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Flexible Sensors for Hydrogen Peroxide Detection: A Critical Review

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diagnostic purposes. This review provides an insight about different types of sensors that have been developed for detection of H₂O₂. Their flexibility, stability, cost, detection limit, manufacturing, and challenges in their applications have been compared. More specifically the advantages and disadvantages of various flexible substrates that have been utilized for the design of H₂O₂ sensors were discussed. These substrates include carbonaceous substrates (e.g., reduced graphene oxide films, carbon cloth,



carbon, and graphene fibers), polymeric substrates, paper, thin glass, and silicon wafers. Many of these substrates are often decorated with nanostructures composed of Pt, Au, Ag, MnO₂, Fe₃O₄, or a conductive polymer to enhance the performance of sensors. The impact of these nanostructures on the sensing performance of resulting flexible H2O2 sensors has been reviewed in detail. In summary, the detection limits of these sensors are within the range of 100 nM-1 mM, which makes them potentially, but not necessarily, suitable for applications in health, food, and environmental monitoring. However, the required sample volume, cost, ease of manufacturing, and stability are often neglected compared to other detection parameters, which hinders sensors' real-world application. Future perspectives on how to address some of the substrate limitations and examples of application-driven sensors are also discussed.

KEYWORDS: flexible sensor, hydrogen peroxide, substrate, nanomaterials, sensing performance, health, environment, food

1. INTRODUCTION

Hydrogen peroxide, H₂O₂, is an inorganic volatile compound with strong oxidizing properties. It is often used as a chemical in bleaching solutions, water treatment, and chemical synthesis. It is also an important biological molecule, taking part in host defense, oxidative biosynthetic reactions, metabolism, oxidative stress, and signaling.^{1,2} Being such an important molecule in both industrial and biological processes, H₂O₂ can be found in the environment, human body fluids, and food. The level of hydrogen peroxide in surface water, such as seas,^{3,4} rain,⁵ snow,^{6,7} rivers,^{8,9} and lakes,¹⁰ determines the state of *flora*, fauna, and air and water quality. In breath and body fluids such as urine and blood plasma, hydrogen peroxide concentration can indicate metabolic disorders associated with diabetes, pulmonary diseases, or other health conditions.¹¹⁻¹⁶ In food and beverages, H₂O₂ residues may be found in the final products after processes such as pasteurization, sterilization, and packaging. However, according to food safety regulation reported by Food Standards Australia and New Zealand, the concentration of H_2O_2 should be below 147 μM in the final products.¹⁷⁻²⁰ In fact, H₂O₂ concentration above specific

thresholds is considered hazardous for human consumption, especially if ingested or inhaled.^{21,22} Thereby, ensuring that the H₂O₂ level is within the safe range is crucial for health, safety, and the environment. As shown in Figure 1, the safe concentration range within which hydrogen peroxide remains benign for biological entities depends on the media and its uptake pathway, spanning from ~ 100 nM for seawater to ~ 1 mM for the aquaculture industry.

Given the diverse role of hydrogen peroxide in industrial and biological processes, H₂O₂ sensors have been at the center of thriving research in recent years.^{23,24} Compared with traditional analytical techniques such as high-performance liquid chromatography (HPLC), which are currently used for most

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Figure 1. Concentration range of H_2O_2 in different media, including the environment, health, and food. The black bars refer to safe ranges (i.e., any value inside this range is considered to be a normal or safe value, while any value outside this range is considered abnormal and unsafe), while the red bars represent the hazardous ranges (i.e., any value inside these ranges is considered to be characteristic of a condition or a disease–note, for example, the differences between the ranges of exhaled air (EA) (black bar) and EA affected by different conditions (red bars)). Levels for diabetes refer to concentration in blood. COPD stands for "Chronic Obstructive Pulmonary Disease". MPL stands for "Maximum Permitted Level". For MPL and wine treatment, the maximum permitted level is indicated. The numbers in the brackets alongside the *y*-axis indicate the corresponding references.

sample analyses in the medical, environmental, and food safety fields, the use of H_2O_2 sensors is favorable, as they are portable, cost-effective, and less time-consuming and can be used by nonspecialized technicians for point-of-care detection and continuous monitoring. Since hydrogen peroxide is a highly active substance, its preferred detection mechanism is through the reaction of H₂O₂ molecules with a substrate on the sensor's surface. In fact, hydrogen peroxide has a strong redox activity, which can be used for its detection via formation or degradation of a dye (colorimetric) or oxidizing a mediator molecule (electrochemical). Although different transduction elements have been employed for detection of hydrogen peroxide, electrochemical sensors, compared with colorimetric sensors, are more favored because of their higher sensitivity and selectivity, cost-effectiveness, relatively shorter response time, and miniaturization capabilities.²³ Among electrochemical sensors, amperometric sensors are less complex and easy to implement. The amperometric sensors are also highly selective as the target molecular compounds (analytes) have specific redox peaks depending on their chemical structure. By selecting the specific redox peak of an analyte as the applied potential, the amperometric sensor can selectively target that specific analyte. The characteristic anodic peak of hydrogen peroxide in a phosphate buffer solution (PBS) measured via cyclic voltammetry can vary from -1.2 V, measured with a cadmium oxide nanoparticle (NP) modified electrode,²⁵ to 0.98 V, measured with a gold NP modified indium tin oxide electrode.^{2,26} Other common electrochemical H₂O₂ sensors are potentiometric,²⁷ chemiresistive, and impedimetric,^{28,29} while colorimetric sensors can include chemiluminescent²⁴ and photoluminescent (which includes fluorescent and phosphorescent sensors). Although colorimetric sensors can achieve detection limits as low as 500 nM, as in the case of Senthamizhan et al. sensor,³⁰ they are not suitable for in situ

quantitative measurement. Colorimetric sensors require the accompaniment of another instrument such as a spectrophotometer, for quantitative detection. Therefore, they are not desirable for the POC applications where the detection of the exact amount of the analyte is critical. Recently, smartphones have been tested as portable devices for semiquantitative detection in conjunction with a colorimetric sensor emitting light within the visible range.³¹ Nevertheless, such semiquantitative measurements lack accuracy compared to quantitative analysis of electrochemical sensors. Moreover, photoluminescent sensors require an external light source, often UV, which may not be practical for food packaging and healthcare POC detection.

Commercial H_2O_2 sensors mostly consist of a metal or plastic-metal composite probe. The probe is connected to a signal reader, which requires an external electrical source and usually relies on an optical or electrochemical detection mechanism.^{32,33} Applications of commercial H_2O_2 sensors range from water quality assessment, such as potable water or a swimming pool, to wastewater treatment, and to vapor sterilization process monitoring.³⁴ Sensitivity of commercial H_2O_2 sensors can be as low as in the order of parts per million. Moreover, they are designed for continuous monitoring and point-of-care detection and hence generally need to be durable, stable, and able to tolerate an intensive water flow rate and corrosive conditions. Therefore, they can work in different conditions of temperature, pH, and water purity, but this also leads to bulky and rigid sensors which usually demand high sample volumes. As such, current commercially available H_2O_2 sensors are not suitable for healthcare and food application.

