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Review article

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FLEXIBLE SENSORS FOR FOOD MONITORING. PART I: PRINCIPLE

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flexible sensor, food monitoring, intrinsically stretchable, mechanical conformability, conductive electrode, electrical property, sensing mechanism, transduction mechanism

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 meet the needs of acquiring information on multiple parameters in small space and more reasonable layout, providing data on mechanical deformations, and conformably attaching to arbitrarily curved surfaces. Flexible sensing materials with a large specific surface area, high carrier mobility and carrier density, dense active sites, outstanding tunability, and processability, such as two-dimensional carbon nanomaterials, conductive polymers, and nanohybrid materials, have further improved the sensitivity, stability, and selectivity of flexible sensors. This article attempts to critically review state-of-the-art developments with respect to materials, fabrication techniques, and sensing mechanisms of devices, as well as the applications of the electrically-transduced flexible sensors. In addition, this review elaborates on the transduction mechanisms of several typical transducers, with a focus on the physics behind, including the modulation of doping level, Schottky barrier, and interfacial layer that typically lead to changes in conductivity, work function, and permittivity. We also highlight the benefits, technical challenges with corresponding solutions of current flexible sensors, and discuss potential strategies to overcome limitations in energy consumption, quantify the trade-offs in maintaining quality and marketability, optimize wireless communication, and explore new sensing patterns.

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ГИБКИЕ СЕНСОРЫ ДЛЯ МОНИТОРИНГА ПИЩЕВЫХ ПРОДУКТОВ: ЧАСТЬ 1 — ПРИНЦИП

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КЛЮЧЕВЫЕ СЛОВА: АННОТАЦИЯ

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

Мониторинг и поддержание качества, безопасности и аутентичности пищевых продуктов являются наиболее важными проблемными вопросами в пищевой промышленности. Самые современные гибкие сенсоры для мониторинга пищевых продуктов точно соответствуют потребностям в получении информации по многим параметрам в небольшом пространстве и более рациональном размещении, обеспечивая данные по механическим деформациям и прилегающей соответствующим образом к произвольно изогнутым поверхностям. Гибкие сенсорные материалы с большой удельной площадью поверхности, высокой мобильностью носителя и плотностью носителя, плотными активными точками, прекрасной настраиваемостью и технологичностью, такие как двумерные углеродные наноматериалы, проводящие полимеры и наногибридные материалы, дополнительно улучшили чувствительность, стабильность и селективность гибких сенсоров. В данной статье предпринята попытка критического обзора передовых разработок в отношении материалов, методов изготовления и сенсорных механизмов устройств, а также применений гибких сенсоров с электрическим преобразованием. Кроме того, в данном обзоре рассмотрены механизмы преобразования некоторых типовых преобразователей с акцентом на лежащую в основе физику, включая модуляцию уровня легирования, барьер Шоттки и межфазный слой, которые обычно приводят к изменениям в проводимости, рабочей функции и диэлектрической проницаемости. Мы также освещаем пользу, технические проблемы с соответствующими решениями современных гибких сенсоров и обсуждаем потенциальные стратегии для преодоления ограничений в потреблении энергии, количественном определении плюсов и минусов в поддержании качества и потребительских свойств, оптимизации беспроводной связи и изучения новых сенсорных паттернов.

1. Introduction

1.1. How important is food safety?

The World Health Organization (WHO) pointed out [1] that contaminated food is responsible for causing an estimated 600 million people fall ill globally, resulting in 420,000 deaths each year [1]. Over the past few years, the increasing awareness among consumers about a healthy lifestyle has sharply raised their familiarity towards food quality and safety [2]. Food quality is a reliable indicator that relates to the consumption needs and expectations of consumers. Common food quality includes factors such as freshness, texture, ingredients, grading of physical appearance and so on. It can lead to taste, health, safety, and pleasure [3]. Food products with high quality are always expected and demanded by consumers [4]. Food safety problems typically include chemical pollution, microbial pollution, and physical pollution [2]. Bacteria, viruses, parasites

and fungi existing in the environment may cause diseases of consumers, and they can also easily contaminate the consumable food materials [5]. During the past decades, pesticides have been widely used for high yield productions [6], while the increasing usage of various pesticides can lead to high levels of residues in foodstuff and accumulation in the food chain, which poses a huge threat to human health [7,8].

1.2. What are the key concerns in food monitoring?

With the globalization of economy and trade, and the rapid circulation of various food products around the world, potential food contamination, and fraudulent food manufacturing have prompted consumers to pay more attention to the quality and safety of their food [9]. All this has generated the urge to develop food monitoring systems that can control and prevent food-borne illnesses, ensure consumer health and

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safety, and promote the development of the food industry. In recent years, significant progress has been made in food quality and safety monitoring based on traditional laboratory analysis and rigid sensor detection. Food quality detection involves measuring various parameters such as temperature [10], humidity [11], pH [12], flavor [13], multi-gas [14] and freshness marking [15]. On the other hand, food safety monitoring involves detecting microorganisms [16], pesticides [17], illegal additives malachite green [18], melamine [19], antibiotic residues [20] and so on. However, traditional rigid sensors exhibit several challenges in food monitoring. First, the intrinsic mechanical non-conformability property of rigid sensors makes them difficult to adapt to arbitrarily curved surfaces and shape changeable parts (or movable parts). Secondly, rigid sensors are large, unbendable, and heavy, which makes effective integration with detection targets difficult. Thirdly, rigid sensors may not maintain consistent contact, resulting in inaccurate or unreliable data collection. Finally, manufacturing process of rigid sensors is extremely complex and contaminated, and they cannot be manufactured in individualized and small batches [21,22].

1.3. Flexible sensing have emerged as a prominent technological advancement in the realm of flexible electronics

Compared to conventional rigid sensors, flexible sensors play a crucial role in the application of flexible electronics, which possess unique advantages such as light weight, portability, great flexibility, stretchability, foldability, and adaptability [23–25]. Flexible electronics refers to circuits and electronic components that can retain their functions under circumstances of bending or stretching [26–28]. The concept of flexible electronics was introduced in the 1960s when copper was patterned on polyimide, resulting in a reduction in the thickness of the solar cell from 400 to 100 μm and leading to an unprecedented leap forward in power density [29–31]. Ever since then, innovations in materials with greater flexibility and large processability, such as conductive polymers [32–34], organic semiconductors [35,36], and amorphous silicon [37,38], have gradually laid the foundation for flexible electronics. With the rapid development of material science, flexible electronics have recently ridden the wave of carbon nanotubes [39,40], graphene platelets [41,42], precious metal nanomaterials (e. g., silver NWs, platinum NPs, etc.) [43,44,45], nanohybrid materials (e. g., CNT-PDMS composites) [46] and even optically transparent hydrogels [24], which brings high sensitivity, fast response, low power consumption, and long lifespan to flexible sensors. With the unique characteristics such as ultrathin, low modulus, light weight, high flexibility, and stretchability [47], flexible sensors precisely meet the needs in food monitoring: acquiring multi-parameter information in a small space, a more reasonable layout, providing data on packaging deformation [48], and conformably attaching to the surface of the skin [47]. Simultaneously, the new generation of information technology represented by wearable Internet of Things (W-IoT) [49], blockchain [50] and cloud computing, have generated immense interest for the nascent flexible sensing because it can integrate and connect personnel, processes, data, and equipment enabling the high adaptability [51], high precision, multiple scale, dynamic non-destructive monitoring [49]. Flexible sensing in food monitoring has become increasingly significant in recent years [44,52,54,55], and is rapidly evolving in its application scenarios [44,48,52,53,54,55], and manufacturing methodology [44,53]. The synergy between flexible sensing and developments in material science [26–46] and microfabrication technology has been instrumental in the success of both fields with innovations in one driving progress and in the other.

