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SUSTAINABLE TECHNOLOGIES IN WATER TREATMENT AND DESALINATION

Laser-induced graphene-based electrochemical biosensors for environmental applications: a perspective

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Abstract

Biosensors are miniaturized devices that provide the advantage of in situ and point-of-care monitoring of analytes of interest. Electrochemical biosensors use the mechanism of oxidation–reduction reactions and measurement of corresponding electron transfer as changes in current, voltage, or other parameters using different electrochemical techniques. The use of electrochemically active materials is critical for the effective functioning of electrochemical biosensors. Laser-induced graphene (LIG) has garnered increasing interest in biosensor development and improvement due to its high electrical conductivity, specific surface area, and simple and scalable fabrication process. The effort of this perspective is to understand the existing classes of analytes and the mechanisms of their detection using LIG-based biosensors. The manuscript has highlighted the potential use of LIG, its modifications, and its use with various receptors for sensing various environmental pollutants. Although the conventional graphene-based sensors effectively detect trace levels for many analytes in different applications, the chemical and energy-intensive fabrication and time-consuming processes make it imperative to explore a low-cost and scalable option such as LIG for biosensors production. The focus of these potential biosensors has been kept on detection analytes of environmental significance such as heavy metals ions, organic and inorganic compounds, fertilizers, pesticides, pathogens, and antibiotics. The use of LIG directly as an electrode, its modifications with nanomaterials and polymers, and its combination with bioreceptors such as aptamers and polymers has been summarized. The strengths, weaknesses, opportunities, and threats analysis has also been done to understand the viability of incorporating LIG-based electrochemical biosensors for environmental applications.

Keywords Electrochemical biosensors · Environmental monitoring · Detection mechanisms · Biorecognition elements · Laser-induced graphene · Electrochemical analysis

Introduction

Detection of various biomolecules, chemicals, heavy metals, organic and inorganic compounds, etc., collectively termed as "analytes," is of high importance in industrial manufacturing as well as scientific research. The detection of the analytes has to be both qualitative and quantitative (Lu et al. 2017). The qualitative aspect refers to "selective" detection of the analyte from amongst a variety of possibly interfering substances that may be present in the same sampling matrix and environment. On the other hand, the quantitative aspect refers to the amount of analyte that can be detected by the particular method under consideration. Biosensors are a special class of sensors that incorporate the use of a biorecognition element such as DNA and proteins to generate the physicochemical signal for measuring the analyte (Bhalla et al. 2016). Biosensors have a wide range of applications depending upon the presence and possible threat posed by the analyte under consideration (Singh et al. 2020). There are numerous areas of application of biosensors such as environmental monitoring, soil quality monitoring for agriculture, food quality monitoring, and clinical diagnostics (Oliveira et al. 2013; Shi et al. 2013; Kaur et al.

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2015; Handford et al. 2015; Bobrinetskiy and Knezevic 2018; Beduk et al. 2021) (Fig. 1). The use of biosensors for environmental monitoring can facilitate the prompt detection of pollutants and the corresponding use of mitigation measures to avoid health and environmental hazards (Dias and Edwards 2003; Halilović et al. 2019). Defining the safety limits for different conventional and emerging pollutants that all stakeholders must follow requires rapid and accurate sensing. The use of dietary medicines, chemical fertilizers, and pesticides for plants and animals can give direct entry to various chemicals into the body, which underlines the need for prompt detection to reduce or eliminate the risks posed by these compounds.

To date, many analytical instrumentation methods have been reported to detect various analytes. These include spectroscopic techniques such as UV–vis spectroscopy (Tahir et al. 2012), inductively coupled plasma optical emission spectrometry (ICP-OES) (Chen et al. 2009), and electrothermal atomic absorption spectrometry (ETAAS) (Anawar 2012). Chromatographic analytical techniques include gas chromatography and high-performance liquid chromatography coupled with capillary electrophoresis or mass spectrometry (Nasraoui et al. 2021). However, these methods often require complicated and expensive instruments, pretreatment and preparation of samples, and trained professionals, making them hard to use in situ (Justino et al. 2017). To overcome the limitations of conventional analytical methods, biosensors have garnered interest for use in the sensing of various analytes in a variety of applications. Food safety, environmental hazards, biomedical applications, etc. are invariably more critical in underdeveloped regions where large-scale analytical laboratories are not easily available or accessible. Hence, considering the field applications, detecting the analytes using low-cost, portable, miniaturized devices is important (Vanegas et al. 2018).