Sensors' performance is often assessed by their detection range, sensitivity, limit of detection (LOD), selectivity, and stability. However, for health, environment, and food point-ofcare applications, additional factors such as cost, toxicity, biocompatibility, low sample volume needed for analysis, ease of manufacturing, and scalability are equally important factors to be considered. Moreover, for the implementation of a sensor for applications in food packaging, wearable devices, and medical devices, another imperative parameter is mechanical flexibility.^{35,36}

Herein, we provide an insight for the recent progress in the development of flexible sensors for the detection of hydrogen peroxide. Their performance is reviewed through assessing their sensing parameters, ease of fabrication, scalability, and their potential for real-world applications in monitoring environmental, health, and food safety conditions. One of the main components of sensors which are pivotal for their scalability, cost, and fabrication, yet mostly overlooked, is the substrate. Therefore, in this review, sensors have been categorized by their substrate materials, as each material is characterized by specific properties that drive the sensor's design and manufacturing. At the end, future perspectives for design and manufacturing of flexible hydrogen peroxide sensors were provided.

2. HYDROGEN PEROXIDE SENSORS AND MECHANISMS OF DETECTION

Hydrogen peroxide is a highly active molecule because of the unstable nature of peroxide bonds. As such, H_2O_2 degradation usually passes through an intermediate step with the formation of two hydroxyl ions or an ion and a radical. Consequently, compounds that are either charged or partially charged are formed. These decomposition routes are utilized in many



Figure 2. A) Illustration of the possible detection mechanism of H_2O_2 on Au–Ag NPS.⁵³ Notice how, being noble metals, hydrogen peroxide is just adsorbed on the substrate and then reacts with hydrogen ions provided by the environment. Reproduced with permission from ref 53. Copyright 2019 Elsevier. B) Proposed reaction mechanism of H_2O_2 on MnOOH NRs.⁴³ Different from noble metal-based sensors, here hydrogen peroxide gets adsorbed on the active material, from which it takes one hydrogen atom, changing the material structure. C) Proposed H_2O_2 detection mechanism for the Dutta et al. fluorescent sensor.³¹ In the colorimetric sensor, hydrogen peroxide has to react with the dye or the luminescence enhancer. Reproduced with permission from ref 31. Copyright 2022 American Chemical Society. D) H_2O_2 detection mechanism schematic. The red and white spheres are oxygen and hydrogen, respectively. Note the two different final steps depending on whether hydrogen or hydroxyl ion is provided.

electrochemical, particularly amperometric, sensing mechanisms.^{2,37} The generic hydrogen peroxide decomposition process can be summarized as in eq 1, where *a*, *b*, *c*, and *d* are the stoichiometric values. The presence of hydrogen and formation of oxygen in the reaction depend entirely on the nature of the reaction, which is driven by the nature of materials used in the sensor probe, as well as the pH of media where the reaction occurs (i.e., acidic or basic). It should be noted that when oxygen is formed, hydrogen is absent from the reaction and vice versa.

$$aH_2O_2 + bH_2 \rightarrow cH_2O + dO_2 \tag{1}$$

Commonly, the degradation of hydrogen peroxide is catalyzed by the active sites on the surface of sensors, where the OH[•] radicals are generated. Here, the radicals gain electrons from the active material, returning to be hydroxyl anions and desorbed as water molecules. 38 This is a common detection system in electrochemical sensors based on noble metals (Figure 2A).^{29,39,40} Alternatively, the hydroxyl ions and radicals originating from hydrogen peroxide can chemically react with the active materials in the sensor such as transition metal-based structures (e.g., iron and manganese).⁴¹⁻⁴³ Contrarily to the sensing mechanism by noble metals, hydroxyl ions are not only adsorbed but react with the active sites of the transition metalbased structures before being released, usually taking with them hydrogen atoms (Figure 2B). Both of these detection dynamics revolve around the reduction-oxidation processes that happen when H_2O_2 dissociation is catalyzed by the sensor. The adsorption-desorption of the hydroxyl groups involves electron transfer, which can be detected in amperometric and voltametric sensors. The same principle can be used for nonelectrochemical sensors,^{31,44} or where an intermediate is used to catalyze the hydrogen peroxide reaction, such as in the case of Prussian blue,⁴⁵ or enzymes (Figure 2C).⁴⁶ On the other hand, the detection mechanism in chemiresistive sensors depends on the nature of the conductive material, as the interaction with hydrogen peroxide results in a change in conductivity.⁴⁷ Nevertheless, the reaction mechanism is similar in both amperometric and chemiresistive sensors.³⁸ The first two steps of the hydrogen peroxide decomposition reaction in

electrochemical sensors are shown in eq 2 and eq 3, where a radical hydroxyl group is generated. The final steps of the reaction are pH- and potential-sensitive as shown in eqs 4a and 4b, respectively.² The schematic of the whole hydrogen peroxide decomposition process in the presence of an active material in a sensor is also presented in Figure 2D.

$$H_2O_2 + e^- \to OH + OH^- \tag{2}$$

$$\dot{O}H + e^- \rightarrow OH^-$$
 (3)

$$2OH^- + 2H^+ \to 2H_2O \tag{4a}$$

$$4OH^- \rightarrow 2H_2O + O_2 + 4e^- \tag{4b}$$

This generic mechanism governs almost all electrochemical sensors. The specific dynamic of detection, however, relies on the active material and the environment, while it is independent from the substrate material used. The latter drives the mechanical properties of the sensor and the decoration and modification techniques that can be used during manufacturing. The main substrate materials used are carbonaceous materials, polymers, and paper.

3. CARBONACEOUS SUBSTRATES

Substrates made from carbonaceous materials such as carbon, graphite, graphene, graphene oxide (GO), and reduced graphene oxide (rGO) are flexible, highly conductive, stable, tough, and biocompatible and usually have a high surface area.⁴³ As such, they are suited for use in electrochemical sensors and in electrode manufacturing where miniaturization and mechanical flexibility must be achieved. Being conductive, their surface modification usually revolves around electrochemical techniques, such as electrodeposition, but other techniques such as chemical modification and hydrothermal processes are also viable. Figure 2A illustrates a schematic summarizing a sensor developed from carbonaceous substrates. Based on the production procedure, such substrates can be produced in different flexible structures, ranging from microfibers to expanded sheets. It is important to note that, although a large number of electrochemical and colorimetric sensors rely on the use of graphene for detection, thanks to its