In this review, we systematically discuss the configurations, sensing mechanisms, and application of flexible sensors in food monitoring. First, we summarize the basic architecture regarding flexible substrates, conductive electrodes, and sensing materials focusing on the unique properties (e. g., mechanical, electrical, chemical properties, etc.) of materials and the fabrication techniques. Secondly, we elaborate the sensing mechanisms and transduction mechanisms of several typical transducers (i. e., resistor, electrochemical sensor, and capacitor). Thirdly, we comprehensively discuss the applications of flexible sensors in the categories of physical, chemical, and biological aspects. Finally, we provide an overview of the benefits and technical challenges of current flexible sensors, and highlight the potential strategies to improve the performance of state-of-the-art flexible food monitoring sensors.

2. Principle of food monitoring flexible sensors

To understand why sensitive detection of physical perturbations, chemical analytes, and microbes can be realized in flexible sensors, and how sensing materials work (e. g., resistive [58], capacitive [59], piezoelectric [44,60], potentiometric [61], amperometric [62,63], impedimetric [63] sensors, etc.), it is necessary to know the physics behind. A typical flexible

food monitoring sensor contains a flexible substrate, and two functional components: a conductive interconnect/electrode [64,65,66,67] (hereinafter referred to as “conductive electrode”) which connect the transducer and the output interface of data and an active sensing material-equipped flexible transducer (i. e., transducing the concentration of analytes or a physical perturbation, such as temperature, into an electrical signal). In this section, we will systematically introduce the configurations, materials, and working mechanisms of flexible sensors for food monitoring.

2.1. Flexible substrates

Flexible sensors have intrinsic characteristics including the ability to bend [44,51–55], fold [51,55], stretch [68,69,70,71], twist [72], and even self-heal [56,73] if damaged. Flexible substrates are the main contributor to the deformation dynamics of the sensors. Conventional rigid substrates such as silicon [74], plastic [75], Al_2O_3 [76], etc., have the advantages of simple structure, convenient preparation and reliable responsiveness. However, the rigidity hinders the capture of analytes and results in poor signal transduction [77], which limits the performance of conventional sensors. Flexible food monitoring sensors, whether attached to the living organisms’ surfaces [44,52,55,78] or integrated inside or outside of packaging [58], need to be lightweight, small, and easy to use [67]. Flexible substrates, taking polymers as an example, are intrinsically or molecularly stretchable materials, which use the materials themselves to accommodate strain [79]. At the molecular level, mechanical softness can be determined by two classifications: tailoring of the chemical structure (e. g., the lengths and composition of the side chains and rigidity of the backbones) and tuning properties familiar to the polymer engineering community (e. g., molecular weight, polydispersity, and cross-linking) [80]. The use of flexible substrates can greatly enhance the functionality, durability, and versatility of flexible devices, making them ideal for use in food monitoring. Among various materials used as substrates for flexible sensors in food monitoring, polymers [51,58,81,82] dominate as the most commonly employed base material, with a few exceptions using materials such as paper [83], carbon paper [84], carbon cloth [84], and others [74]. In this section, we will discuss several typical types of polymer substrates and paper-based substrates. Although paper and carbon-paper substrates are infrequently used as substrate materials, we will introduce them together in the same section to provide an overview of their potential use.

2.2. Polymer substrates

In food monitoring, polymers represent the predominant substrate material for flexible sensors, accounting for a significant majority of their composition. In addition to high flexibility, adaptability and low cost, polymer substrates offer a range of unique advantages for flexible sensor components, including biocompatibility (e. g., PEDOT: PSS, HPU, PLA, PDMS, PET, etc.) [63,85,86,87], elasticity (e. g., PDMS, PEDOT: PSS, HPU, rubber, etc.) [44,58,63,88], and intrinsic stretchability (e. g., PEDOT: PSS, PDMS, HPU, rubber, nitrile, etc.) [44,89,90]. Biocompatibility is highly advantageous for direct contact measurements between sensors and living organisms; elasticity and intrinsic stretchability allow sensors to adapt to irregular deformations of the analytes’ surfaces, obtaining more reliable data. The polymers used for flexible substrates can be classified into the following categories based on their chemical structure and properties: (1) polyimide: PI (polyimide) [51,81,91]; (2) polyester: PET (polyethylene terephthalate) [59,92], PEN (polyethylene naphthalate) [93]; (3) polysiloxane: PDMS (polydimethylsiloxane) [44,58]; (4) biodegradable: PLA (polylactic acid) [87]; (5) rubber: rubber [88], nitrile (nitrile rubber) [89,90]; (6) polyurethane: HPU (hydrophilic polyurethane) [63]; (7) epoxy: epoxy (epoxy resin) [82]; (8) polyethylene: PE (polyethylene) [82], PVC (polyvinyl chloride) [58], OPP (oriented polypropylene) [62]. Herein, we will discuss several typical polymers regarding their physical, chemical, and possibly biological properties.

Polyimide (PI): In the polymer family, polyimide (PI) has some notable properties. For example, PI is a high-temperature resistant polymer, with excellent mechanical and electrical properties. In the field of laser direct scribing for flexible PCB [53,58,81], PI is a commonly preferred substrate material due to its favorable properties. Xiao and his team [51] proposed and developed a flexible battery-free wireless electronic system (FBES) for food monitoring. The FBES was fabricated by laser direct scribing on commercial PET/PI/Cu film. PI also has good chemical resistance and can withstand exposure to many solvents and chemicals. Schöning et al. [94] developed calorimetric gas sensors on PI films for more precise detection of gaseous H_2O_2 over a wide H_2O_2 concentration range. In addition, the nature of the PI chemical repeat units plays a key role in the fabrication of LDG electrodes [95]. Ever since it was first discovered in 2004 by Geim and Novoselov, graphene has garnered significant attention from the scientific community owing to its unparalleled properties [96,97,98].