Biosensors are analytical devices that convert a biochemical reaction into a measurable physicochemical signal, which provides a quantitative assessment of the analyte concentration (Bhalla et al. 2016). An analyte may be a chemical compound like a pollutant, a hormone, a biomolecule, a pharmaceutical compound, or any other chemical that is to be detected. This analyte interferes with the metabolic activity of the biorecognition element such as biomolecules or distorts their original conformation (e.g., DNA) (Ozsoz et al. 2003; Labuda et al. 2005). Due to this interference, a biochemical signal is generated, which is then converted to a processible signal by the transducer. For developing a biosensor, the use of material science and the mechanism of interaction of the analyte with biomolecules are critical (Zhang et al. 2000; Justino et al. 2017). Biosensors can sensitively and selectively detect a wide range of compounds and macromolecules strongly relevant to applications such as human health diagnosis, food safety, and environmental monitoring (Kumaravel and Chandrasekaran 2015; Peña-Bahamonde et al. 2018; Taniselass et al. 2019; Chai and Oh 2020; Khanmohammadi et al. 2020). A biosensor is a detection device having three major components: (1) a biorecognition element alternatively called a bioreceptor, (2) a transducer, and (3) a signal amplifier. Thus, a biosensor is a synergistic device combining biotechnology and electronic sensing. Based on the transduction principle, the biosensors are mainly classified as (a) electrochemical; (b) thermal; (c) piezoelectric; and (d) optical biosensors (Bhalla et al. 2016).

Electrochemical biosensors typically convert the biochemical signal into electrical signals. Electrodes are key components of electrochemical biosensors that support the bioreceptor immobilization or the site for direct adsorption of analytes and facilitate the transfer of electrical charge through the sensor circuit (Rezaei et al. 2016; Vanegas et al. 2018). Electrochemical oxidation or reduction of the analyte under consideration is carried out at the transducer interface. The redox process results in the generation of an electric charge, which is carried through the electrode and measured at the output signal processing device like a potentiostat (Yang and Yan 2021). Modification of the electrode using metal nanoparticles, polymers, selective enzymes, antibodies, etc. is a common approach to improve the electrocatalytic activity of the transducer (Shao et al. 2010; Morales and Halpern 2018). This results in enhanced signals and hence better sensitivity for the analyte. Electrochemical biosensors have received increased attention due to their low detection limits, adjustable selectivity, and fast responses (Thévenot et al. 2001; Ronkainen et al. 2010; Saei et al. 2013). They are less expensive as compared to conventional analytical methods, are easy to handle, and can be fabricated in miniaturized forms. This allows for portability of the device and hence easy in situ application in sensing of the analytes.

This article explores the possibilities of laser-induced graphene (LIG), a novel form of graphene that has garnered increasing interest for electrochemical biosensors, for application in environmental monitoring. The scope of the article encompasses the detection of pollutants in the ecosystem that concern environmental safety. To understand the possibilities of electrochemical biosensor applications in this area, first, an overview of existing LIG-based biosensors in the field of environment, biomedical, and food safety applications is done. Based on different receptors, the electrochemical techniques used for these LIG-based biosensors, and the combination of nanomaterials, an inference is drawn for the possible development of environmental biosensors. The major challenges in the context of these potential sensors lie in the complexities and variable constituents of the environment. Possible solutions are identified for the future of environmental biosensors that can address these complexities while giving industry scalable and reliable results.

