16 kPa and 16–100 kPa, which is equal to are 17.82 and 5.75 MPa-1 respectively. This is approximately double the sensitivity levels observed in a sensor produced with flat copper foil electrodes under similar conditions. Moreover, the output of the sensor's capacity remains stable even after being subjected to 5,000 load cycles.

1.2. Flexible chemical sensors

1.2.1. pH

During the food spoilage processes the original chemical compounds like glucose, lactic acid and certain amino acids are catabolized by microbes or microflora [20]. The pH of the chemical environment of food can be altered by acidic or basic metabolites produced during microbial spoilage [21]. These markers, such as liquids (e. g., fatty acids, lactic acid, etc.) and gases (e. g., amines, carbon dioxide, etc.), can form acidic or basic substances when dissolved [22]. Therefore, monitoring pH profiles in food provides a tool for food quality measurement [23].

Xiao and his team [24] developed a flexible battery-free wireless electronic system (FBES) equipped with electrochemical pH sensor. Double-electrode pattern is implemented on a thin commercial PET/ITO film (~125µm) using a carbon dioxide laser for its scribing. Then, Ag/Cl is

applied to the reference electrode through a drop-coating process to finalize the production of a flexible electrode. However, the first issue was the inflexibility of ITO film, which makes it unsuitable for its application in flexible devices because of the oxide material brittleness [25,26,27]. However, growing ITO into nano-branched structures has solved this issue [28]. At second, the sensing mechanism can be analyzed as follows: this two-electrode pH sensor can utilize the activity of target ions (H^+ , OH^-) as an input signal, and the potential as an output signal, for effective conversion of ions to electrons [29]. According to Nernst equation, when the activity of measured ions increases by 10 times, the potential/electromotive force (EMF) increases by $59.2 \text{ mV}/z_i$ (z_i is the ionic charge of the ion " i ") [30]. The activity of ions has a close correlation with the concentration of hydrogen ions or hydroxide ions. Ag/Cl electrode has made major contribution to the stable potential difference between the working electrode and the reference electrode: ion-to-electron transduction proceeds through the reversible redox reaction between $AgCl(s) + e^- \rightleftharpoons Ag(s) + Cl^-(aq)$, which provides stable interfacial potential between the AgCl/Ag electrode and the inner filling solution [30]. It is worth emphasizing that the analysis is run under thermodynamic equilibrium conditions, which means that the system can be observed without applying the potential that can induce chemical reactions, and thus can detect chemical substances without involving any redox processes [31].

1.2.2. Gases

Oxygen (O_2): Oxygen is essential for life but also can spoil food through its oxidation (lipid and ascorbic acid), enzymatic reactions (accelerated ripening, darkening or browning), and microbial growth [32,33]. Thus, it is crucial to keep track of the levels and variations of oxygen within the food packaging to ensure food quality and safety.

Seoyeon et al. [34] designed a cut-the-edge self-powered flexible oxygen gas sensor for the smart food packaging. This self-powered sensor is based on metal-air batteries designed to generate power by oxidizing metals with the air oxygen. This flexible oxygen sensor was fabricated through a layer-by-layer process. Firstly, an adhesive gel electrolyte (thickness 600 μm) was attached to a zinc sheet (anodic adhesion, thickness 100 μm), followed by coating with Ag-OPP (oriented polypropylene) film on the electrolyte. Silver, serving as catalyst (thickness 0.1 μm), was deposited onto the OPP film (25 μm). Second, the three layers were laminated with an automatic adhesion roller. Compared to gel electrolyte, both the Ag-OPP film and zinc sheet extended 1 cm as conductive electrodes in the opposite direction. The sensing mechanism of this self-powered flexible potentiometric/voltammetric/amperometric sensor is based on a metal-air battery capable to generate energy sufficient for monitoring analytes through electrochemical reactions. They are composed of a metal anode (Zn), an air cathode (O_2), and an electrolyte. The pure metal zinc is oxidized into Zn^{2+} at the anode by providing electrons, which are then captured by oxygen to diffuse through a gas diffusion layer from the air at the cathode. The slow redox reaction at the cathode is catalyzed with the help of Ag as an electrocatalyst. The electrochemical reactions of zinc-air battery in acidic or neutral electrolytes are described by the following equations: Zinc anode: $Zn \rightarrow Zn^{2+} + 2e^-$ ($E^\circ = -0.762 \text{ V}$), air cathode: $1/2O_2 + 2H^+ + 2e^- \rightarrow H_2O$ ($E^\circ = +1.229 \text{ V}$). Under potentiometric mode, according to Nernst equation, the open circuit voltage (OCV) between the two electrodes is proportional to the logarithm of the analyte concentration. Under voltammetric/amperometric mode, when the two electrodes are connected to the load resistor to form a closed circuit, the flow of electrons from the anode to the cathode gets proportional to the analyte concentration. Moreover, according to Ohm's law, the voltage of the closed circuit is proportional to the current. In addition, it should also be emphasized that (1) a sensitivity of 18 mV/% O_2 and good linearity ($R^2 = 0.999$) were achieved within the range of 0–21% O_2 gas, (2) the silver catalyst was deposited on a thin and transparent OPP film in the gas diffusion layer because it is oxygen-permeable. Silver is not only non-toxic but also has antibacterial activity. (3) The gel electrolyte is slightly acidic, so it is more environmentally friendly and less sensitive to CO_2 in comparison with the strongly alkaline electrolyte widely used in metal-air batteries (4). In the bending test the initial OCV of the sensor has hardly decreased after 250 bending cycles ($R_c = 3.3 \text{ mm}$).