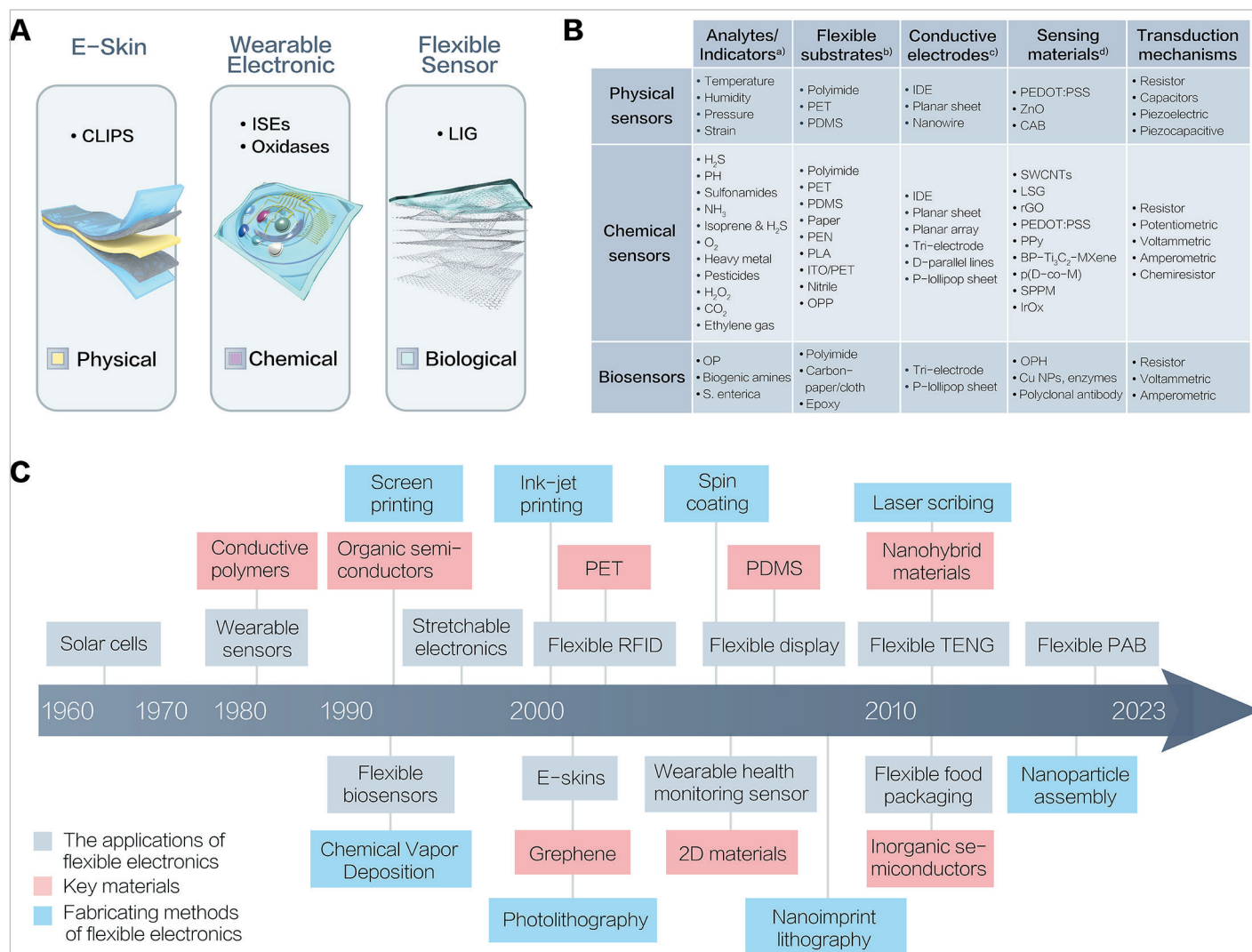


Figure 1. Flexible sensors application architecture. (A) Three typical categories of flexible electronics using physical, chemical, and biological sensing mechanism, respectively. E-Skin: Adapted with permission [56]. Copyright 2022, Nature Publishing Group. CLIPS: Cl-functionalized iontronic pressure sensitive material. Wearable electronics: Adapted with permission [57]. Copyright 2016, Nature Publishing Group. ISEs: Ion-selective electrodes. Flexible sensor: The PI film was patterned using laser scribing to obtain 3D porous LIG. Then, liquid PDMS was drop-casted and heated for a specific period of time. Finally, the side of PDMS/LIG was peeled off to obtain the stretchable LIG-based electrode. LIG: Laser induced graphene. (B) Summary of monitoring indicators, materials, transduction mechanisms, etc., of three types of flexible sensors (i. e., physical, chemical, and biological) used for food monitoring. (a) OP: Organophosphate. (b) PET: Polyethylene terephthalate, PDMS: Polydimethylsiloxane, PEN: Polyethylene naphthalate, PLA: Polylactic acid, ITO: Indium Tin Oxide, OPP: Oriented polypropylene. (c) IDE: Interdigitated electrodes, Tri-electrode: three electrode system, D-parallel lines: Double parallel lines, P-lollipop sheet: This is a new definition here that refers to a flat shape resembling a lollipop. (d) PEDOT: PSS: Poly(3,4-ethylenedioxythiophene) polystyrene sulfonate, CAB: Cellulose acetate butyrate, SWCNTs: Single-walled carbon nanotubes, LSG: Laser scribed graphene, rGO: Reduced graphene oxide, PPY: Polypyrrole, BP-Ti₃C₂-MXene: Two-dimensional phosphorene (BP) nanohybrid with graphene-like titanium carbide MXene (MXene, 2D material, 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)), p(D-co-M): poly(N-[3-(dimethylamino)propyl]-methacrylamide-co-2-N-morpholinoethyl methacrylate), SPPM: SWCNT-PdNP-polystyrene microsphere, IrOx: Iridium oxide, OPH: Organophosphorus hydrolase, NPs: Nanoparticles. (C) Timeline of major events in flexible electronics development. PAB: Polyclonal antibody biosensor, TENG: Triboelectric nanogenerator.