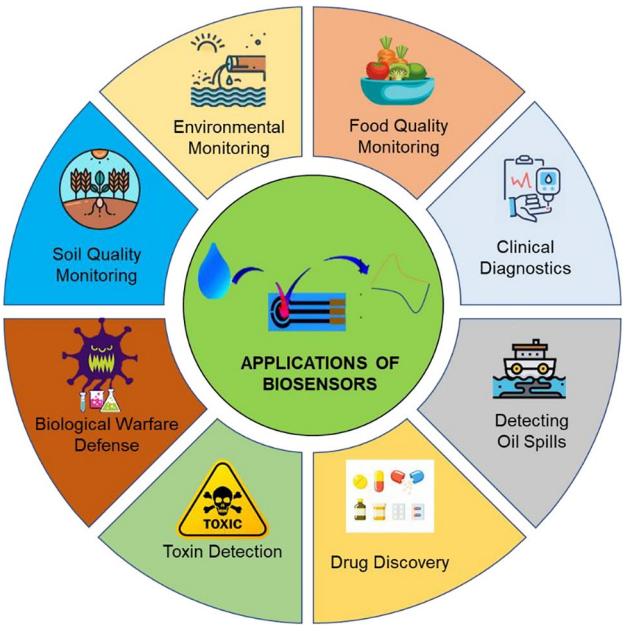

Fig. 1 Applications of biosensors

LIG-based electrochemical sensors

Graphene, a 2D nanostructured allotrope of carbon, has been a very effective material for electrochemical analysis (Wu et al. 2013; Wan et al. 2020a). This is owed largely to its outstanding electrochemical properties and nanosized sheet structure. The nanostructure allows for the measurement of signal currents of nanoamperes and picoamperes, which has permitted the sensing of substances at very low concentrations. Many electrochemical sensors have been developed with graphene in different forms. For example, a sensor for clinical diagnostics was developed using graphene and carbon nanotubes on screen-printed carbon electrodes to detect bilirubin in the blood (Thangamuthu et al. 2018). Another sensor employed graphene and iron oxide (Fe_3O_4) on a glassy carbon electrode to detect the biomarker dopamine and ascorbic acid (Peik-See et al. 2014). An environmental application of a graphene-based sensor has been demonstrated for the detection of the pharmaceutical drug naproxen in treated effluent (Qian et al. 2020). These graphene-based electrochemical sensors exhibit improved performance than the corresponding base electrodes, such as screen-printed carbon electrodes and glassy carbon electrodes. However, the conventional fabrication methods of graphene, viz., mechanical exfoliation, chemical vapor deposition, or reduction of graphene oxide, are time-consuming and expensive. This impacts the scalability of the sensors being fabricated using graphene. Also, difficulties in testing new architectures of biosensors are faced, limiting the advent of new and improved sensing technologies, including multifunctional sensors. Laser-induced graphene (LIG) is a simple and scalable form of graphene developed by direct laser writing on commercial polyimide (PI) or other polymeric material (Lin et al. 2014). It provides a high-resolution, mask-free, and low-cost method to fabricate electrodes for electrochemical sensing. The patterning is done using a computer-controlled machine using different wavelength lasers (Singh et al. 2017). These include the CO_2 laser (10.6 μm) (Singh et al. 2017), near-infrared laser (1064 nm) (Zhang et al. 2016), and semiconductor laser (405 nm) (Cai et al. 2018). SEM images of the fabricated LIG reveal highly porous foamlike 3D structures (Fig. 2a). Also, nanosized ripples are observed in the structure which is attributed to the edges of graphene layers. The high localized temperatures produced by laser induction break the polyimide's C-O, C = O, and C-N bonds (PI). This results in the oxygen and nitrogen atoms being released in the gaseous forms creating the porous structure of LIG. The residual carbon chains are rearranged in sp^2 hybrid form, creating graphene layers (Pimenta et al. 2007). The Raman spectrum of the fabricated LIG shows a 2D band around 2700 cm^{-1} which is a characteristic of 3D multi-layered graphene sheets (Fig. 2a). LIG-based biosensors have shown improved performances as compared to conventional electrodes based like screen-printed electrodes (SPE), glassy carbon electrodes (GCE), or metal electrodes. In a recent study, LIG electrodes have been observed to exhibit higher sensitivity than SPCE and GCE in the detection of paracetamol (Ghanam et al. 2020). LIG holds its advantage over conventional electrode materials due to excellent flexibility, good porosity, large surface area, high conductivity, easy fabrication, and effective cost. LIG is highly electrochemically active due to the abundant edge planes present in its structure (Ye et al. 2018). Hence, it has great promise in the development of electrochemical biosensors.