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	$\begin{array}{llllllllllllllllllllllllllllllllllll$	1.62	378.1	N.A.	OA (2), paracetamol	Ş	N.A.	55
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Ag WVs arprenentcic $200-1.5 \times 10^3$ 46 749 30 accorbic acid, glucose, sodium oralate 2 001 39 DNN W/s + Ag NPs dremiseitite 1.7×10^3 4.1 N.A N.A 3.4×10^3 $3.4 \times $	Ag NWsamperometric $200-1.5 \times 10^3$ Ag NWsamperometric 1.7×10^3 PANI NWs + Ag NPschemiresistive 1.7×10^3 Cu MOFamperometric $10-1 \times 10^3$ Cu MOFamperometric $10-1 \times 10^3$ LIG + Pt NPsamperometric $10-1 \times 10^3$ LIG + Pt NPsamperometric $10-29 \times 10^3$ LIGamperometric $10-29 \times 10^3$ Ag porous µfilmamperometric $10-29 \times 10^3$ MnS@MoS2amperometric $2-500$ AgarMnS@MoS2amperometric $2-500$ MnS@MoS2amperometric $0.5-5 \times 10^3$ PEDOT-PSS-HRPchemiresistive $61.3 \times 10^{-3}-61.3$ $0.5-5 \times 10^3$ MnS@MoS2amperometric $10-10 \times 10^3$ MnS ODcolorimetric $10-10 \times 10^3$ $0.5-5 \times 10^3$	0.485	21.93 ^c	21	sugars (2), urea, ascorbic acid, ethanol	<1	S	29 ^d
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	PANI NWs + Ag NPs chemiresistive 1.7×10^3 - 3.4×10^3 Cu MOF amperometric $1 \times 10^3 - 20 \times 10^3$ Cu MOF amperometric $10^{-1} \times 10^3$ - 6.8×10^3 LIG + Pt NPs amperometric $10^{-2} \times 10^3$ LIG amperometric $10^{-2} \times 10^3$ Ag porous μ film amperometric 1^{-10} Ag porous μ film amperometric 1^{-10} Ag porous μ film amperometric 2^{-500} MA. α MnS(β MoS ₂ amperometric $0.5^{-5} \times 10^3$ PEDOT-PSS-HRP chemiresistive $61.3 \times 10^{-3} - 61.3$ (chitosan + TMB colorimetric $10^{-1} 0 \times 10^3$	46	749	30	ascorbic acid, glucose, sodium oxalate	\$	0.01	39
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	PANI NWs + Ag NPschemiresistive $1 \times 10^3 - 20 \times 10^3$ Cu MOFamperometric $10-1 \times 10^3$ Cu MOFamperometric $1.4 \times 10^3 - 6.8 \times 10^3$ LIG + Pt NPsamperometric $10-29 \times 10^3$ LIGamperometric $10-29 \times 10^3$ Ag porous μ filmamperometric $1-10$ Ag porous μ filmamperometric $2-500$ AgarMnS@MoS2amperometric $2-500$ MnS@MoS2amperometric $0.5-5 \times 10^3$ PEDOT-PSS-HRPchemiresistive $61.3 \times 10^{-3}-61.3$ $0.5-5 \times 10^3$ MAC CDcolorimetric $10-10 \times 10^3$ 0.500		1640					
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	Cu MOF amperometric $10-1 \times 10^3$ 1.4×10^3 - 6.8×10^3 LIG + Pt NPs amperometric $1-29 \times 10^3$ 8.8×10^3 LIG amperometric $1-29 \times 10^3$ Ag porous µfilm amperometric $1-10$ Ag porous µfilm amperometric $2-500$ Agar MnS@MoS_1 amperometric $2-500$ MnS@MoS_2 amperometric $0.5-5 \times 10^3$ PEDOT-PSS-HRP chemiresistive $61.3 \times 10^{-3}-61.3$ (chitosan + TMB colorimetric $10-10 \times 10^3$ MAD CP AD CON AD CON A MAD CP AD AD CON AD CO	1	N.A.	N.A.	N.A.	<180	N.A.	38
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	$\label{eq:relation} \begin{array}{c} 1.4 \times 10^3 - 6.8 \times 10^3 \\ 10-29 \times 10^3 \\ 10-10 \\ 10-10 \\ 10-10 \\ 10^3 \\ $	4.1	N.A.	N.A.	ascorbic acid, 1-histidine	<60	N.A.	57
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	LIG + Pt NPs amperometric $10-29 \times 10^3$ LIG $1-10$ amperometric $1-10$ Ag porous μ film amperometric 110 Agar α amperometric $0.5-5 \times 10^3$ MnS@MoS ₂ amperometric $0.5-5 \times 10^3$ PEDOT-PSS-HRP chemicesistive $61.3 \times 10^{-3}-61.3$ (chitosan + TMB colorimetric $10-10 \times 10^3$ MAC CD 0.0000							
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Ag porous μ filmamperometricN.A.N.A.S37.4N.A.glucose, OA (2), ethanolN.A.A.A.40M.A.Pl, andeGR + PkNPsamperometric2–5001.9164.3N.A.N.A. $\sim (10^{\circ} 30^{\circ} 60^{\circ} 60^{\circ})$ 3060/AgarMnS@MoS_3amperometric2–5001.9164.3N.A.N.A. $\sim (10^{\circ} 30^{\circ} 60^{\circ} 60^{\circ})$ 3060/AgarMnS@MoS_3amperometric0.5–5 × 10^30.1265060sugars (4), metal ions (3), OA (2), amines (2), $\sim (5^{\circ} NA)$ 47PEDOT-PSS-HRPchemiresistive61.3 × 10 ⁻³ - 61.30.0613N.A.7N.A.7N.A.42chitosan + TMBcolorimetric10–10 × 10^31.55N.A.N.A.N.A.N.A.4247chitosan + TMBcolorimetric10–10 × 10^31.55N.A.N.A.N.A.1443chitosan + TMBcolorimetric10–10 × 10^31.55N.A.N.A.N.A.4740mhO_{T}CPpotentiometric110–10 × 10^31.55N.A.N.A.4740cholate hydrogelamperometric110-5021778.85N.A.N.A.4747cholate hydrogelamperometric9.1–16.5 × 10^37.70.178N.A.N.A.4746056027cholate hydrogelamperometric9.1–16.5 × 10^37.70.074N.A.N.A.47 <td< td=""><td>Ag porous µfilm amperometric N.A. PI, and eGR + PtNPs amperometric 2–500 /Agar MnS@MoS₂ amperometric 0.5–5 × 10³ PEDOT-PSS-HRP chemicesistive 61.3 × 10⁻³–61.3 (chitosan + TMB colorimetric 10–10 × 10³ MAO CP 200000000000000000000000000000000000</td><td>0.3</td><td>20^{c}</td><td>N.A.</td><td>N.A.</td><td>N.A.</td><td>S</td><td>59</td></td<>	Ag porous µfilm amperometric N.A. PI, and eGR + PtNPs amperometric 2–500 /Agar MnS@MoS ₂ amperometric 0.5–5 × 10 ³ PEDOT-PSS-HRP chemicesistive 61.3 × 10 ⁻³ –61.3 (chitosan + TMB colorimetric 10–10 × 10 ³ MAO CP 200000000000000000000000000000000000	0.3	20^{c}	N.A.	N.A.	N.A.	S	59
η_{and} eGR + PtNPs amperometric 2-500 1.91 64.3 N.A. N.A.	IJ, and eGR + PtNPs amperometric 2–500 /Agar MnS@MoS2 amperometric 0.5–5 × 10 ³ PEDOT-PSS-HRP chemicesistive 61.3 × 10 ⁻³ –61.3 0 ohitosan + TMB colorimetric 10–10 × 10 ³ 0 M.O. Ch non-non-non-non-non-non-non-non-non-non	N.A.	537.4	N.A.	glucose, OA (2), ethanol	N.A.	N.A.	40
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	$MnS(@MoS_2 mtext{ amperometric } 0.5-5 \times 10^3$ $PEDOT-PSS-HRP mtext{ chemicesistive } 61.3 \times 10^{-3}-61.3 mtext{ chitosan + TMB } colorimetric mtext{ 10-10 \times 10^3 } 0.000 mtext{ MoO Cm } 0.000 mtext{ mod + 100 } 1.000 mtext{ mod + 100 \ mtext{ mo$	1.91	64.3	N.A.	N.A.	<10	30	60
PEDCT-PSS-HRP chemiresistive $61.3 \times 10^{-3} - 61.3$ 0.0613 N.A. 7 N.A. < 3600 0.02 47 $^{-7}$ $^{-7}$ N.A. $^{-7}$ $^{-3}600$ 0.02 47 $^{-7}$ $^{-7}$ $^{-7}$ $^{-3}600$ 0.02 47 $^{-7}$ $^{-7}$ $^{-3}600$ 0.02 47 $^{-7}$ $^{-7}$ $^{-7}$ $^{-3}600$ 0.1 61 $^{-61}$ $^{-10-10} \times 10^3$ 1.55 $^{-7}$ $^{-8}$ $^{-3}$ $^{-3}600$ 0.1 61 $^{-7}$ $^{-7}$ $^{-7}$ $^{-7}$ $^{-6}00$ $^{-1}$ $^{-1}$ $^{-1}$ $^{-1}$ $^{-1}$ $^{-1}$ $^{-1}$ $^{-1}$ $^{-1}$ $^{-1}$ $^{-1}$ $^{-1}$ $^{-1}$ $^{-7}$ $^{-1}$ $^{-1}$ $^{-1}$ $^{-1}$ $^{-1}$ $^{-1}$ $^{-1}$ $^{-1}$ $^{-1}$ $^{-1}$ $^{-1}$ $^{-1}$ $^{-1}$ $^{-1}$ $^{-1}$ $^{-1}$ $^{-1}$ $^{-1}$ $^{-1}$	PEDOT-PSS-HRP chemicesistive $61.3 \times 10^{-3}-61.3 \text{ 0}$ chitosan + TMB colorimetric $10-10 \times 10^3$ M.O. Ch	0.12	650	60	sugars (4), metal ions (3), OA (2), amines (2), glutathione, amino acids (4)	\$	N.A.	42
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	chitosan + TMB colorimetric $10-10 \times 10^3$	0.0613	N.A.	7	N.A.	<3600	0.02	47
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	$M_{n} \cap \Omega \qquad \qquad$	1.55	N.A.	N.A.	N.A.	<3600	0.1	61
$ \begin{array}{ccccccc} 2-\text{modified Tb}^{3+} & \text{fluorescent}^h & 10-50 & 2 & 1778.85 & \text{N.A. water, salts (S), amino acids (2), tert-buryl & 1 h & \text{N.A. } & 31 \\ \text{cholate hydrogel} & & \text{hydroperoxide, HO} \bullet & \text{hydroperoxide, HO} \bullet & & & & & & & & & & & & & & & & & & $	1×10^{-1} potentionitetity 1×10^{-1} 1×10^{-1}	4×10^{-5}	26.98 ^g	N.A.	N.A.	12	0.05	27
	2-modified Tb ³⁺ fluorescent ^h 10–50 cholate hydrogel	2	1778.85	N.A.	water, salts (5), amino acids (2), <i>tert</i> -butyl hydroperoxide, HO^{\bullet}	1 h	N.A.	31
7.7–42.6 × 10 ³ 7.7 0.074 <a>360 r PB - nanoporous Au amperometric 1–17 × 10³ 0.22 708 60 min sugars (3), OA (3) <4 N.A. 41 film	t glass carbon graphite ink amperometric $9.1-16.5 \times 10^3$	9.1	0.168	N.A.	N.A.	<600	≈0.08	62 ^f
er PB - nanoporous Au amperometric 1–17 × 10 ³ 0.22 708 60 min sugars (3), OA (3) <4 N.A. 41 film	$7.7 - 42.6 \times 10^3$	7.7	0.074			<360		
	er PB - nanoporous Au amperometric $1-17 \times 10^3$ film	0.22	708	60 min	sugars (3), OA (3)	4>	N.A.	41