Рисунок 1. Архитектура применения гибких сенсоров. (A) Три типичных категории гибкой электроники с использованием физического, химического и биологического сенсорного механизма, соответственно. Электронная кожа (E-Skin): адаптировано с разрешения [56]. Copyright 2022, Nature Publishing Group. CLIPS: Cl-функционализированный ионотронный чувствительный к давлению материал. Носимая электроника: адаптировано с разрешения [57]. Copyright 2016, Nature Publishing Group. ISEs: ионоселективные электроды. Гибкий сенсор: Пленка PI была структурирована, используя лазерное скрайбирование для получения 3D пористого LIG. Затем был нанесен жидкий PDMS методом литья каплями и нагрет в течение определенного периода времени. Наконец, была отделена сторона PDMS/LIG для получения растяжимого электрода на основе LIG. LIG: лазерно-индуцированный графен. (B) Краткое представление мониторинговых индикаторов, материалов, механизмов преобразования и т. д. трёх типов гибких сенсоров (т. е., физический, химический и биологический), используемые для мониторинга пищевых продуктов (a) OP: органофосфат. (b) PET: полиэтилентерефталат, PDMS: полидиметилсилоксан, PEN: полиэтиленнафталат, PLA: полимолочная кислота, ITO: Оксид индия-олова, OPP: ориентированный полипропилен. (c) IDE: гребенчатые электроды, Tri-electrode: трех электродная система, D-parallel lines: двойные параллельные линии, P-lollipop sheet: Это новое определение здесь, которое относится к плоской форме, напоминающей леденец на палочке. (d) PEDOT: PSS: поли(3,4-этилендиокситиофен) полистиролсульфонат, CAB: ацетобутират целлюлозы, SWCNTs: одностенные углеродные нанотрубки, LSG: лазерно-скрайбированный графен, rGO: восстановленный оксид графена, PPY: полипиррол, BP-Ti₃C₂-MXene: наногибрид двумерного фосфорена (BP) с графено-подобным карбидом титана MXene (MXene, 2D материал, может быть синтезирован путём травления “A” из фазы MAX (“M” представляет собой переходные металлы, “A” представляет собой группу элементов IIIA/IVA и “X” представляет собой C и/или N элементы)), p(D-co-M): поли(N-[3-(диметиламино)пропил]-метакриламид-ко-2-N-морфолиноэтил метакрилат), SPPM: SWCNT-PdNP-полистироловая микросфера, IrOx: оксид иридия, OPH: фосфорорганическая гидролаза, NPs: наночастицы. (C) временная шкала основных событий в разработке гибкой электроники. PAB: биосенсор поликлональных антител, TENG: трибоэлектрический наногенератор

Gomes et al. [99] developed an impedimetric immunosensor based on LIG electrodes functionalized with specific antibodies for the detection of *Salmonella enterica* using laser-induced porous graphene produced from PI, showing potential in the sensing field.

Polyethylene terephthalate (PET): PET has gained widespread popularity as a platform for food monitoring sensors, owing to its excellent mechanical properties (i. e. bendability, durability, etc.) [100], low cost, commercial availability [59], and outstanding adhesion with nanowires (mixed with PDMS, ZnO, etc.) [100] and functional ink materials (silver-nanoparticles ink) [59]. Duan et al. [100] developed a flexible ammonia sensor with highly aligned conducting polymer nanowires fabricated by a capillary filling-based soft lithography technique on a PET substrate. Owing to the PET substrate's superior mechanical stability and adhesion ability to nanowires, after 1200 bending cycles, no physical damage to the sensing material (flexible nanowire) was caused by mechanical fatigue failure of the substrate, which could have resulted in unfavorable resistance changes of a sensor. Rooij et al. [59] developed humidity sensors equipped with planar interdigitated electrodes (IDE) capacitors. The extraordinary adhesion between silver ink and PET substrate permitted the growth of thicker (up to 15 μm) layers of Ni on substrate. However, PET substrates are unsuitable for stretchable sensing platforms due to their relatively higher tensile strength (2–4 GPa) [65,67], and high temperature (>100 °C) [101] material processing is not possible on it because of less thermal stability.

Polydimethylsiloxane (PDMS): PDMS is a commonly used flexible substrate material of flexible sensors for food monitoring due to its unique mechanical properties [102], remarkable transparency (under visible spectrum) [58], extraordinary biocompatibility, and resistance against chemicals such as water and the majority of alcohols and bases [58]. Escobedo et al. [58] fabricated a smart strain tag through injecting an active material into a microchannel made from PDMS. This flexible strain sensor exhibited a considerably good performance with an average gauge factor of up to 13000, and an average degree of hysteresis (DH) within the range of < 9%. PDMS was also applied as a protective layer for flexible PCB (preventing the oxidation of the Cu circuit) [51] and NFC tag (encapsulating and protecting against moisture, liquid, etc.) [58]. Xia et al. [44] fabricated a flexible dual-mechanism pressure sensor, where PDMS was employed as the substrate due to its exceptional elasticity, as well as the cross-linking matrix due to its chemical stability and low relative permittivity. This sensor demonstrates a long-term stability of over 5000 pressure cycles and exhibits satisfactory linearity, repeatability, and stability when detecting pressures in the range of 0–100 kPa.

2.3. Paper-based substrates

Just like the knight Edgar, who has fewer appearances but is essential in Shakespeare's King Lear, paper-based substrates also play a crucial role in the fabrication of flexible food monitoring sensors. Paper-based substrates possess excellent properties including flexibility [109], breathability [66], hygroscopy [110], ionic conductivity modulation [110] and easily forming cross-linked structure with nanosized sensing materials [83], which makes them ideal for use as a lightweight, low-cost, recyclable, biocompatible starting substrate [83,111]. Tang et al. [83] designed a paper-supported H_2S sensing electrode through loading Cu_2O -polypyrrole conductive aerogel (Cu_2O -PPy). The flexible electrode exhibits excellent H_2S sensing and egg spoilage monitoring performance owing to the synergistic effect between the different components and 3D porous architecture. Padalkar et al. [84] developed a ZnO-based biosensor on flexible and porous carbon paper/ cloth to detect organophosphates (e. g. paraoxon). Due to the three-dimensional substrate and the morphology of the nanomaterials, the path length for charge carriers was significantly reduced, leading to a decrease in recombination losses and an increase in electrical

conductivity. As a result, the device exhibited higher sensitivity and lower LOD values. Table 1 summarizes the mechanical, physicochemical and thermal properties of the four most used materials of substrates in fabrication of food monitoring sensors [104,105].

2.4. Conductive electrodes

Conductive electrodes play a crucial role in the fabrication of flexible sensors [67,118]. The function of the conductive electrode is to collect electrical signals from the sensor and transmit them to a processor or other circuit for analysis and processing. They are usually made of conductive materials such as metal nanoparticles/nanowires (i. e. Au [100,113], Ag [58], Ni [59], etc.), conductive ink compound (e. g., Ag/Cl ink, carbon ink, etc.) [114], conductive polymer (e. g. PEDOT: PSS [63], polypyrrole [83], etc.), laser-derived graphene (LDG) [91,99], graphene-carboxymethyl cellulose(G-CMC) [119], etc. The design and fabrication of conductive electrodes have a significant impact on the performance and sensitivity of sensors.

The fabrication of conductive electrodes and device arrays have been investigated via three different approaches:

1. flexible electrodes were fabricated with a micrometer-scale thickness [59] using non-stretchable materials such as metal [58,100,113], polymer [91,99], and paper [83] through various processing techniques including inkjet printing [59], screen printing [87], laser irradiation [92], photolithography [113], etc.;
2. stretchable electrodes were obtained by intrinsically stretchable materials (e. g. PEDOT: PSS [63] and polypyrrole [83]);
3. stretchable electrodes were developed through i) geometric engineering of non-stretchable but flexible materials and ii) mixing conductive ink with intrinsically stretchable polymers such as silicone elastomer and polystyrene-block-polyisoprene-block-polystyrene (PS-PI-PS) [90].