Carbon dioxide (CO_2): carbon dioxide can inhibit the growth of bacteria and fungi, while also lowering the pH level in the food environment [11]. Carbon dioxide inhibits the growth of gram-negative aerobic bacteria, such as *Pseudomonas* spp, which can produce bad odors and harmful chemicals such as sulfides and acids during their growth and metabolism in food, thus leading to food spoilage [35]. The antimicrobial effect of CO_2 is attributed to its ability to create an anaerobic environment that prevents enzymatic decarboxylation [11]. Accumulation of CO_2 may also disrupt the permeability of certain microbial membranes [36]. Any change

in gas concentration/composition can indicate the deterioration of food quality and signal the presence of microbial activity undesirable for the consumers [37]. Monitoring and controlling the amount of CO_2 is, therefore, crucial for extending the shelf life of food [38].

Shahrbabaki et al. [39] developed a flexible CO_2 sensor through depositing poly(N-[3-(dimethylamino)propyl]-methacrylamide-co-2-N-morpholinoethyl methacrylate) (p(D-co-M)) on planar double-parallel-lines CB electrode which was stenciled on a PET substrate. The less basic monomer 2-N-morpholinoethyl methacrylate (MEMA, pKaH of homopolymer = 4.9) [40,41] provides more free amine sites for p(D-co-M), especially at lower pH. This solves the issue of non-linear and irreversible response by allowing for the adjustment of the basic parameters of pDMPAM [42]. The sensing mechanism of this chemiresistor can be described as follows: (1) In transduction, the CO_2 -responsive amine side groups of p(D-co-M) can alternate between the neutral and charged states depending on presence or absence of CO_2 [43]. This switchable behavior in the charge state of amine side groups is translated to the corresponding changes in the electrical resistance of the polymer. (2) For reverse procedure the polymer chains of p(D-co-M) contain primary, secondary and tertiary amines, which are the organic bases that can react with carbonic acid generated by CO_2 in the presence of water or wet organic solvents. In the presence of CO_2 , the functional groups in the polymer chains change from neutral to charged state (or vice versa) as this equation: $B + CO_2 + H_2O \rightleftharpoons [BH^+][HCO_3^-]$. For reversible CO_2 sensors, switchable CO_2 -responsiveness is required which involves hydrogen protonation and deprotonation. The deprotonation process is often referred to as good reversibility. Higher pKaH values indicate stronger bases and produce higher degrees of protonation (DOP). However, the deprotonation process gets more complicated. Higher pKaH values make charged bases more difficult to deprotonate [44]. The pKaH of tertiary amines is approximately 6.0–7.0 [45,46], which means that they are mild bases and are expected to have good conversion properties, i. e., they are easy to protonate and have good reversibility. Primary and secondary amines possess N-H bonds that react with CO_2 in water, thus forming carbamate salt [47]. Carbamate formation is not desired for sensing applications because it is not reversible unless exposed to high temperatures, which requires additional heating, expenses and time [43]. Therefore, the tertiary amine in the polymer chains plays a significant role in improving the reversibility and reducing operating temperature of the sensor.

Ammonia (NH_3): the spoilage of high-protein food, like fish and shrimp, releases nitrogen-containing compounds, which are the source of the unpleasant odor produced during meat decomposition [48,49,50]. Volatile amines such as ammonia, dimethylamine, and trimethylamine are produced by amino acids decomposition and activity of *Pseudomonas* bacteria, usually referred to as total volatile basic nitrogen (TVBN) [51]. Ammonia, as the main component of TVBN, may serve as potential indicator of protein-rich food spoilage [52]. Real-time measurement of ammonia emitted from food has great significance for evaluating the food quality, reducing food waste, controlling the occurrence of foodborne diseases, and studying the relationship between ammonia generation and food quality [48].

Tang et al. [53] designed a NH_3 sensor based on flexible PEDOT: PSS nanowire. The NH_3 sensing system that was integrated showed improved performance, with the ability to detect as low as 100 parts per billion (ppb) of NH_3 , along with high selectivity and reproducibility. Additionally, the flexible nanowire sensor consumed very little power, as low as 3 μW . Firstly, a soft nanomold containing well-defined parallel nanogrooves was prepared by thermal nanoimprint lithography (NIL) on a flat PDMS substrate. Second, after soft bonding to a plasma-treated PET substrate, the nanogrooves turned into parallel nanochannels. Third, a few drops of aqueous solution containing functional materials PEDOT: PSS were applied at the edges of the nanochannels to shape the sensitive PEDOT: PSS nanowires. Finally, the Au interdigital electrodes (IDE, $N = 9$, length = 8 mm, width = 300 μm) were deposited by vacuum evaporation through a designed shadow mask. The sensing mechanism of this flexible NH_3 chemiresistor can be explained as follows: during the interaction between the analyte and the sensing material, the distance between the target molecule and the sensing material becomes close enough to allow electron transfer between them [54]. The analyte NH_3 acts as an electron donor, increasing or compensating for the doped charge density. This process is commonly referred to as secondary doping of the sensing material [31]. The modification of the doping density thus changes the state density in the barrier area and consequently alters the conductivity of the sensing material PEDOT: PSS. By modulating the doping level, the change in conductivity of the sensor is closely related to the rate of the analyte molecule occupancy on the sensing material surface. Specifically, when being in contact with NH_3 , the holes in the valence band of the con-

ducting polymer PEDOT: PSS are depleted by the electron-donating gas NH_3 , leading to a significant decrease in conductivity and a macroscopic increase in its resistance. This mechanism is described by the binding site hypothesis, where the atoms on the surface of sensing material can serve as binding sites for molecule adsorption [53,55]. It is worth to mark that even after 1,200 bending cycles this flexible device still demonstrates excellent mechanical flexibility and durability with no significant decrease in performance. And in a dynamic response test to VOCs containing NH_3 , ethanol, IPA, p-xylene, acetone, n-heptane, and n-hexane, the flexible chemiresistive sensor showed a preferential response to NH_3 (with sensitivity up to 0.25), while the response to other VOCs can be neglected (with a maximum sensitivity of 0.065) [56].