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not weighted on the electrode area. Hence, sensitivity is shown as $\mu A m M^{-1}$ and not $\mu A cm^{-2} m M^{-1}$. ^{*d*}Data for ref 29 refers to the amperometric mode. ^{*e*}Selectivity is tested only when detecting glucose with the addition of glucose oxidase. ^{*f*}The two data sets refer to the two-electrode and three-electrode setup, respectively, ^{*g*}Sensitivity is in mV log(M)⁻¹. ^{*h*}Detection range and LOD refers to the RGB mode of detection. Sensitivity is in directed and the environmetric over concentration of H₂O₂ an μM^{-1} .

Review

biocompatibility, high conductivity, and toughness, the vast majority of these sensors is not free-standing and requires the deposition on glassy carbon electrodes or similar substrates.^{48,49} Hence, even though research in graphene-based sensors has progressed greatly in recent years, it is not considered as a substrate and only used as a functionalization material. Contrarily, reduced GO paper, carbon cloth, and carbon fibers can be considered flexible free-standing substrates. The sensing parameters of flexible H_2O_2 sensors are summarized in Table 1.

3.1. Reduced Graphene Oxide Paper. Freestanding reduced graphene oxide (rGO) paper is produced by dispersing GO in a solvent, usually deionized water, followed by deposition in layers on a surface. Different techniques, such as spin coating, vacuum filtration, 46,50,63 layer-by-layer assembly,⁵² and mold-casting,⁵¹ have been used for the fabrication of rGO papers. The formed GO layers are then reduced, chemically or electrochemically, and peeled off to obtain freestanding rGO paper (Figure 2B). The film thickness can be tuned by changing the solution concentration and volume.⁵⁰ rGO paper shows improved mechanical properties compared with graphite, while being highly conductive, tough, and uniform and exhibiting a high surface to volume ratio.⁵ The cost of manufacturing is relatively low when using techniques such as vacuum filtration, but processing might be time-consuming depending on its surface functionalization.⁵² rGO alone proved to be weakly responsive to H_2O_2 as the related oxidation and reduction peaks expected were not observed. Therefore, rGO is commonly functionalized with nanostructures or other compounds to make it sensitive enough to allow detection.⁵¹ As an example, Song et al. electrodeposited Pt NPs on rGO paper to improve H2O2 detection in a three-electrode system.⁵⁰ This amperometric sensor exhibits two linear detection ranges from 0.2 μ M to 2 mM and from 2 mM to 8.5 mM with an LOD of 100 nM. Moreover, the sensor proved to retain 92% of its sensitivity after bending and was able to detect H₂O₂ in milk. Similarly, Fe₃O₄ nanocrystals (NCs) can be electrochemically deposited on rGO.⁴⁶ Here, the modified electrode was decorated with catalase, an oxidoreductase enzyme that catalyzes hydrogen peroxide decomposition, to selectively react and detect it.⁴⁶ However, the electrode modification involves multiple steps, of which the catalase enzyme immobilization takes up to 8 h with an efficiency as low as 33%. To optimize the sensor production step, Zhang et al. decorated a layer-by-layer assembly of rGO papers with Au NPs coated in PB, which were added directly to the GO solution before the filtration process.⁵² The sensor developed by this method showed a linear detection range between 1 μ M and 30 μ M, which might not be broad enough for many applications, and its selectivity and stability were not reported. On a similar note, rGO can be decorated with other active compounds while being reduced. In Dong et al. work,⁵¹ the MnO₂ precursor was added to the GO solution where the electrochemical reduction was occurring. The presence of the MnO₂ precursor allowed a one-step electroreduction-deposition. Such sensors could detect H2O2 in a biological cell environment in the range of 0.1 mM to 45.4. mM.⁵