LIG-based sensors for environmental application

LIG has shown great potential in improving electrochemical sensors due to its superior electrocatalytic properties and fast electron transfer rates (Grossman et al. 2020). These properties make LIG a promising alternative for the application in sensors for environmental monitoring, including sensors for estrogenic compounds such as bisphenol A, pathogens or pathogen-indicating organisms, heavy metals, nutrients, and emerging contaminants including pesticides and antibiotics. A diverse array of pollutants exists, and many new ones are continuously emerging due to new materials in science and technology. A review of the available literature on LIG-based biosensors is important to correlate the development of sensors for different analytes. The study and

understanding of mechanisms of detection can lead to the exploration of possible mechanisms for potential environmental biosensors.

Antibiotics Increased use of antibiotics for medicinal and commercial purposes in the meat industry has led to contamination of wastewater and natural water streams. Traces of antibiotics can be found in food products as well as the waste streams coming from industries. Therefore, trace-level detection of antibiotics is an important requirement for proper monitoring of these wastes and food products. A biosensor fabricated with LIG was demonstrated for detecting chloramphenicol, an antibiotic that can be used against many bacteria. The use of a highly selective recognition technique, i.e., molecularly imprinted polymer (MIP) with Eriochrome Black T (EBT) as a monomer, was done in this sensor (Fig. 3a). MIPs are a type of bioreceptor that has garnered widespread attention in electrochemical sensing of pollutants in recent times. These receptors are based on the concept of creating tailor-made sensing sites for an analyte using functional monomers. The functional monomers are first allowed to bind with the analyte molecules used as templates (Fig. 3) non-covalently. A polymeric layer is then formed around the monomers to immobilize them at specific positions, followed by the removal of the analyte molecules from their respective positions, creating a selective binding site from a sample to be analyzed. In the chloramphenicol biosensor developed using this MIP technique, the limit of detection attained by the sensor was 0.62 nM and a linear response range of 1 nM to 10 mM. These analytical features achieved with LIG as transducer material are much higher than those achieved by comparative commercial graphene and graphene-based screen-printed electrodes (Cardoso et al. 2019). The MIP was also used as a bioreceptor in a dual molecule system with two working electrodes. The two analytes were ascorbic acid and amoxicillin, commonly used in aquaculture, and can be traced into surface waters and wastewaters. The sensor depicted a detection limit of 11.98 nM for amoxicillin with a linear response range of 50 nM to 100 µM. Ascorbic acid was detected in a linear range of 1.5 to 4 mM with 1.356 µA/decade sensitivity. The fabrication steps for these MIP-based sensors have been shown in Fig. 3b (Marques et al. 2020). Thus, the use of MIP on LIG electrodes is shown to be successful in detecting antibiotics and micronutrients in aquaculture. With specific engineering of MIPs and improved analytical performance provided by LIG, the detection of other emerging antibiotic compounds may be possible with improved sensitivity.