Ethylene(C_2H_4): ethylene is the colorless and odorless plant hormone that plays a crucial part in fruit ripening, especially in climacteric fruits [57]. The sharp increase in climacteric ethylene production at the ripening onset is considered as controlling the beginning of changes in color, aromas, texture, flavor, and other biochemical and physiological parameter [58]. Generally, climacteric fruits are harvested before the climacteric stage, and the ripening process is managed during transportation and storage by controlling the level of ethylene to maintain optimal freshness when displayed for sale [59]. Meanwhile, controlled atmosphere was developed to allow longer storage periods and, subsequently, to provide a wide variety of fruits to the consumers over the entire year. Therefore, it is extremely important to monitor and control the ethylene emission in growth chambers, greenhouses and storage facilities to optimize fruit freshness [60].

Yan et al. [57] developed a wearable paper-based chemoresistive ethylene gas sensor which consists of a paper-based substrate, gold interdigitated electrodes (IDE), sensing layer, and encapsulation layer. Firstly, laser direct writing was carried out to pattern the interdigitated electrodes (IDE) on gold foil which was glued to high-temperature resistant paper using polyvinyl alcohol glue based on local heating with steam iron. Second, a liquid polyimide solution (50 wt%) was coated on both sides of the interdigitated electrodes as an encapsulation layer, which only left the sensing area exposed to the target ethylene gas. Finally, modify the sensing area of IDE through pipetting 10 μL of the SWCNT-PdNP-polystyrene microsphere (SPPM) dispersion and drying at room temperature. Sensing material SPPM contains the following characteristics: (1) The unique properties of CNTs (i. e., high carrier mobility, excellent physical properties, ease of modification, sizeable surface-area-to-volume ratio, etc.) enable the sensor to have a fast response and substantially higher sensitivity at room temperature. More detailed discussion on the electrical, physical, mechanical, and chemical properties of SWCNTs can be found in section 2.3.1 'Single-Walled Carbon Nanotubes' part of the Carbon Nanomaterials chapter. (2) As a potential C_2H_4 acceptor and catalyst, PdNPs are expected to achieve a high and rapid response and a low detection limit [61], wherein PdNPs can both provide a larger surface area and catalyze the cleavage of carbon-carbon double bonds [62]. Hydrocarbons can have a good affinity for transition metals, which are located in the middle of the periodic table, have partially filled d-orbitals that can interact with hydrocarbons in various ways. (3) Polystyrene microspheres further improved the sensitivity by increasing the surface area of the SWCNT network, upgrading the charge distribution and transfer in the SPPM for ethylene gas sensing, and possibly enhancing the local concentration of ethylene in the device as it partitions into the polystyrene beads [63,64]. The sensing mechanism can be analyzed as follows: (1) The work function of Pd (5.12 eV) is higher than that of SWCNT (4.7-4.9 eV) [65], forming a Schottky junction between them, which causes electrons to flow from SWCNT to PdNP. Oxygen molecules quickly adsorb onto the surface of the SPPM and, with the help of the Schottky junction, the adsorbed oxygen molecules combine with the electrons provided by SWCNT to form adsorbed oxygen O_2^- as shown in the equation: $\text{O}_2 + e^- \rightarrow \text{O}_2^-$ (a). This helps to create an electron depletion layer around the SWCNT [66], leading to an increase in resistance [67,68,69]. (2) The adsorbed oxygen O_2^- reacts with the target ethylene molecules as shown in the equation: $2\text{C}_2\text{H}_4 + \text{O}_2^- \rightarrow 2\text{C}_2\text{H}_4\text{O} + e^-$ (b), which releases electrons. This weakens the electron depletion layer, leading to a decrease in resistance. (3) The intermediate product $\text{C}_2\text{H}_4\text{O}$ continues to react with the adsorbed oxygen O_2^- as shown in the equation: $2\text{C}_2\text{H}_4 + 5\text{O}_2^- \rightarrow 4\text{CO}_2 + 4\text{H}_2\text{O} + 5e^-$ (c). Reaction (c) releases more electrons, further reducing the resistance of the SPPM. Finally, it should be emphasized that the concentration of C_2H_4 can be detected down to 100 ppb using the SPPMs/FWPCS at 25 °C.

1.2.3. Pesticides

Pesticides are extensively used in modern agriculture in order to meet the global demand for food [70,71]. As the pesticides (organophosphorus, organochlorine, carbamate, dithiocarbamate, etc.) are carcinogenic,

they are harmful for humans when being included into the food chain [72] which resulted to the requirement to monitor the availability of pesticide remnants in soil, their effluents from wastewater, surface water, primary sources of drinking water, and food items [73,74]. Analytical techniques such as gas chromatography (GC) [75] and high performance liquid chromatography (HPLC) [72] demand expensive equipment and time-consuming processing by the skilled personnel. On the one hand, enzyme-based sensors are limited in terms of non-specific interactions with interferents, storage and long-term stability [71,76]. On the other hand, immunosensors face issues concerning storage, stability, and high cost when it comes to the use of antibodies [77].