3.2. Carbon Cloth. Carbon cloth consists of carbon fiber threads woven together to form a fabric. As such, while showing the chemical and electrical characteristics of carbon-based materials, it also shows larger surface area, high porosity, relatively low cost, and mechanical robustness.⁴³ Carbon cloth's performance on its own toward the detection of H_2O_2 is

poor. Hence, its surface needs to be modified with electrocatalytic materials to improve sensitivity and selectivity. In Gowthaman et al. work,⁵³ carbon cloth was immersed in a solution containing Ag and Au precursors in the presence of ascorbic acid. By an electroless deposition, Ag NPs were initially formed and attached to the cloth, and then Au-Ag NPs were produced. Through these simple production steps, a sensor was produced that could detect H₂O₂ in the range between 500 nM and 2 mM (Figure 2C). Moreover, the sensor's performance was tested in human urine and blood serum, where it was able to detect 10 μ M hydrogen peroxide. Analogously, MnOOH nanorod (NR) arrays can be directly grown on carbon cloth by hydrothermal autoclaving. Although time-consuming, this process was quite straightforward, and the fabricated sensor displayed a linear detection range from 20 μ M to 9670 μ M and retained its sensitivity for up to 30 days. Mani et al. manufactured a 3D interconnected network, made from NiCo2S4@CoS2 on carbon cloth that allowed hydrogen peroxide detection between 12.64 nM and 2.104 mM, with real-time detection in living mammalian cells.⁵⁴ However, the production of the NiCo₂S₄@CoS₂ nanostructure on carbon cloth proved to be complex and involved four hydrothermal steps: one sulfurization step followed by three calcination steps over a total of more than 43 h of production time.⁵⁴

3.3. Carbon and Graphene Fibers. Fiber substrates in sensing applications have gained increasing interest over the past decade. Fiber-shaped electrodes show many advantages over two-dimensional systems, such as the lower sample volume needed for analysis, improved signal-to-noise ratio, and increased current density.⁵⁵ Carbon fibers (CFs) can be produced via different routes, such as carbonization or graphitization of carbon-rich materials, e.g., cellulose, pitch, and polyacrylonitrile.⁶⁴ Ag NPs embedded into Nafion decorated with carbon microfibers (C μ Fs) have been used to detect hydrogen peroxide.²⁹ Although the sensor showed a response in less than 1 s, a wide detection range both in amperometric and impedimetric applications, and good selectivity and stability,²⁹ the fiber electrode had to be kept inside a glass capillary tube, making it not fully flexible anymore. In the development of CFs, graphene fibers (GFs) have become the focus of different studies. Contrary to CFs, which are polycrystalline, GFs are produced by carefully stacking graphene sheets, which grants them better mechanical and electrical properties.⁶⁴ Gold nanosheet and MnO₂nanowire (NW) @Au-NPs have been electrodeposited on two GF-based sensors respectively to detect hydrogen peroxide in cell culture (Figure 2D).^{55,56} The greatest advantage of carbonaceous fiber sensors is the remarkable reduction of the sample volume required to less than 5 mL, while for carbon cloth and rGO paper sensors, the required sample is commonly above 20 mL in three-electrode systems.⁵⁵ From the research reviewed, this advantage seems to come at the cost of an average increase in LOD and a shift of the detection ranges toward higher concentration of H₂O₂ compared to the twodimensional carbonaceous substrates.

4. POLYMERIC SUBSTRATES

Contrarily to carbonaceous materials and paper, polymers do not directly contribute to the detection chemistry or to the sensing material immobilization. Polymeric substrates, such as polyethylene terephthalate (PET) and polyimide (PI), provide physical support for the sensor, are flexible, have good



Figure 3. A) Schematic demonstrating the carbon differentiated materials followed by the most common techniques and functionalization of the carbonous materials. B) Electrochemically reduced GO paper (depicted as ERGO paper) is produced from graphene oxide and its cross section (SEM image).⁵¹ Reprinted in part with permission from ref 51. Copyright 2015 Elsevier. C) An image of a carbon cloth-based sensor used as a working electrode in a three-electrode system in liquid.⁵³ Reprinted in part with permission from ref 53. Copyright 2019 Elsevier. D) Figures representing a) a schematic, b) and c) photos, and d) the SEM image of a GF knot of the sensor developed by Peng et al.⁵⁵ Reprinted in part with permission from ref 55. Copyright 2018 Elsevier.

mechanical properties, thermal and chemical stability, and are relatively inexpensive (PI cost is ≈ 20 USD m⁻² and PET cost is ≈ 5 USD m⁻² compared to ≈ 30 USD m⁻² for carbon cloth).^{58,60} Moreover, using polymers as substrates may improve the sensor adherence to other surfaces, such as packaging. As such, modifying PET or PI with a sensing material is a promising approach to create flexible and scalable

sensors in food, health, and environmental monitoring. It is also worth noticing that the sensors' performance is dependent on the substrate and on what and how the sensing material is attached to the polymer surface.

The most common techniques that have been used to decorate polymer films with sensor components were casting or coating^{39,57,60,65} and printing (Figure 3A).³⁸ Manufacturing



Figure 4. A) Schematic summarizing the common techniques with which plastic substrates are modified: laser, printing, and stamping. Each method requires a further step of functionalization before being active. In the case of printing, this last step can be avoided as functionalization can be carried out at the same time as substrate preparation. B) Device developed by Vahidpour et al. The two sensors, one active and one passive, are produced via different casting steps.⁴⁴ Reproduced with permission from ref 44. Copyright 2018 John Wiley and Sons. C) Photos of eGR and nGN stamped on the PET substrate from Stromberg et al.⁶⁰ Reprinted in part with permission from ref 60. Copyright 2019 Springer Nature. D) Schematic representation of the manufacturing steps used by Lu et al. to produce their sensor: (a) PI sheet undergoes laser engraving; (b) formation of LASER engraved porous graphene, (c) its flexibility, (d) and what it looks like; (e) electrodeposition of Pt NPS; (f) drop casting of glucose oxidase (GOD in the figure); (g) mechanism of detection of the produced sensor; and (h) proof that different shapes of electrodes can be produced via laser engraving. Notice how after laser engraving, a common carbonaceous substrate functionalization technique such as electrodeposition can be used.⁵⁸ Reproduced with permission from ref 58. Copyright 2021 Elsevier.

based on casting is usually more complex compared with printing, as it usually requires multiple steps. The presence of an adhesive layer, photolithography steps, electrode coating along with resistive layers, and multiple casting steps may be required to avoid sensor delamination. Once a conductive material is deposited on the plastic substrate, electrodeposition can be carried out to decorate the sensor with H_2O_2 -sensitive metallic nanostructures. Despite the extra step, manufacturing remains quite simple and scalable, and a detection limit as low as 1.91 μ M was achieved with Pt NPs decorated on expanded graphite electrodes deposited on a plastic substrate (Figure 3C).⁶⁰ Laser-induced graphene (LIG) is an alternative

approach to introduce the conductive tracks on plastic films. In LIG, graphene is obtained by ablating a PI sheet with a laser beam. Here, the high temperature caused by a laser induces a local carbonization which is accompanied by graphene formation.⁵⁸ The physical properties of this graphene layer can be changed by tuning the laser patterning parameters. The production of LIG does not involve any further chemical modification, making it more eco-friendly and less hazardous than traditional methods.⁵⁸ The sensors obtained by this method can be used for different applications, including hydrogen peroxide detection. As an example, Kothuru et al. used LIG to manufacture flexible sensors for detection of H₂O₂ at concentrations ranging between 1 μ M and 10 μ M.⁵⁹ To broaden the range of detection, H₂O₂-sensitive nanoparticles such as Pt NPs can be electrodeposited on the graphene generated by LIG on PI substrates (Figure 3D).⁵⁸ Similarly, a laser can be used to deposit metal structures on plastic substrates. A Ag 3D porous structure made of nano sized Ag particles was directly deposited on a PI film by pulsed-laser deposition. The developed sensor was able to electrochemically detect hydrogen peroxide while being insensitive toward common interferents such as glucose and ascorbic acid.⁴⁰ Additionally, using a laser allows a green synthesis approach, which eradicates the use of other chemical reagents, compared with traditional methods of Ag nanostructure synthesis. Table 1 also lists the sensing parameters and sensing mechanism of flexible H₂O₂ sensors based on plastic substrates in more detail.