Molina-Lopez et al. [59] developed a functional humidity sensor with planar interdigitated electrodes (IDE) using an all-additive approach. They achieved this by ink-jetting the silver nanoparticle ink onto a PET substrate and electrodepositioning nickel on silver with nickel thickness ranging from a few hundred nanometers to 15 μm on PET. High sensor stability was obtained by passivating printed silver electrodes with nickel. This prevented silver from oxidizing or releasing any possible trapped organic residue after interacting with humidity, and also provided extra stiffness to reduce strains on the electrode plane. Escobedo et al. [58] fabricated a strain sensing tag by injecting intrinsically stretchable conductive polymer PEDOT: PSS into the microchannel shaped by a copper mold in a PDMS substrate. Mishra et al. [90] designed a wearable organophosphorus (OP) glove biosensor via printing Ag/AgCl and carbon-based serpentine structures on the glove. This excellent geometric structural design allows the electrode to withstand extreme mechanical deformations while detecting organophosphate (OP) nerve-agent compounds on suspicious surfaces and agricultural products. Table 2 summarizes the materials, fabricating methods and geometry forms of conductive electrodes in fabrication of flexible sensors for food monitoring.

3. Flexible sensing materials

Flexible sensors require the integration of various crucial components, including sensing materials that can be incorporated within or stacked onto flexible substrates [120]. Sensing materials play a vital role in the performance of flexible devices [67], enabling them to detect and respond to stimulation of analytes or physical perturbation. The electrical and physicochemical properties of sensing materials determine the sensitivity, stability, and selectivity of the sensors. Therefore, the development and incorporation of advanced sensing materials are essential for the fabrication of flexible sensors. Conventional sensing

Table 1. Summary of the mechanical, physicochemical and thermal properties of typical flexible substrates used in food monitoring sensors

Таблица 1. Краткое представление механических, физико-химических и термических свойств типичных гибких субстратов, используемых в сенсорах для мониторинга пищевых продуктов

Materials ^{a)}	Flexibility/ Stretchability	Young's modulus [MPa]	Tensile strength [MPa]	Thermal stability / Coefficient of thermal expansion [K ⁻¹]	Chemical resistance	Transparency	Ref.
PI	Flexible	2–3 × 10 ⁵	70–150	Resist temperature (<450 °C) 3–6 × 10 ⁻⁵	Weak acids, alkali	Low	[101,103, 104,105]
PET	Flexible	2–4 × 10 ⁵	80	Resist temperature (<100 °C) 2–8 × 10 ⁻⁵	Dissolvable in acetone	High (>85%)	[101,103, 104,105]
PDMS	Stretchable	1.8 ^{b)}	6.7 ^{b)}	Resist temperature (<100 °C) 30–31 × 10 ⁻⁵	Ethanol and acetone	High (>95%)	[106,107]
Paper	Flexible	2.1–2.8 × 10 ⁵	40–60	Resist temperature (<100 °C) 0.2–1.6 × 10 ⁻⁵	No	No	[101,108]

^{a)} PI: polyimide; PET: polyethylene terephthalate; PDMS: polydimethylsiloxane; ^{b)} SYLGARD184 silicone elastomer.

Table 2. **Materials, fabricating methods and geometry forms of conductive electrodes in fabrication of food monitoring sensors**
Таблица 2. Материалы, методы изготовления и геометрические формы проводящих электродов при изготовлении сенсоров для мониторинга пищевых продуктов

Analytes/Monitoring indicators ^{a)}	Materials ^{b)}	Methods of fabrication ^{c)}	Geometry forms ^{d)}	Ref.
Temperature	Silver conductive paste	Stencil printing	Planar serpentine structures	[58]
Humidity	Ag NPs ink Ni	Inkjet printing Electroplating	IDE	[59]
Pressure	PDMS Ag NWs	Spinning coating Drop-casting	Planar sheet	[44]
Strain	PEDOT: PSS	Injecting	Nanowire	[58]
H ₂ S	Cu	Commercial Cu foil	Planar sheet	[83]
PH	Au, Cr IrOx Ag/AgCl	e-beam evaporation Sol-gel process Electroplating	Planar double electrode system	[81]
	ITO	Laser exfoliating and Evaporating patterning	Planar lollipop sheet	[61]
	PDDA-IrOx	LbL IJP and alternate deposition	Planar sheet	[60]
Sulfonamides	LIPG	Laser scribing	Three-electrode system	[112]
NH ₃	Au	Evaporating through copper shadow mask	IDE	[100]
Isoprene and H ₂ S	ITO Au	Photolithography Magnetron sputtering	IDE	[113]
O ₂	Ag-OPP cathode Zinc anode	Commercial available silver-deposited OPP film Commercial available zinc film	Planar sheet	[62]
Heavy metal	Ag/Cl conductive ink Carbon conductive ink	Printing	Planar array	[114]
Pesticides	Carbon ink Ag/AgCl	Screen printing Coating	Three-electrode system	[87]
	LIPG	Laser scribing	Three-electrode system	[115]
	Carbon ink and CSS/PCBN	Screen printing and drop-casting	Three-electrode system	[88]
	ZnO nanostructures and Ag/AgCl	Electrodepositing	Three-electrode system	[84]
H ₂ O ₂	PEDOT: PSS/HPU/HRP	Screen printing	Three-electrode system	[63]
CO ₂	LSG Ag/AgCl	Laser irradiation Electrodeposition	Three-electrode system	[92]
	Carbon black	Printing	Planar double parallel lines	[116]
Ethylene gas	Gold foil	Laser direct writing	IDE	[117]
OP	Ecoflex, carbon ink, and PS-PI-PS	Screen printing	Three-electrode system with serpentine structures	[90]
Biogenic amines	LSG	Laser scribing	Three-electrode system with	[91]
Salmonella enterica	LSG	Laser scribing	Planar lollipop sheet	[99]

^{a)} OP: Organophosphate.

^{b)} Ag NPs: Ag nanoparticles, Ni: Nickel, PDMS: Polydimethylsiloxane, Ag NWs: Ag nanowires, PEDOT: PSS: Poly(3,4-ethylenedioxythiophene) polystyrene sulfonate, IrOx: Iridium oxide, ITO: Indium Tin Oxide, PDDA: Polydiallyldimethylammonium, LIPG: Laser induced porous graphene, OPP: Oriented polypropylene, CSS: Carbon spherical shells, PCBN: Printex carbon nanoballs, HPU: Hydrophilic polyurethane, HRP: Horseradish peroxidase, LSG: Laser scribed graphene, Ecoflex: Ag/AgCl ink with platinum-catalyzed silicone elastomer, PS-PI-PS: Polystyrene-block-polyisoprene-block-polystyrene.

^{c)} LbL IJP: Layer-by-layer inkjet printing.