Zhu Xiaoyu and colleagues [78] created a sensor for detecting very small amounts of the plant growth regulator α -naphthalene acetic acid (NAA). This sensor was made of combination of two-dimensional phosphorene (BP) and graphene-like titanium carbide MXene (Ti_3C_2 -MXene) materials (MXene, 2D material, it can be synthesized by etching "A" from MAX phase ("M" represents transition metals, "A" represents group IIIA/IVA elements and "X" represents C and/or N elements)), which were applied onto a flexible substrate made of laser-induced porous graphene (LIPG). Using ultrasonic assistance in an organic solvent, a stable nanohybrid of BP- Ti_3C_2 -MXene was created by liquid-phase exfoliation of black phosphorus with cuprous chloride and Ti_3C_2 -MXene, which had been obtained by etching the aluminum layers from Ti_3AlC_2 . MXene features distinct thin-layered nanostructure that offers abundant room for accommodating the other functionalized nanomaterials. Nonetheless, the efficiency of MXene can be deteriorated when its nano-layers stack too much, leading to damaging of the active surface area [79]. In order to resolve this issue, a noncovalent nanohybrid of BP and MXene has been proposed as a solution to prevent the aggregation of either material while also leveraging the benefits of both substances [80]. This electrochemical sensor operates on the following sensing mechanism: The working electrode, which has been modified with Ti_3C_2 -MXene/BP biomimetic enzymes, demonstrates oxidase-like properties (acting as a nanozyme) in amperometric mode when the zymolyte NAA is electrocatalytically oxidized. As a result of electron transfer reactions, detectable alterations in current take place. As the magnitude of the current generated is directly proportional to the quantity of NAA molecules present in the solution, the concentration of the molecules can be tracked in a time-dependent manner [81]. This sensor exhibits an extremely low detection limit (LOD) of 1.6 nM and an expansive linear range within 0.02–40 μM .

1.2.4. Antibacterial substances

Sulfonamides (SAs) are the derivatives of sulfanilic acid (p-aminobenzenesulfonic acid) and are included into the list of the most widely used antibiotics. Residues of these antibiotics in food pose a serious health hazard [82]. The antibacterial mechanisms of SAs can be described as: The sulfanilamide part is structurally similar to p-aminobenzoic acid (PABA), which is involved in the biosynthesis of dihydrofolic and folic acids, as well as other substances utilized by microorganisms. SAs act as competitive inhibitors of the enzyme dihydropteroate synthase, inhibiting the catalytic conversion of p-aminobenzoic acid (PABA) to dihydropteroic acid, which is a precursor compound of dihydrofolic acid. Dihydrofolic acid is further reduced down to tetrahydrofolic acid by dihydrofolate reductase. Tetrahydrofolic acid participates in the synthesis of nucleic acids. The lack of nucleic acid synthesis would result in an inability to produce the associated proteins. Therefore, SAs competitively bind with the enzyme dihydropteroate synthase, inhibiting the synthesis of dihydropteroic acid, which in turn inhibits the synthesis of nucleic acids and ultimately protein synthesis, leading to the antibacterial effect [83,84,85]. Sulfamethoxazole (SMZ) is an important antibiotic obtained from the sulfonamide class that is used to prevent and treat infections. However, it is considered a persistent pollutant that can have significant toxicological effects on both the food of animal-origin and aquatic environments [86,87]. Exposure to low levels of antibiotics over a long period of time can increase the risk of development of antibiotic resistance and even thyroid cancer in humans [88,89]. Therefore, it is crucial to develop efficient and convenient methods to detect residual antibiotics in the food and water [90].

Zeng et al. [91] developed the portable electrochemical sensor for fast detection of sulfonamides (SAs). At first, the original laser-induced porous graphene (LIPG) with an ordered-porous flake foam-like structure was prepared and applied on a polyimide (PI) film according to the design drawing. As the laser stencil scribing was carried out in air, the local availability of oxygen and moisture could cause the burn-off of some carbon during graphitization, thus releasing CO and CO_2 gases and leading to porosity [92]. At second, the working electrode was electrochemically modified with two-dimensional hexagonal boron nitride (2D h-BN), and

then Ag/Cl ink was drop-coated on the reference electrode. The sensing mechanism can be described as follows: this voltammetric sensor produces a measurable response by the variation of the current at different potentials [93]. The surface of the working electrode was functionalized with an electroactive layer of hexagonal boron nitride (h-BN) to induce selectivity towards the targeted analytes. The applied potential is the driving force for electron transfer reactions, which generate measurable current changes. As the magnitude of the measured current is directly proportional to the number of oxidizing/reducing molecules in the solution, the relevant concentrations of the molecules can be monitored on physiological timelines [81]. It is worth emphasizing that this voltammetry-based sensor displayed a good linear range from 0.5 to 362.5 μM , a low limit of detection was 0.011 μM , and satisfactory recoveries of SMZ in the range of 97.5%–101.3% in milk and lake water. It also exhibits excellent repeatability with a 4.01% RSD value for the repeatable peak currents under the 20th successive measurement, and features good reproducibility with a 4.65% RSD value. Moreover, after 50 bending cycles, the sensor still demonstrated good mechanical stability. This sensor was also applied for the detection of four other SAs, including sulfanilamide, sulfapyridine, sulfadimidine, sulfisoxazole.

1.3. Flexible biosensors

1.3.1. Foodborne pathogens

Food has been considered as a potential carrier for intentional contamination that could result in human or animal illnesses and deaths, as well as economic losses [94]. *Salmonella enterica*, which is found in raw/undercooked eggs, poultry meat, etc., is one of the most prominent foodborne pathogens [95,96,97,98,99]. Therefore, monitoring of *Salmonella enterica* in food is the solution for prevention and recognition of the issues related to health and food safety.