5. PAPER SUBSTRATES

Being easy to source and handle, paper is an optimal substrate material for sensing applications as it is easily sourced and handled, cost-effective, and flexible and can be used for developing disposable sensors.⁴⁵ Contrary to carbon-based materials, paper is not conductive. Hence, it does not electrochemically participate in the detection mechanism. Moreover, paper is not as tough as carbonaceous materials or plastics, given their low tear resistance—the tear strength of paper ranges from 3 N mm⁻¹ to 7 N mm⁻¹, compared with the 32.4 N mm⁻¹ on average for PET.⁶⁶ The stability of paper deteriorates even more in the presence of water, limiting its application in wet environments.⁶⁷

Like other substrate materials, paper can also be decorated with nanostructures to make it conductive and sensitive to H_2O_2 (Figure 4A). For instance, tea bag filter paper was used as a substrate and modified with MnS microcubes enveloped in a MoS₂ nanosheet to detect H₂O₂.⁴² More importantly, the porous structure of paper makes it suitable for enzyme immobilization.⁴⁷ Given its high porosity and biocompatibility, enzymes, and even polymers, can easily be trapped inside paper's 3D structure. For example, conductive polymers such as PEDOT:PSS can be adsorbed by paper, making it conductive. Furthermore, the immobilization of horseradish peroxidase (HRP), an enzyme that catalyzes hydrogen peroxide degradation, on paper can also make it sensitive to H_2O_2 (Figure 4B).⁴⁷ Enzyme-based sensors can be highly sensitive and selective, thanks to the enzyme intrinsic properties. Moreover, a peroxidase enzyme, such as catalase or HRP, can be coupled with an oxidase enzyme, allowing the detection of different analytes, e.g., glucose, amines, and lactose.⁶⁸ On the downside, enzymes add further restrictions to the sensor applications, as they are active only within the defined range of environmental conditions. To overcome the

restrictions posed to the sensors' working conditions by enzymes, materials mimicking the catalytic activity of enzymes have been studied and developed. As an example, chitosan can catalyze the oxidation of TMB in the presence of hydrogen peroxide, leading to a change in color.⁶¹ Although the chitosan catalyzed reaction allows the avoidance of the use of a peroxidase enzyme, its catalysis mechanism is still unclear and needs to be studied further. Moreover, in the work of Ravagan et al., the H₂O₂ detection reaction did not take place on chromatography paper but in liquid, and it was only after the reaction had taken place that the chemicals were deposited on paper.⁶¹ As such, further experiments are required to ensure the activity of reagents in the solid state. Similarly, Dutta et al. developed a Tb³⁺ cholate-based hydrogel which included their own developed molecule (called 2 or pro-synthesizer in the paper and that includes a boronate group) to detect hydrogen peroxide.³¹ Here, H₂O₂ reacts with the boronate group, allowing then the energy transfer between the pro-synthesizer and Tb³⁺, resulting in a fluorescent emission and, hence, detection. This change in emission is measured with a spectrometer but can also be quantified via an imaging software as it is visible with the naked eye. However, this change of the light quantification method has a negative impact on both the LOD and detection range: the LOD increases from 0.7 μ M to 2 μ M, while the saturation concentration drops from 300 μ M to 50 μ M, drastically reducing the detection range. In addition, the reaction occurs before the solution is deposited onto paper; subsequently, more experiments are required to confirm its activity on paper. Moreover, the production of the pro-synthesizer requires the use of a wide range of hazardous chemicals, such as lanthanides, of which toxicity is still debatable,⁶⁹ which could hinder their applications in the design of sensors that are directly exposed to the human body or foods.

Another advantage of paper compared to polymers and carbonaceous substrates is the feasibility of building microfluidic systems. Microfluidic devices rely on the manipulation of small volumes of fluids within a μ L range, hence reducing the consumption of both reagents and sample size, while offering high sensitivity and fast response.⁷⁰ Pesaran et al. developed a microfluidic electrochemical paper-based device (ePAD) to detect hydrogen peroxide.²⁷ The ePAD was built by creating hydrophilic and hydrophobic areas on filter paper, which was then folded including modified carbon paste electrodes. Then, the variation in potential after the addition of H₂O₂ is measured. These microfluidic devices exhibit a high performance by showing a remarkably low LOD of 0.4 nM and a response time of 12 s. Their main drawback is their high sensitivity to external forces, and although flexible, they need to be placed in a solid case to achieve a reliable response. For example, the ePAD developed by Pesaran et al. was placed between two glass slides to stabilize the signal reading. Along with more details, in Table 1, it is possible to see how paperbased sensors minimize the sample volume required while still maintaining a comparable LOD to carbonaceous and polymer substrate-based sensors.

6. OTHER SUBSTRATES

In addition to carbonaceous materials, paper, and polymers, as shown in Table 1, other flexible materials have been used as the substrate. For example, flexible glass has been utilized as a substrate for printing graphite and Ag/AgCl inks in the production of an H_2O_2 sensor.⁶² Although the absence of

pores and the nonconductive nature of glass make it similar to polymeric substrates, glass substrates exhibit a higher thermal and chemical stability. Enhanced stability is beneficial for different applications and also for surface modification and decoration techniques that are performed under harsh environmental conditions. However, glass is not only more expensive than PI and PET (\approx 5000 USD m⁻² against \approx 20 USD m⁻² and ≈ 5 USD m⁻², respectively) but also not as flexible. As such, the glass substrate is used for specific applications where chemical stability is needed, and subsequently, the increase in cost is justified. A silicon wafer is another flexible material which has been attempted as a substrate. Huang et al. used a Si wafer as a substrate for sputtering of a Au-Ag alloy, which was then exposed to nitric acid, obtaining a porous gold film.⁴¹ Prussian blue was then electrodeposited on the sensor to enable detection of H_2O_2 in a range between 1 μ M and 17 mM with an LOD of 0.22 μ M. The surface modification procedure, and especially the use of nitric acid, was possible due to the Si wafer chemical stability. Despite the optimal detection performance, the presence of Cr, which may form toxic compounds, and the high cost of Si wafer (\approx 3000 USD m⁻²) hinder the use of this sensor in some applications. Additionally, Si has a high stiffness, hence the mechanical flexibility of its wafer is determined by the thickness of the wafer. Therefore, to fabricate a truly flexible H₂O₂ sensor based on Si wafers as substrates, thickness must be reduced considerably, which, in turn, leads to deterioration of mechanical robustness.