Pathogens Pathogen detection is an essential criterion for establishing water quality and, hence, for environmental monitoring. A biosensor was demonstrated for *E. coli* detection using an even distribution of gold, silver, and platinum nanoparticles on LIG that led to a limit of detection of 100 CFU/ml (You et al. 2020). For selective detection of these indicator organisms, an antibody specific to the *E. coli* strain O157:H7 was employed as the biorecognition element. Antibodies are commonly used as receptors for

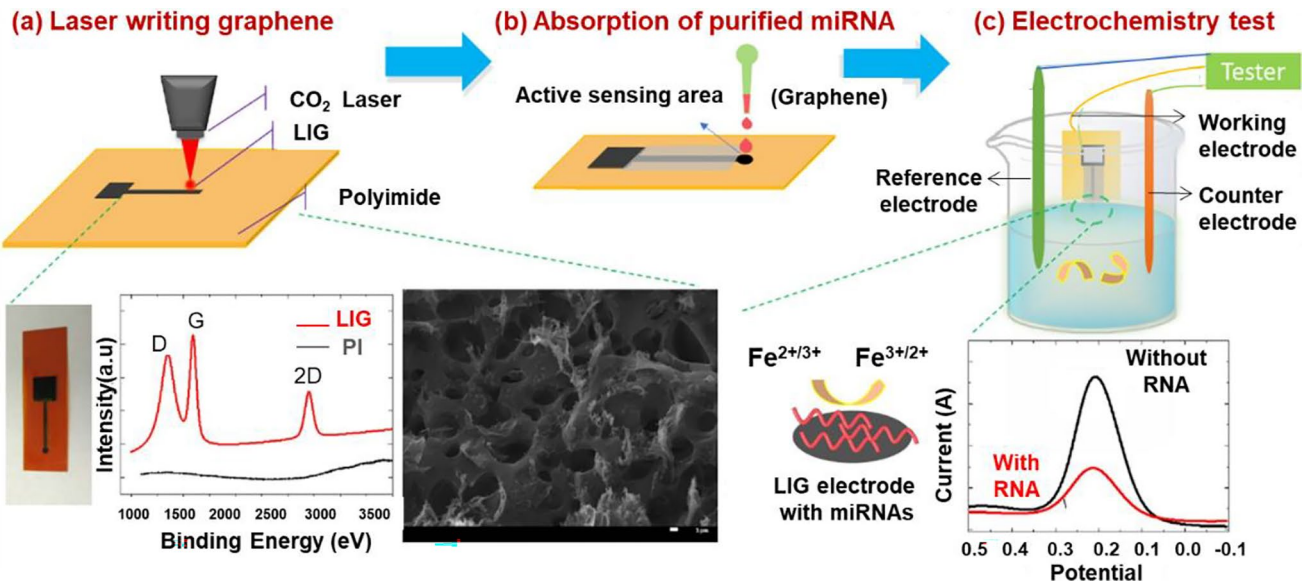

Fig. 2 Typical scheme for LIG-based sensor fabrication, characterization, and electrochemical signal generation. **a** Fabrication of LIG using CO_2 laser with Raman spectrum and SEM image, **b**)introduction of analyte onto LIG surface, **c** electrochemistry test of the analyte using three-electrode system to observe the change in current with and without analyte. Adapted with permission from ref (Wan et al. 2020b). Copyright 2020, Elsevier

pathogenic organism detection. *Salmonella enterica* detection with LIG electrodes has also been done by antibodies as receptors. This pathogen is typically found in food products and is an important marker for food safety. Detection limits of less than 5 CFU/ml are required as a standard for maintaining food quality (Soares et al. 2020). Using this biosensor for *Salmonella enterica* detection, a limit of detection of 13 ± 7 CFU/ml was achieved, which is substantially less than previously developed sensors of a similar application. Developing sensors for food-borne and water-borne pathogens is of major concern for human health and environmental safety. The use of LIG has shown improvement in the performance of electrochemical sensors for pathogen detection (Soares et al. 2020; You et al. 2020). Immunosensors are a class of biosensors defined as such due to the use of antibodies specific to a pathogen for its detection (Vidic and Manzano 2021). There is potential for the development of such immunosensors using bioreceptors specific to particular pathogens. Pathogens of interest for environmental monitoring include protozoa, other bacteria such as *Yersinia enterocolitica*, which causes a type of diarrhea, and viruses for hepatitis A, E, and enteroviruses (Ramadan and Gijs 2012).