Soares et al. [100] designed a LIG-based impedimetric immunosensor which functions with polyclonal anti-salmonella antibody to detect *Salmonella enterica*. The LIG electrodes were produced by laser induction on the polyimide (PI) film in ambient conditions. And then the working area was separated from the connector ends of the working electrode with a passivation layer (fast-drying varnish) to cover the non-active areas of the electrodes. During the process of electrochemical sensing, passivation was run to ensure that the working electrode retained a consistent surface area exposed to the redox solution [101]. The antibody functionalization of LIG electrode can be described as follows: firstly, 1-ethyl-3-(3-dimethylaminopropyl) carbodiimide (EDC) and N-hydroxysuccinimide (NHS) are fixed on the surface of the LIG working electrode in a ratio 3:1. Then, the polyclonal anti-salmonella antibody is incubated at 4 °C for a certain period of time, and the biofunctionalization of the sensing material is completed through carbodiimide cross-linking chemical reaction with EDC/NHS. The sensing mechanism of this impedimetric immunosensor can be described as follows: the change in charge transfer resistance (R_{ct}) on the electrode surface is directly proportional to the adhesion of bacterial cells to the biofunctionalized area of the electrode [102]. This “bio-barrier” hinders the entry of electrolytes, acting as an electron-blocking agent, thus increasing the R_{ct} [103,104,105]. Furthermore, the presence of adhered bacterial cells acts as an electron kinetic barrier and steric hindrance [103], thus reducing the electronic transmission path between the electrolyte solution and the electrode, leading to increase of R_{ct}. According to this technique, a larger diameter corresponds to a larger R_{ct}, thus indicating a higher number of bacteria bound to the antibodies on the electrode surface [102]. It is worth emphasizing that this LIG biosensors showed the capability to detect live *Salmonella* in chicken broth within a broad linear range (25 to 105 CFU mL⁻¹) and at low detection limit (13 ± 7 CFU mL⁻¹; n = 3, mean ± standard deviation). These results were obtained within an average response time of 22 minutes, without the requirement for preconcentration of the sample or methods of redox labeling. Furthermore, the LIG immunosensors demonstrated high selectivity, as it was proven by the absence of significant responses to other strains of bacteria.

1.3.2. Pesticides

Organophosphate (OP) nerve-agent are routinely used as pesticides in agriculture and household purposes [106,107]. It represents a serious concern as they can be weaponized as chemical warfare agents (CWA) [108,109], which represents a major security challenge [110]. Therefore, there is an urgent need for rapid and selective pesticides monitoring due to the high toxicity of OP nerve-agents and pesticides [108,111,112].

Mishra et al. [113] developed a stretchable glove biosensor for monitoring of the organophosphorus chemical threats. Firstly, the reference electrodes and long serpentine structures of three-electrode system were fabricated on the index fingers of nitrile gloves using intrinsically

stretchable Ag/AgCl ink with platinum-catalyzed silicone elastomer Ecoflex through screen printing. Secondly, the working electrode and counter electrodes were printed by carbon inks mixed with polystyrene-block-polyisoprene-block-polystyrene (PS-PI-PS). Third, the serpentine interconnects were coated with a precise layer of insulation to create a dielectric barrier between the three electrodes and to prevent any potential short-circuits in the device while exposing the sensing area to square contact pads. Finally, the working electrode was modified by drop-coating with the mixture of solutions of organophosphorus hydrolase (OPH) and Nafion onto it (on the index finger) after cleaning it by cyclic voltammetry (CV) in the range of 0 to +1.0 V using 0.01 M sodium acetate buffer (pH 4.6) with a scan rate of 0.1 V/s for 20 cycles. Square-wave voltammetry (SWV) can be used for the analysis of catalytic reactions. The increased ratio of faradaic current to non-faradaic current allows for a lower limit of detection and for a higher sensitivity [114]. It features the following advantages: at first — it can be operated at high frequency [115] allowing for quick experiments that conserve electroactive species [116] and tend to result in less hindrance induced by non-electroactive products on the electrode surface [114]; at second — there is no need to exclude oxygen from the solution, because the reduction of oxygen is included in the background current [117]. The sensing mechanism of this biosensor can be described as follows: organophosphate (OP) nerve agents diffuse through the conductive semisolid gel matrix on the index finger sensing area and reach the OPH enzyme layer on the working electrode, where they are catalytically hydrolyzed by the organophosphate hydrolase (OPH) to yield p-nitrophenol. The presence or absence of the analyte is determined based on the presence or absence of the characteristic SWV peak (in this case it occurs at +0.85 V) which corresponds to the oxidation peak of p-nitrophenol. It should be noted that this glove sensor is intended for single use, which effectively eliminates the risk of the cross-sample contamination. The special serpentine structures provide sensors with superior intrinsic stretchability and mechanical deformation adaptability, with more detailed discussion in paragraph 2 of the section 2.2 Conductive electrodes.