7. CHALLENGES AND FUTURE PERSPECTIVES

The library of materials used to fabricate flexible sensors is generally selected by considering the physical, chemical, and electrochemical properties of the components. However, in many applications such as the environment, health, and food monitoring, it is pivotal to take into consideration other factors such as cost, biocompatibility, sample volume, and ease of handling. The required sample volume, for instance, is one of the key factors that has been widely neglected, as shown in Table 1. Most of the flexible H₂O₂ sensors from the reported studies did not report the sample volume used for analysis. On the other hand, a few studies reported a sample volume above 5 mL. In applications where the amount of sample is limited, such as tears, saliva, and blood, developing a sensor that requires a low sample volume for detection is crucial. In this regard, carbonaceous and polymeric substrates often use the sample volume in the milliliter range, which might not be suitable for all applications. On the other hand, paper substrates commonly require the sample volume within the range of microliters. The reduction of the sample volume by paper-based sensors is due to their high surface to volume ratio granted by their porous structure. Stability is another important parameter for comparing the performance of sensors that has not been thoroughly reported. Among the studies reviewed in this paper, only nearly 50% reported the sensor's stability. However, further complications arise as for stability a sensor's shelf life and the working period (i.e., the duration a sensor could function continuously) are reported interchangeably. As a result of this, it is difficult to make a conclusive comparison between the performance of sensors. Moreover, it is even more difficult to understand whether a sensor is suitable for a specific application. For example, in the case of smart food packaging, it is important that the sensor remains stable during storage (shelf life) and also active for the whole

time the food is inside the package to monitor its spoilage rate and quality. For medical and diagnostic applications, the shelf life under different storage conditions is critical for determining their expiration date for safe application of devices. Nevertheless, it is anticipated that the functionalization of a substrate can make a significant impact on the stability of a sensor. For example, functionalization by labile biomolecules such as an enzyme decreases the stability of sensors due to faster degradation, denaturation, and loss of their activity.

As much as cost is concerned, PET is the cheapest substrate material, followed by paper (Paper cost varies greatly with its quality, so an average quality filter paper cost is used for comparison.), carbon fibers, carbon cloth, and PI. However, polymers do not show the same porous structure as paper or the same conductivity as carbonaceous materials. Hence, there is a trade-off between cost and utility. The same can be said for manufacturing. Carbonaceous substrates benefit from being conductive, which make electrochemical functionalization processes available and allow one-step synthesis, such as in Dong et al. work,⁵¹ thus lowering the cost of functionalization. Paper and polymers show similar functionalization techniques, such as 3D printing or casting, which are scalable and affordable. However, polymer substrate-based sensors are at risk of delamination. This risk is low with paper, thanks to its 3D porous structure. Therefore, the most appealing technique for polymers is laser engraving.58,59 Although the risk of delamination could still be present, the ability of producing graphene directly on polymer substrates opens an avenue for electrochemical functionalization, adding functionality to the cost-effectiveness of polymers. LIG on polymers is a promising technique to produce sensors, even though it still needs further research in the sensor field.

Figure 5 shows how polymer-based sensors struggle in terms of LOD, which is usually in the micromolar order, and the detection range compared with carbon cloth- and rGO-based sensors, which can have an LOD as low as 2 nM. The same can be claimed for CF sensors. The importance of a low LOD depends entirely on the application range: if this is narrow, a low LOD gains value, as the difference between an acceptable hydrogen peroxide concentration and an unacceptable one can be very small.

From these data it can be concluded that, due to the high cost of carbonaceous substrates, their application is justifiable for the design of sensors that are used in healthcare. In medical applications, high performance and a low detection level are critical for diagnostic purposes. Hence, the higher detection performance of the carbonaceous substrate is preferred and justifies the increase in overall cost. However, the major obstacle of using this class of material is the large sample volume for analysis. CC-based sensors reviewed reported a sample volume required of more than 20 mL, which can possibly only be viable for environmental testing. To tackle this issue, it is recommended to use carbonaceous fibers (GFs and $C\mu$ Fs), as they require a smaller sample volume for detection. Moreover, their toughness makes it possible to produce fabrics with integrated carbonaceous fibers that can be used for developing wearable sensors. Nevertheless, while fibers reduce the sample volume for analysis, these sensors also experience a lower detection level. Therefore, there is a trade-off between the detection performance and sample volume required, which should be investigated based on a case-to-case application.

Paper-based sensors are cost-effective, biocompatible, and porous. Its three-dimensional porous structures grant it high



Figure 5. A) Schematic of paper advantages and common ways to modify it. Notice that before functionalization paper needs to be patterned to create hydrophilic and hydrophobic areas (via wax printing, lithography, use of mask, etc.). Alternatively, paper can be cut and then assembled in a final device. B) Photo of the inkjetprinted sensor developed by Giaretta et al.47 Reprinted in part with permission from ref 47. Copyright 2021 John Wiley and Sons. C) Schematic representing the production steps of the device developed by Maier et al.⁴⁵ Notice how paper is first patterned via wax printing, then the electrodes are screen printed on top of it, and finally it is assembled in the final device. Reprinted in part with permission from ref 45. Copyright 2019 American Chemical Society.

permeability, which allow liquids and gases to infiltrate through its pores. As such, paper-based sensors are characterized by high active surface and surface to volume ratios, which improves their detection. These characteristics make them suitable for vapor sensing in many applications such as masks and food packaging, as demonstrated by Giaretta et al. and Vahidpour et al.^{44,47} However, the drawbacks of paper-based sensors include their limit of applications for detection of gases in a wet environment and that they are not reusable. In addition, paper substrates may be more adaptable in food packaging made from paper or cardboard material, as they can easily be attached to their surfaces. Their use is also preferred in microfluidic applications for POC detection as the sample volume can be reduced due to its high porosity and high active surface.²⁷

PET and PI are the most common polymers used as substrates in the development of H_2O_2 flexible sensors. Polymers are characterized by reasonable cost and amenable properties. They are ideal for the design of smart food packaging, as they can easily adhere to the packaging materials.⁷¹ Compared with paper-based sensors, polymerbased substrates struggle to achieve detection of vapors due to their minimal porosity and permeability, that hinders fluid adsorption. To overcome this obstacle and allow the construction of sensors for the detection of biomarkers and diagnostic devices for medical applications, the surface of polymer substrates needs to be functionalized. Alternatively, conductive polymers could be included in the sensors' constructs, to achieve a more efficient design by rendering the substrate electroactive.^{72,73} Finally, the high cost of flexible glass and silicon wafers, compared with other substrates, restrain their applications to only a few specific cases where mechanical strength and chemical stability are pivotal, such as environmental monitoring of waste waters and other industrial applications (Table 2). Further technological advances could tackle some of the shortcomings of current substrates. The emerging field of hydrogels and fibrous polymers might open a new avenue for the development of porous substrates with

substrate	advantages	disadvantages	applications	future proposed development
carbonaceous materials	• electrochemically active	 high cost 	• wearable	• reduce cost
	• tough	 usually functionalized with nanomaterials 	• healthcare applications	 integration into fabric for wearable application (GF, CμF)
	• biocompatible can be fiber-shaped and woven into fabric (CC, GF, $C\mu$ F)	• tedious manufacturing	 environmental applications 	 reduce sample volume (CC, rGO)
polymers	• low cost	• not electroactive	• wearable sensors (small dimensions)	 improve porosity (hydrogel, fibrous polymers, etc.)
	• amenable	• low porosity	 food packaging (especially plastic) 	 functional polymers (e.g., conductive polymers)
	• adhere to various surfaces	 may not be recyclable 		 hybrid materials
	 possible green synthesis (LIG) ease of manufacturing 			
paper	• 3D porous structure	• low tear resistance	 food packaging (especially paper) 	• hybrid materials
	• biocompatible	 not suitable for wet environment 	 vapor sensing (e.g., masks) 	
	• suitable for vapor detection	 not reusable 	 POC disposable 	
	• low cost		healthcare (microfluidica)	
	• simple to manufacture		(interonations)	
	• low cost, easy to manufacture			
others (flexible glass and Si wafer)	 high mechanical and chemical stability 	 high cost 	 specific environmental 	• N.A.
		 low flexibility depends on thickness 	applications	