Ions Heavy metals and other inorganic ions form a major class of environmental pollutants. Interconvertible oxidation and reduction of these ions are conducive to their detection by electrochemical biosensors. A sensor for ions sensing for copper (Cu^{2+}) detection was developed using LIG and laser-induced metal sulfide nanocomposite. Copper is an essential micronutrient for many organisms but is toxic at higher concentrations. The electrochemical reduction of Cu^{2+} to Cu^{+} leads to the formation of Cu_xS ($x = 1,2$) on the laser-induced nanocomposite electrode surface. This, in turn, leads to a reduction in current that is proportional to the amount of Cu^{2+} which allows for a limit of detection of 2 nM (Ge et al. 2019). Electrochemical detection of another heavy metal

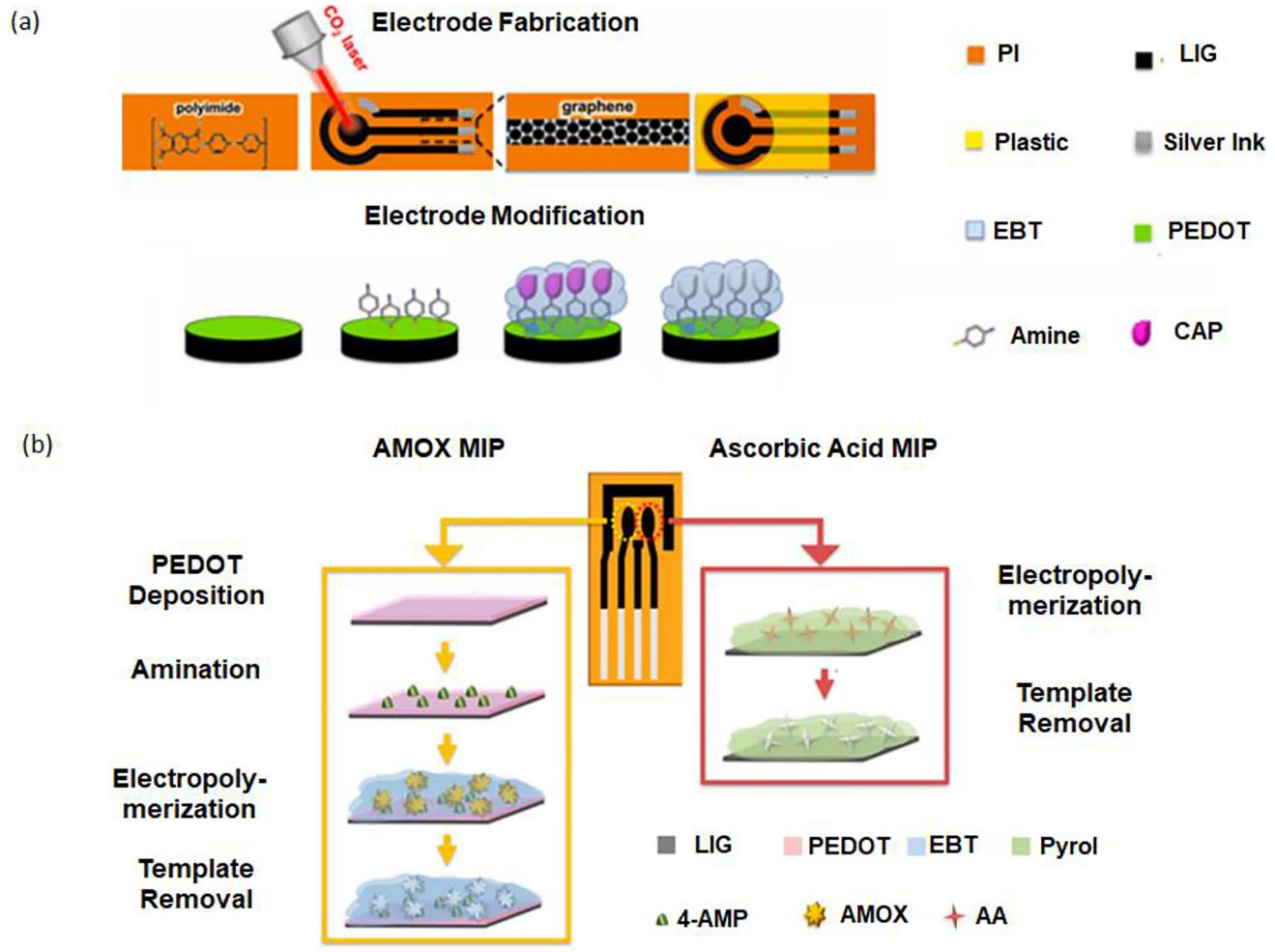

Fig. 3 **a** Fabrication of LIG electrodes and modification for chloramphenicol molecularly imprinted polymer (MIP) biosensor. Adapted with permission from ref (Cardoso et al. 2019). Copyright 2019, Elsevier. **b** MIP technique for selective detection of amoxicillin and ascorbic acid. Adapted with permission from ref (Marques et al. 2020). Copyright 2020, American Chemical Society

ion, i.e., lead (Pb^{2+}), was done by a biosensor with LIG as electrode modified with the polymer poly-L-cysteine and the ionic liquid N-butyl pyridinium hexafluorophosphate or [BuPy]PF_6. The sensor achieved a detection limit of 0.17 μgL^{-1} with high selectivity against interference due to other cations and anions (Lu et al. 2019). Electrochemical sensing of heavy metals can also be performed using the anodic stripping voltammetry (ASV) technique without the use of a bioreceptor. One such LIG-based sensor for lead ion (Pb^{2+}) detection was proposed by Grossman et al. (2020). This sensor involved the use of the ASV technique to generate a distinct peak of lead with a detection limit of around 50 ppb. The comparison of LIG precursors in polyimide (PI) and polyethersulfone (PES) has been made to establish the higher sensitivity of PI-based electrochemical sensors in lead detection (Grossman et al. 2020). Another sensor with a nanocomposite of Bismuth on LIG covered with a Nafion film was used for simultaneous detection of lead and cadmium. This sensor involved square wave ASV to detect the heavy metals with a detection limit of 0.5 and 0.8 µg/L for Cd(II) and Pb(II), respectively (Zhao et al. 2022).