Ahmad et al. [118] developed zinc oxide (ZnO)-based biosensor arranged on porous and flexible substrates (i. e., carbon paper and carbon fabric) assigned for the detection of organophosphate (OP) such as paraoxon. This sensor runs three-electrode system: firstly, Ag/AgCl wire saturated in 1M KCl was used as the reference electrode, and 2 mm diameter platinum wire was used as the counter electrode. Secondly, ZnO nanostructures were electrodeposited onto carbon paper/fabric after being treated with acetone bath and ultrasound, followed by oxygen plasma treatment to enhance the wettability of the substrate for manufacturing of the biosensor. Thirdly, the working electrode was immersed in acetylcholinesterase (AChE) solution to immobilize the AChE enzyme on Zn O. The sensing mechanism of the sensor can be described as follows: firstly, the three-electrode system and buffer phosphate saline electrolyte (PBS, pH=7.4) made up an electrochemical cell. Upon the addition of AChCl to the electrolyte, the amperometric current was measured. Here, the following reactions were observed: acetylthiocholine + H₂O → AChEthiocholine + acetic acid (with AChE catalyzing, a); 2thiocholine − 2e[−] → dithio-bis-choline + 2H⁺ (b). The amperometric measurement obtained at the oxidation potential of AChCl was equal to 0.4 V. Secondly, when the sensing material was exposed to OP, AChE interacted with OP. This led to the inactivation of AChE, which could no longer catalyze the hydrolysis of AChCl to generate thiocholine. As a result, reaction (a) was inhibited, thus leading to a decrease in the rate of reaction (b) eventually manifested as a decrease in the amperometric response. It is worth to note that the ZnO biosensors produced on the carbon fabric featured the more than sufficient results, including increased sensitivity, improved stability along with the detection range for OP of 0.5 nM–5 μM .

1.3.3. Biogenic amines

High concentrations of biogenic amines (BA) found in food are formed by microbial metabolism, which can be influenced by storage conditions and temperature. Consequently, the certain amount of BA is frequently utilized as an indicator for the food quality and safety [119]. High levels of BA in food can cause food poisoning, while low levels may lead to food intolerance [120]. Therefore, it is crucial to monitor BA to gain a useful insight into the real quality and preservation conditions of food products, and as a consequence, enable risk-reduction of food waste and foodborne illnesses [121,122].

Vanegas et al. [119] developed Laser scribed graphene (LSG) amperometric biosensor for the detection of BA levels in food samples. Firstly, the three-electrode system was fabricated using a UV laser stencil engraver (laser energy density was 1.7 J cm⁻²). Ag/Cl ink was applied to the reference electrode and the solder pads of the three electrodes to pre-

vent electrode breakage due to repeated application. Subsequently, a passivation layer of fast-drying nitrocellulose lacquer (approximately 200 μm thick) was applied between the electrodes and the connectors to insulate the non-active features. Secondly, electroplated copper nanocubes were applied on the surface of the working electrode. Thirdly, the working electrode was biofunctionalized with purified diamine oxidase (DAO). The sensing mechanism can be described as follows: DAO enzyme catalyzes the deamination of primary amines, diamines, and substitutes amines via oxidation to generate aldehyde, ammonia and hydrogen peroxide (H_2O_2) [123]. H_2O_2 is then easily decomposed at the working electrode polarized at +500 mV, leading to production of an oxidative current. The following two equations can describe the chemical reaction process: $\text{R-CH}_2\text{-NH}_2 + \text{H}_2\text{O} + \text{O}_2 \rightarrow \text{R-CHO} + \text{NH}_3 + \text{H}_2\text{O}_2$ (with catalyzing DAO, a); $\text{H}_2\text{O}_2 \rightarrow 2\text{H}^+ + \text{O}_2 + 2\text{e}^-$ (at 500mV, b). This electrical response can be further correlated to the concentration of BA in the electrochemical cell via the calibration plots. It is worth to highlight that this amperometric biosensor performs well in regards of its electrochemical properties, with an average sensitivity to histamine of 23.3 $\mu\text{A}/\text{mM}$, low limit of detection of 11.6 μM , and with response time of 7.3 s. These results are comparable to those of biosensors made from materials of analytical grade.

2. Discussion

The increasingly prominent importance and huge market demand for food quality and safety, coupled with the fatal flaws exposed by traditional rigid sensors, including mechanical non-conformability, inaccurate or unreliable data collection caused by intermittent contact, and large space requirements, have led to numerous studies devoted to flexible sensors for food monitoring. Firstly, not inferior to rigid sensors, the flexible sensors also contain abundant number of transducers, such as resistors, electrochemical sensors, capacitors, etc. When flexible sensing materials encounter physical perturbations or chemical/biological analytes, their electrically-related physical properties like conductivity, work function and permittivity correspondingly change in one or more way, which changes are subsequently transformed into detectable signals by the transducer. The diversity of transduction mechanisms enables flexible sensors to operate according to a variety of sensing mechanisms, making the design of flexible sensors in principle almost unlimited. The detection indicators cover a wide range of physical parameters like temperature, humidity and pressure, as well as chemical parameters such as pH, multi-gases, pesticides, as well as the biological parameters such as foodborne pathogens and biogenic amines. Secondly, the flexible sensing materials with large specific surface area, high carrier mobility and carrier density, dense active areas, outstanding adjustability and processing capacities such as 2D carbon nanomaterials, conductive polymers and nanohybrid materials have additionally improved the sensitivity, stability and selectivity of flexible sensors. However, there are still some limitations such as high Young's modulus, poor biocompatibility and poor responsiveness [124], which make the design and development of flexible sensors quite challenging task. In this discussion we will explore the benefits, challenges and prospects of flexible sensors application for food quality monitoring.

2.1. Benefits

Flexible sensing in food monitoring has emerged as a game-changer that can control and prevent food-borne diseases, ensure consumers' health and safety, and promote the development of the food industry. Flexible sensors possess unique features, including light weight, portability, great flexibility, stretchability, foldability and adaptability, which make them more efficient and effective in food quality monitoring [124,125,126]. Flexible sensors, which refer to the combination of circuits and electronic components that can retain their functions under circumstances of geometrical bending or stretching, have a long history of innovation and still keep evolving along with the development of materials science [125,127]. Innovations in carbon nanomaterials, conductive polymers, metal-oxide based semiconductors (MOXs), 2D nanostructured materials [128,129,130,131], nanohybrid materials [78,132–138], and even optically transparent hydrogels [139], provide high sensitivity, fast response, low power consumption and long lifespan for the flexible sensors. The synergy between the material science and microfabrication technology has served as propulsion force in the advancement of flexible sensor design, with innovations in one and driving progress in the other. Flexible sensors in food monitoring precisely meet the needs of acquiring multiple parameter information in a small space, with more reasonable layout, being exposed to mechanical deformation, and conformably attaching to uneven surfaces. The application scenarios of flexible sensing in food monitoring are rapidly evolving, starting from measuring temperature, humidity, pH, multi-gases, microorganisms, pesticides, antibi-

otic residues etc. till grading of ripeness, freshness marking and so on. Herein, we systematically summarize and analyze the critical advantages of flexible sensors in the latest representative research in the field of food quality monitoring.