^{d)} IDE: Interdigitated electrodes, Planar lollipop sheet: This is a new definition here that refers to a flat shape resembling a lollipop, formed by a combination of a circle and a rectangle.

materials are modular, generally semiconducting and conducting inherently, changing their electrical properties upon exposure to various application scenarios [67]. Although their electronic performance exhibits high carrier mobility and stability, compared to novel nanomaterials such as conductive polymers, they show weaknesses in physical and chemical properties: high Young's modulus, poor controllability and tunability of chemical composition and molecular structure [120]. The sensing materials need to be lightweight, low-cost, and well compatible with large scale processing [67]. With advancement in nanotechnology, carbon nanomaterials [67,121], conductive polymers [122], nanohybrid materials, metal nanomaterials (e. g., AuNPs, BiNPs, etc.) [114] have been demonstrated to be promising sensing materials in flexible electronics applications. Numerous materials such as laser-scribed/induced graphene (LSG/LIG) [92,95,99,123,124,125], laser-induced porous graphene (LIPG) [112,115], graphene-CMC (carboxymethyl cellulose) [119], SWCNT-PdNP-polystyrene microsphere (SPPM) [117], carbon spherical shells (CSS) [88], CuxO-PPy@GO aerogel [83], PEDOT: PSS [44,58,126,126,128], poly(N-[3-(dimethylamino)propyl]-methacrylamide-co-2-N-morpholinoethyl methacrylate) (p(D-co-M)) [116], Ti3C2-MXene/BP nanohybrid [112], AuNPs [114], iridium oxide (IrOx) nanoparticles [60], etc., have exhibited remarkable performance in various research settings. In this section, we focus on three categories of nanomaterials: carbon nanomaterials, conductive polymers, and

nanohybrid materials. The electrical, physical, mechanical, and chemical properties, fabrication methods, and sensing mechanisms of sensing materials are explained.

3.1. Carbon nanomaterials

The unique electronic, mechanical, and chemical properties of carbon nanomaterials make them very interesting for developing the new generation of miniaturized, low-power, ubiquitous sensors [121,129]. Low-dimensional carbon structures, with the majority of their atoms exposed to the surroundings, provide a large specific surface area that can be advantageous in achieving high sensitivity [121,130]. In the last few years, nanomaterials like carbon spherical shells (CSS), single-walled carbon nanotubes (SWCNTs), laser-scribed graphene (LSG), laser-induced porous graphene (LIPG), laser-scribed graphene oxide (LSGO), reduced graphene oxide (rGO), etc., have become the most studied carbon-based materials for developing food monitoring sensors. In this part, we will discuss two types carbon-based nanomaterials systematically.

Single-walled carbon nanotubes (SWCNTs): After their discovery in 1991 by Iijima and Ichihashi, carbon nanotubes (CNTs) have been extensively researched for their unique electrical, physical, mechanical, and chemical properties in the development of high-performance sensing devices [131]. Sensors equipped with SWCNTs exhibit reversible stretchability, fast response and substantially higher sensitivity at

room temperature [66,132], owing to their high carrier mobility, excellent physical properties, ease of modification, and sizeable surface-area-to-volume ratio, which provides a huge number of active sites for analytes [133,134,135,136]. Yan et al. [117] developed a flexible paper-based chemoresistive sensor by modifying a SWCNT-PdNP-polystyrene microsphere (SPPM) composite (SPPM/FWPCS) for the low-cost and online detection of C_2H_4 from banana. SWCNT-PdNP-PM composites were synthesized by a method of self-assembly under van der Waals forces, and then was pipetted on the sensing area of the interdigitated electrodes and dried at room temperature. The sensing mechanism involves the release of trapped electrons due to the oxidation and cleavage of ethylene, resulting in a decrease in resistance. More specifically, the Schottky barriers between SWCNTs, PdNPs, and adsorbed oxygen in the sensing composite create an electron depletion layer, and adsorbed oxygen (O_2^-) is formed during the process. The concentration of C_2H_4 can be detected down to 100 ppb (subppm level). The detection range of a rigid sensor is the total amount of ethylene in the entire space, which, coupled with diffusivity of gases, leads to hysteresis for the monitoring of fruit ripeness and corruption. Flexible sensors can be used to overcome this limitation by placing them inside fruit packaging.

Laser-scribed graphene (LSG): The discovery of graphene in 2004 by Geim and Novoselov generated great scientific interest because of its excellent properties [137–141], including rapid electron mobility, self-assembly behavior, excellent conductivity, large specific surface area, high mechanical strength and stability, etc. [141–149]. Graphene is a single layer of sp^2 -hybridized carbon atoms covalently bound together in a honeycomb lattice [140]. First, its outstanding electronic properties, such as high carrier mobility and high carrier density, make it a promising candidate for fabricating high-performance electrically-transduced analytical devices [64,148]. Secondly, the surface properties of graphene enable it to interact with a wide range of analytes through different mechanisms such as van der Waals forces, electron transfer, or covalent bonding [140,149], which can lead to changes in the conductivity of the sensing layer. Thirdly, graphene's high surface-to-volume ratio implies that each carbon atom in graphene functions as a surface atom, resulting in the maximum possible surface area per unit volume. As a result, the charge transport in graphene is extremely sensitive to its chemical surroundings [149,150]. Fourthly, flexible electronic devices often undergo mechanical stress and deformation during operation, so they require materials that can withstand these stresses without breaking or losing their functionality. Graphene, which has high mechanical strength and stability, is therefore a promising material for use in flexible electronic devices. By using graphene as a base material, flexible electronics can be designed with improved durability and reliability, even under harsh operating conditions [146,151]. Traditional methods for producing graphene, such as thermal decomposition [152,153], mechanical exfoliation [154,155], and chemical vapor deposition (CVD) [156,157] are time-consuming and complex. In contrast, laser-derived graphene (LDG) technology offers an easy, mask-free, and low-cost alternative for the production of graphene-based materials. The products are now mainly termed laser-scribed graphene (LSG) or laser-induced graphene (LIG), etc. [95]. Aparicio-Martínez et al. [92] developed a novel, flexible and non-enzymatic electrochemical H_2O_2 sensor based on a laser-scribed graphene (LSG) electrode decorated with silver nanoparticles (LSG-Ag). They fabricated a graphene electrode on a PET substrate that was coated with GO, using a 780 nm 5 mW infrared laser from a DVD unit. And then, they decorated the LSG electrode with silver nanoparticles (LSG-Ag). The changes in morphology caused by the laser treatment, such as increased roughness, expansion, surface defects, and exposure of edge planes, have a significant impact on the electrochemical performance of the material [123,158,159]. Specifically, during silver electrodeposition, defects and features of LSG acted as nucleation sites and induced a high density of non-spherical nanoparticles (sizes from 70 to 120 nm) with uniform distribution over the LSG layers. The sensing mechanism of this amperometric sensor can be described as follows: the sensing material's role is to interact with the analyte through the active sites on its surface, while the electrode is maintained at a fixed potential and the cathode current is monitored over time. The cathode current magnitude is related to the concentration of the analyte present. The morphology features of LSG such as 3D architecture, surface defects, exfoliation, edge plane exposure and oxygen removal were combined with the electrocatalytic activity of AgNPs, resulting in an overall enhanced non-enzymatic H_2O_2 detection. The LSG-Ag sensor demonstrated low LOD of 7.9 μM , rapid amperometric response within 3 seconds, as well as high repeatability of 3% R.S.D. and reproducibility of 4.5% R.S.D. It also showed minimal loss of performance even after continuous bending.