Table 2. Different Types of Substrates Used for Designing Sensors, Their Advantages, Disadvantages, and Applications

improved functionality,^{74–76} hence making them more suitable for designing sensors. Although these technologies have been already applied for the development of sensors, they are often used as functionalization materials rather than substrates, making them inflexible and not self-standing. Decreasing the cost of carbonaceous fibers and the creation of hybrid materials are other alternative strategies for the future design of sensor devices.^{77–79}

Regarding the functionalization, from Figure 5 it becomes clear how most of the flexible H2O2 sensors reported in the literature employ at least one type of nanostructure (patterned bars). Although a direct comparison is hard to make due to the concurrent change in the substrate and sensing material, nanostructures do not seem to provide a clear improvement in sensitivity. It can be concluded that, while nanomaterials show many advantages, they are not strictly necessary for detecting H_2O_2 in the concentration range required by the many realworld applications. Furthermore, nanomaterials are often associated with toxicity, interference with neurotransmitters, complicated synthesis, cumbersome assembly, lack of reproducibility, and oxygen sensitivity.² Hence, the development of nanomaterial-free sensors is advisable for applications where cost, ease of fabrication, or biocompatibility are the main design criteria. The use of bioreceptors such as enzymes and the use of non-nanostructured functionalization materials, such as MnO₂ powder, are sufficient in that regard.^{27,44,47,61} Figure 5 also highlights that the detection range of most flexible H₂O₂ sensors reported in the literature already spans over multiple orders of magnitude, easily satisfying the required LOD and detection range of numerous applications, especially food and health domains. Indeed, in some examples, the developed sensors have been overengineered to reach an extremely low LOD or wide detection range that are not necessary requirements for most applications. Therefore, rather than further improving the sensing performance of flexible H_2O_2 sensors, it is recommended that future research focuses on addressing the manufacturing obstacles, sustainability, scalability, and device design. As an example, Vahidpour et al. developed a calorimetric sensor capable of detecting gaseous hydrogen peroxide in concentrations ranging from 0 to 7.7% v/ v using MnO₂ powder.⁴⁴ The sensor takes advantage of the heat released when H₂O₂ decomposes on a heating element. The released heat alters the electrical resistance of the circuit proportional to the level of H₂O₂. The sensor consists of two Pt/Ti elements, one of which is decorated with MnO₂, while the second serves as a passive element to eliminate the effect of variation in temperature caused by sources other than H₂O₂ decomposition on the active element. The relative ease of manufacturing, flexibility, and wide working temperature range of the sensor make it suitable for inline monitoring of the sterilization process of food packages (Figure 3D). Addressing health monitoring, Maier et al. developed a flexible sensor able to detect hydrogen peroxide in simulated breath.⁴⁵ The sensor was produced by screen printing and placed in a filter extension at the end of a respiratory mask (Figure 4C). Although the sensor could not be used in real application as the detection range (5 μ M-320 μ M) is above the H₂O₂ concentration in human breath (0.1 μ M–1.5 μ M in the case of exhaled breath condensate), the designed device functioned as a proof of concept of a commercial sensor. Although such application-driven sensors might be less sensitive than those developed with a focus on sensing performance alone, they possess an edge over the latter in terms of feasibility and

commercialization viability. Many studies on the flexible H_2O_2 sensor however overlook the fundamental aspects of sensor production hindering their chances for commercialization. Therefore, it is critical to consider a greater engineering approach in design of flexible H_2O_2 sensors to tailor sensors parameters for real-world applications (Figure 6).



Figure 6. Ranges of sensors referenced in this review. The red dots indicate their respective LOD. Sensors' substrate and functionalization are listed on the left, while references and the substrate class are indicated on the right. The pattern in the bars indicates which sensors make use of nanostructured compounds. The colored shades define the concentration range of interest for the environment (blue), food (purple), and health (green) applications. These ranges are created by choosing the maximum and minimum H_2O_2 concentration of the applications listed in Figure 1. HyG stands for hydrogel. * The sensor should have two ranges, which separation is not visible in the logarithmic scale. The LOD shown is the one for the lowest range. ** The sensor's range and LOD shown are the ones for the amperometric mode. *** The sensor was used also with other polymeric substrates (Table 1).

8. CONCLUSION

The substrate of sensors provides flexibility, which is necessary for the development of novel applications such as wearable sensors and smart food packaging (Figure 7). They also play a key role in determining fabrication, stability, and design of sensors. In this review article, flexible H2O2 sensors are categorized based on their substrates, which include carbonaceous materials (i.e., reduced graphene oxide, carbon cloth, carbon and graphene fiber), polymers, paper, flexible glass, and silicon wafers. Polymeric substrates are the cheapest substrate, but they are inferior to carbonaceous materials and paper due to their lower electrical conductivity and poor permeability/ lack of porosity, respectively. These characteristics affect how each substrate can be functionalized to further improve sensing performance such as limit of detection (LOD) and detection range. Although a simple comparison is hard to make, twodimensional carbonaceous substrates seem to present the best sensing performances, considering both LOD and detection range. However, it is important to note that even sensors with simple constructs already satisfy the detection requirements for



Figure 7. Parameters that need to optimize to achieve a sensor commercialization. Both science (green) and engineering (light brown) aspects need to be assessed and optimized for a sensor to be suitable for real-world application.

most applications in the environmental, medical, and foodrelated fields for the detection of hydrogen peroxide. In other words, excessive emphasis on improving sensing performance has led to designs which are overcomplicated and overengineered, limiting their chance for commercialization. Therefore, it is recommended to consider other key factors for the future design of sensors such as improving the technological aspect of manufacturing, cost, and sample volume reduction that are also critical for sensor performance and application.

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Notes

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ABBREVIATIONS

- CB = carbon black
- CC = carbon cloth
- CF = carbon fiber
- $C\mu F$ = carbon microfiber
- CP = carbon paste
- eGR = expanded graphite
- GF = graphene fiber
- GO = graphene oxide
- GR = graphite
- HRP = horseradish peroxidase
- LIG = laser induced graphene
- LOD = limit of detection
- MOF = metal oxide framework
- NA = nanoarray
- NC = nanocrystal
- nGN = nanoplatelet graphene
- NP = nanoparticle
- NR = nanorod
- NS = nanosheet
- NW = nanowire
- OA = organic acid
- PANI = polyaniline
- PB = Prussian blue
- PCL = polycaprolactone
- PEDOT:PSS = poly(3,4-ethylenedioxythiophene) polystyr-
- enesulfonate
- PET = polyethylene terephthalate
- PI = polyimide
- rGO = reduced graphene oxide
- TMB = 3,3',5,5'-tetramethylbenzidine

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