- Mechanical conformability: The flexibility absolutely stipulated by the flexible substrate contributes to the deformation dynamics of the sensors. Flexible substrates, represented by polymers, use these materials to accommodate strain in molecular level through the fabric of the chemical structure or adjusting the properties familiar to the polymer engineering community of professionals. Escobedo et al. [140] designed a strain sensor for monitoring the swollen food packages which is caused by food spoilage. The superior flexibility and excellent stretchability provided by substrate PDMS and sensing material PEDOT: PSS allow the sensor to be applied onto food packaging of various sizes and arbitrarily curved surfaces.
- Sensitivity: Flexible sensing materials such as 2D carbon nanomaterials, conductive polymers and nanohybrid materials provide sensors with higher sensitivity, better selectivity and lower threshold of detection due to their large specific surface area, high carrier mobility and carrier density, dense active areas, outstanding adjustability and processing capacities. Tang et al. [53] developed an ammonia sensor with Au interdigital electrodes (IDE) using PEDOT: PSS nanowires as the sensing material. The integrated NH₃ sensing system showed the enhanced performance, with a detection limit of 100 ppb, as well as high selectivity and reproducibility. Zhu Xiaoyu and colleagues [78] fabricated a self-assembled phosphorene/Ti₃C₂-MXene nanohybrid-based flexible sensor to detect ultra-trace phytohormone α -naphthalene acetic acid. It can work in a wide linear range of 0.02–40 μM with a low LOD of 1.6 nM. Yan et al. [57] developed a wearable paper-based ethylene gas sensor modified with SWCNTs-PdNPs-polystyrene microspheres composite for the real-time monitoring of fruit ripeness and corruption with a low threshold of detection of 100 ppb.
- Durability: The loading-unloading cycles imposed onto the flexible substrates can cause fatigue failure. And the mechanical deformation of flexible sensing materials can cause changes in their electrical properties as well, such as conductivity decrease. Therefore, durability and reliability is one of the key factors determining the prospects for the development of flexible sensors. Xia et al. [18] created a pressure sensor based on Ag nanowire/PDMS electrodes and a ZnO nanoparticle membrane used for grading of avocado ripeness. The sensor demonstrated stable functioning under 5,000 cycles of pressure in piezoelectric mode. The substrate did not reveal any mechanical fatigue failures that could have been caused by physical damage of the flexible sensing material that led to unfavorable changes to its resistance.
- Lightweight and thin: Flexible sensors' structure feature a very minimalist design due to direct exposure of the sensor to the analyzed material, due to mixing or stacking it on the conductive electrodes and flexible substrates without the need for additional redundant components. The flexible substrates are mostly micrometer-scale polymer films or paper-based materials. The production methods for conductive electrodes such as LSG, inkjet-printing, drop-casting, screen print, spinning coating, dip coating, electrochemical-assisted deposition (ECAD), etc., do not significantly increase the thickness of the sensor. Sensing materials, such as carbon nanomaterials, conductive polymers, nanohybrid materials, etc., are modified on conductive electrodes by laser induction, printing, drop-casting, etc., and the gained increase in thickness still remains at the micrometer level only. All of these engineering design and manufacturing methods have brought significant progress in weight and thickness of the flexible sensors, thus distinguishing them from traditional rigid sensors.
- Wireless communication: If traditional wired technology is used in the lightweight flexible sensors for data transmission, their advantages in miniaturization and portability will be greatly lost. The flexible PCB antenna-based RFID/NFC communication is currently one of the quite refined solutions of data transmission for flexible sensors. Xiao and his team [1,24] has made significant progress in integrating flexible RFID into flexible sensor systems. Escobedo et al. [140] also used NFC technology for data communication within flexible sensors, as well as the achieved operation without batteries by using an NFC reader to power the cell through a custom-designed planar coil antenna.

Overall, the benefits of the flexible sensors in food quality monitoring are immense. This concept offers an innovative solution to the challenges posed by traditional rigid sensors, and its unique properties make it well-suited for food quality and safety monitoring. As the technology still keeps developing, it holds the potential to revolutionize the food industry and improve the health and safety of the consumers worldwide.

2.2. Challenges

Although flexible sensors have great potential and advantages in food quality monitoring, yet numerous issues and challenges remain in the future research and development of high-performance flexible sensors.