3.2. Conductive polymers

Conductive polymers (CPs) have been acknowledged as a distinctive category of organic materials possessing exclusive electrical and optical characteristics comparable to those of inorganic semiconductors and metals [160]. CPs can be synthesized through simple, versatile, and cost-effective methods. Furthermore, they can be easily assembled into supramolecular structures with multifunctional capabilities through electropolymerization processes [161]. The development of various methodologies has enabled the modification and tuning of CPs to incorporate them into the fabrication of stretchable sensors, which include both physical and electrochemical sensors. Such novel innovations are highly desired and sought after in various fields of flexible sensing, including temperature monitoring, strain sensing, and the high-sensitivity active gas sensing (e. g., H_2S), as they hold the potential to pave the way for future breakthroughs. Conjugated π polymers are a class of materials with electrons held in their backbones [162], such as poly(3,4-ethylenedioxythiophene) (PEDOT) and polypyrrole (PPy). The unsaturated backbone of conductive polymers allows for the delocalization of π -electrons, which facilitates the movement of charge carriers along an electrical pathway [163,164]. Flexible sensing materials that incorporate CPs exhibit RT-sensing capabilities and ease of chemical modification. Specifically, the high flexibility of polymeric materials enables the sensing materials to be easily deposited or patterned onto flexible or stretchable substrates, and they can withstand mechanical deformation during use, thus maintaining the integrity of the sensing layer [67]. As typical examples of conductive polymers, PEDOT: PSS and polypyrrole (PPy) will be discussed here.

Poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT: PSS): polythiophene is a fascinating material due to its persistent conductivity and impressive electrical conductivity ($10^3 S cm^{-1}$), which is influenced by the dopant and polymerization type [165,167,167]. PEDOT can be polymerized through electrochemical or oxidative chemical methods, and it is a polymer with high electrical conductivity [160]. Compared to PPy, PEDOT possesses superior chemical [168–170] and thermal [170] stability and has been postulated for possible use as an interfacing agent [160]. PEDOT can be hybridized with poly(styrenesulfonate) (PSS) to improve electrical conductivity [171]. PSS was used for doping the PEDOT for film fabrication by spin coating and vapor phase polymerization [160]. The conductivity mechanism of PEDOT: PSS is based on the synergistic effect between PEDOT and PSS, where PEDOT serves as the conductive backbone providing the pathway for electron transfer, while PSS acts as the electrolyte stabilizing the polymer chains of PEDOT and regulating the conductivity. The highly ordered hybridization between PEDOT and PSS results in PEDOT: PSS composite materials with high conductivity. Conductive polymers are a popular choice for temperature sensing due to their ease of processing and excellent electrical properties, although they may be unstable at high temperatures [172]. However, PEDOT: PSS stands out as one of the most stable organic conductive polymers with electrical properties similar to those of a metal or semiconductor [173,174]. Escobedo et al. [58] designed a temperature sensing tag by printing silver electrodes on a flexible polyvinyl chloride (PVC) substrate and then drop-casting PEDOT: PSS on silver electrodes with a gap of 2 mm. With an increase in temperature, the mobility of carriers within the sensing layer is predicted to rise, resulting in a decrease in resistance. This PEDOT: PSS based temperature sensor showed a 70% change in resistance for a temperature change of $\sim 60^\circ C$. Jacopo et al. [79] developed a dual-mode highly flexible hydrogel-based H_2O_2 sensor, which is based on poly(3,4-ethylenedioxythiophene): polystyrene sulfonate (PEDOT: PSS) as the transducer, hydrophilic polyurethane (HPU) as the hydrogel matrix, and horseradish peroxidase (HRP) as the H_2O_2 specific redox enzyme. The solution containing PEDOT: PSS/HPU/HRP was applied onto the work electrode of a three-electrode system using a drop-casting method. The sensor operated in both amperometric and chemiresistive dual modes. The sensing mechanism can be explained as follows. In the amperometric mode, H_2O_2 undergoes electrochemical reduction in the presence of HRP to produce water and oxygen on the surface of the modified electrode [175]. The process entails the alteration of the oxidation state of the iron ion within the heme group present in HRP, resulting in the discharge of electrons that are subsequently transmitted to the electrode, thereby inducing a current peak [176,177]. Conductive PEDOT: PSS plays a key role in the direct electron transfer between the enzyme and the electrode [178]. In the chemiresistive mode, the sensing mechanism relies on the conformational change of PEDOT between two molecular structures, quinoid and benzoid, in response to H_2O_2 . The quinoid group, which has alternating double bonds and more charges, is more conductive form leading to an increase in PEDOT: PSS conductivity. The change to the fibrillary-like structure of quinoid increases the delocalization of charges and π -electrons in the PEDOT backbone, resulting in higher conductivity. The conformational change to the coil-like benzoid structure reduces elec-

tron mobility [126,179]. The HRP and H_2O_2 reaction initiates the PEDOT conformational change to the quinoid structure, resulting in higher conductivity [178]. The amperometric sensor showed a rapid response time of less than 6 seconds, selectivity towards common interferents, and a broad detection range spanning from 100 μM to 101.6 mM.