- ❑ Additional contamination: For flexible sensors that come in direct contact with food or living organisms, or those that are located in the same confined space, long-term toxicity of analyses and secondary contamination due to the passive transfer of the flexible substrates (e. g., metal oxide-polymer foil, carbon paper/cloth, etc.) and sensing materials (e. g., functionalized ink, LSG, heavy metal nanoparticles, etc.) through peeling or diffusion are critical areas of research in the future. The emergence of new materials, re-design of the sensor structures, appropriate arrangement, encapsulation, and innovation in surface/interface engineering can cope with these challenges.
- ❑ Performance improvement: Enhancement of both stretchability and sensitivity is another challenge for cutting-edge flexible sensors. The emergence of new intrinsically stretchable materials, the transformation from non-stretchable materials to stretchable materials through improvements with the help of geometric engineering methods, the combination of the most recent sensing materials, the fusion of multimodal information obtained from comprehensive indicators, the fusion of multiple transduction mechanisms for single indicators, combinations of the novel processing technologies can optimize the balance between sensitivity and stretchability.
- ❑ Fabricating tolerance control: The modification or assembly of sensing materials for flexible sensors requires the use of micro-nanofabrication techniques, like inkjet printing, screen printing, thin film deposition like electrochemical-assisted deposition (ECAD), spin coating, photolithography, etc. In the mass production process it is pretty challenging to maintain consistent geometric parameters and electrical performance of the sensing materials. This leads to emergence of new challenges for the practical application and commercial production of high-performance flexible sensors according to high-precision and high-sensitivity application scenarios. The stable and reliable calibration systems and advanced data processing are viable options to cope with this challenge.
- ❑ Energy consumption and regeneration: Energy consumption and regeneration are ongoing challenges in regards to the flexible sensors. In long-term monitoring activities such as food spoilage gas detection and cold chain temperature/humidity monitoring, the reversibility of sensing materials and the energy consumption of sensing systems are crucial issues to face with. The development of novel materials with good regeneration rate and the combination of miniaturized solar cells and supercapacitors are effective choices to overcome this challenge. Shahrbabaki et al. [39] have achieved the outstanding results in the study of reversibility of flexible CO₂ sensors and provided reliable analysis of the working mechanism, which involves protonation and deprotonation. Xiao and colleagues [1,24] have achieved significant breakthroughs in the field of miniaturized power consumption and self-powered systems for the flexible sensors.

2.3. Prospects

The development of state-of-art flexible sensors for food quality monitoring is an emerging research field. Quality grading, monitoring of food spoilage, detection of foodborne pathogens, maintaining of the refrigerated supply chain marketability etc., have significant market potential in the field of food quality monitoring. Here we discuss several prospects of combining the flexible sensors for food quality monitoring with several emerging technologies. These prospects may represent feasible research directions for coping with significant demands and achieving practical development in food quality monitoring.

- ❑ Digital twins: Fresh fruits, seasonally available seafood obtained from natural wild sources/artificial aquaculture, and other typical perishable foods with a short term of expiry require strict monitoring of their quality and marketability. Digital twins technology has opened up

possibilities for real-time coupling with flexible sensor data, thus enabling the quantification of the trade-offs in maintaining food quality along with its marketability within the refrigerated supply chain [141].

- ❑ TENG-coupled: The triboelectric nanogenerator (TENG) generates electric power based on the triboelectrification (i. e., contact electrification) of two heterogenous materials and the electrostatic induction. Compared to electromagnetic and piezoelectric mechanisms, the TENG technology shows great promise due to its advantages such as simple and diverse configurations, excellent flexibility, high output performance and independence from some specific materials [142]. During food storage, lightless and dark environment is inevitable. By generating electricity through mechanical motion or friction, TENG provides the required power for the operation of the flexible sensors, and stores the excessive power in the supercapacitors. The combination of this technology enables power self-sufficiency, thus making flexible sensors more convenient and reliable in the practical applications.
- ❑ Flexible NFC chip: Installation of wireless communication chips enclosed in bulky rigid materials on the lightweight and thin flexible substrates significantly reduces the flexibility and stretchability of the sensors. Placing the flexible PCB antenna on the outer periphery of the flexible substrate, while positioning the flexible electrodes, sensing materials and flexible NFC chip within the circular area enclosed by the antenna, allows maintaining the least dimensional scales of the sensors. This highly integrated flexible sensing-communication system can greatly enhance the practicality of the flexible sensors. One midway approach is to integrate silicon-based microprocessor chips into a flexible substrate and thinning the silicon chips [143,144]. However, this method still relies on traditional high-costs manufacturing processes. Additionally, the island-like configuration cannot provide complete flexibility and stretchability to the chip. Another approach is to develop the intrinsically flexible processor. John Biggs and his team [145] at Arm have taken a groundbreaking step in the design of a flexible chip that is fabricated from metal-oxide TFTs arranged on a flexible substrate.
- ❑ Flexible on-chip spectral sensor: Vis/NIR light can penetrate into the flesh of malus, prunus, pyrus, and berry fruits, and by diffuse reflection it can collect physicochemical information such as soluble solids content (SSC), moisture content, firmness, titratable acidity (TA), extractability of anthocyanins (EA), and others with the help of the spectral information. Analyzing optical information is an important tool for food safety evaluation, quality control, and prediction of the optimal fruits harvesting date. Currently, due to the absence of flexible optical chips, researches on flexible optical sensors for food monitoring are still lacking. The emergence of flexible optical chips will bring new opportunities and possibilities in the field of flexible sensing used for food quality monitoring.

3. Conclusion

As one of the most important research direction in the field of flexible electronics, the flexible sensors have achieved remarkable results in wearable tools of health monitoring, high-sensitive detection of physicochemical parameters and the other fields. However, the development of flexible sensors for food quality monitoring still remains an emerging research area. The enormous, complex, and crucial demand for food quality monitoring encourages the urgent research on the novel cutting-edge flexible sensors. It is worth noting that the development in this emerging field greatly benefits from the cross-fertilization between the materials science, electrical engineering, information science, chemistry, physics, and energy research fields. In general, the flexible sensing and wearable technology have the potential to revolutionize the field of food quality monitoring. By providing more accurate and timely information about the quality and safety of food products, as well as assisting the individuals in making the informed decisions about their diets, they can significantly enhance the industry.

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