Polypyrrole (PPy): PPy can be traced back to the 1919 studies by Angeli and Pieroni, who described the synthesis of pyrrole blacks from pyrrole magnesium bromide [180]. PPy is unique among the electronically conductive polymers in that the pristine form is oxidized by molecular oxygen [181]. PPy exhibits impressive conductivity, outstanding redox properties, biocompatibility, tunability and processability, as well as environmental stability, and could be widely applied in the field of chemical sensing [182,183,184]. Conductive polymer aerogels are considered a promising option to create versatile electronics due to their mechanical deformability and their 3D porous structures [185]. Compared to conventional porous materials, conductive polymer aerogels possess advantages such as regulated conductivity, low density, large specific surface areas, and interconnected channels, making them suitable for gas sensing applications [83]. Shu et al. [83] developed a paper substrate-based flexible H_2S sensor by incorporating CuxO-polypyrrole conductive aerogel (CuxO-PPy) into the sensor design. The aerogel is prepared by in situ polymerization of pyrrole with a “big-macromolecular surfactant” of graphene oxide (GO). GO is composed of both aromatic regions consisting of benzene rings and aliphatic regions consisting of oxygen-containing groups, which give it amphiphilic properties [186]. The surface of graphene oxide was improved by the self-assembly of positively charged pyrrole monomers through π - π stacking and electrostatic interactions, resulting in a better pyrrole dispersion. Additionally, the 3D porous structure of the coated aerogel was maintained on the paper substrate, and due to the combined mechanical properties of the aerogel and paper, the resulting paper electrode was mechanically flexible and could be easily cut into desired shapes. The sensing mechanism of this chemiresistive gas sensor can be speculated as follows. First, when the sensor was exposed to H_2S , some of the absorbed gas molecules dissociated into H^+ and S_2^- (HS^-). Secondly, the S_2^- readily reacted with semiconducting CuxO, transforming it into metallic CuxS with good conductivity [187,188]. This process facilitated the continuous dissociation of H_2S molecules, generating more H^+ ions. Thirdly, the dissociated H^+ ions protonated PPy, serving as the host conducting pathway, and facilitated electron transfer through multidimensional paths, including 2D nanosheets and 3D cross-linking frameworks, ultimately improving conduction [189]. The CuxO promoted the protonation of PPy. Fourthly, the aerogel's inherently hierarchical and porous micro-nanostructures of CuxO-PPy@GO, with a large surface area and pore volume, provided abundant reaction sites (such as vacancies, defects, functional groups, and sp^2 -bonded carbon) for gas molecule adsorption [190]. Moreover, the loose and porous structure facilitated rapid gas diffusion and worked simultaneously on both the internal and external surfaces. Overall, the exceptional sensing performance was attributed to the synergistic function of the micro-nanostructures, the strong chemical interaction between CuxO and H_2S , and the effective proton acid doping with PPy.

3.3. Nanohybrid materials

Nanohybrid materials are distinct combinations of inorganic and organic materials [191]. Compared to single-component materials, hybrid materials possess a diverse range of functionalities as well as enhanced chemical and physical properties. Advanced nanostructures that are based on organic/inorganic composites play a significant role in driving innovations across various fields [192,193]. Hybrid materials exhibit enhanced properties such as higher conductivity, increased porosity, improved catalytic activity, and greater optical and electrical potential compared to their single-component counterparts [194,195]. When polymeric materials are combined with metal oxides or other inorganic materials, the resulting composite can exhibit enhanced selectivity and unique sensing properties. This is due to the synergistic and geometrical effects of the different components [196,197]. Polymer materials and 2D graphene-based materials have been shown to have a large surface area and good electrical conductivity, but the performance of these devices can be further improved by synthesizing nanohybrid materials.

BP-Ti3C2-MXene: Zhu Xiaoyu et al. [115] designed an ultra-trace analysis phytoregulator α -naphthalene acetic acid (NAA) sensor fabricated by two-dimensional phosphorene (BP) nanohybrid with graphene-like titanium carbide MXene (Ti3C2-MXene). MXene, 2D material, 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) on the flexible substrate surface of laser-induced porous graphene (LIPG). A BP-Ti3C2-MXene nanohybrid with excellent ambient stability is produced through liquid-phase exfoliation of black phospho-

rus with cuprous chloride and Ti3C2-MXene, which is obtained by etching Al layers of Ti3AlC2, using ultrasonic assistance in an organic solvent. MXene has a unique thin-layered nanostructure that provides ample space for supporting other functionalized nanomaterials. However, the performance of MXene is compromised when its nanosheets restack too severely, which can damage the effective area [198]. To address this issue, a noncovalent nanohybrid between BP and MXene is expected to resolve the aggregation of MXene or BP and combine the advantages of both materials [199]. The sensing mechanism of this electrochemical sensor is as follows. In the amperometric mode, the working electrode modified with Ti3C2-MXene/BP biomimetic enzymes exhibits oxidase-like characteristics (nanozyme) when the zymolyte NAA is electrocatalytically oxidized. Electron transfer reactions generate measurable changes in current. Since the size of the measured current is proportional to the number of NAA molecules in the solution, the relative concentration of the molecules can be monitored on a physiological timeline [200]. The LOD of the sensor is as low as 1.6 nM with a wide linear range of 0.02–40 μM .

4. Principle of flexible sensing

The sensing layer of a flexible sensor interacts with physical disturbances or analytes, causing changes in its own physical properties, which are then transduced into variations of electrical signals (e. g., current, voltage/ Nernst potential, capacitance, etc.) or others by the transducer. In this section, we will discuss the sensing principle of electrically-transduced analytical flexible sensors from the aspects of sensing and transduction mechanisms.

4.1. Sensing mechanism

A typical sensing device consists of two primary elements: sensing material and the transducer [201]. The sensing material is accountable for responding to physical perturbation (e. g., temperature, humidity, pressure, mechanical deformation, etc.) or chemical/biological analytes (e. g., gases, pesticides, foodborne pathogens, etc.). This interaction results in a modification of one or more properties of the sensing material, which is subsequently transformed into detectable signals by the transducer [202–204]. Sensors capable of detecting physical changes have been achieved through the collaborative advancements in material development, involving the synthesis of materials with novel electrical, optical, and mechanical properties and refinement in the methods of integrating materials into devices [205]. In contrast, to advance electrically-transduced chemical sensing, there is an added level of complexity posed by the chemical interfaces between the sensing material and the analyte [201]. The chemical interfaces between the material and the analyte are crucial in determining the sensitivity, selectivity, stability, and biocompatibility of chemical sensing devices [206–208]. The interaction between sensing materials and analytes is a prerequisite for the operation of sensors.

We can classify these interactions as:

1. Non-covalent interactions, including van der Waals forces, hydrogen bonds, coordination bonds, and π - π interactions.

This may result in reversible or partially reversible reactions. For example, the large, electron-rich π -surface of graphene can interact with target analytes through van der Waals forces, charge transfer, and π - π interactions.

2. Covalent bonding.

This leads to irreversible reactions but brings improvements in selectivity and sensitivity. For example, metal oxides contain chemically adsorbed oxygen molecules on their surface, which are responsible for the interaction with gaseous analytes through oxygen-involved chemical reactions [64].

4.2. Transduction mechanism

The sensing material serves a dual purpose in chemical sensing. Firstly, it should have the ability to covalently or noncovalently interact with the analyte on its surface. Secondly, it should react to this interaction by altering its electrically related physical properties. The transduction mechanism relies on conductivity, work function, or electrical permittivity, which can be transformed into a change in resistance, capacitance, or inductance [203]. These transduction events, which involve changes in resistance/impedance, capacitance, current, and voltage/electrical potential, can be detected and measured using various devices such as resistors, electrochemical sensors, capacitors, diodes, and field-effect transistors. The magnitude, frequency, and phase of the resulting signal can provide important information about the sensing event [64]. In flexible sensing for food monitoring, the main transducers are resistors and electrochemical sensors, while capacitors are uncommon. Diodes and field-effect transistors are rarely seen. The modulation of doping level, Schottky barrier, and the formation of dipole and interfacial layer are the basic mechanisms that typically lead to changes in conductivity, work function, and permittivity [64].

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