Nitrite (NO_2^-) ion sensors using LIG have also been reported. Nitrite is of importance in the pharmaceutical and chemical industry. It is also an important part of the nitrogen removal process from wastewater and thus an important marker for water quality monitoring. The sensor involved modification of the LIG electrode with -COOH functionalized carbon nanotubes and gold nanoparticles which increased the available electrochemical surface area and hence, the performance of the electrode. Nitrite is oxidized to nitrate with two-electron transfer electrochemical signal generation which resulted in a limit of detection of 0.9 µM (Nasraoui et al. 2021). Another sensor for the detection of nitrogen in the soil, i.e., nitrate (NO_3^-) and ammonium (NH_4^+) ions, was demonstrated by Garland et al. (2018). This sensor employed the use of LIG without any additional nanoparticles. The limit of detection for the sensor was 28.2 ± 25.0 µM for ammonium and 20.6 ± 14.8 µM for nitrate (Garland et al. 2018).

Pesticides Pesticides are a common class of environmental pollutants that are introduced into natural streams due to agricultural activities. Due to the dispersal of pesticides into plant systems and the aquatic environment, many non-target organisms are adversely affected by their exposure and consequent neurotoxic effect (Morrissey et al. 2015). An enzyme-based strategy on LIG with horseradish peroxidase enzyme as the bioreceptor for selective detection of atrazine was employed by Kucherenko et al. (2021). The sensor achieved selectivity against other possible interfering pesticides, viz., glyphosate, dicamba, and 2,4-dichlorophenoxyacetic acid. The sensor exhibited a detection limit of 1.6 µM and a sensitivity of 28.9 nA/µM (Kucherenko et al. 2021). In another study, a non-enzymatic approach was developed to detect a group of neonicotinoid pesticides. Square wave voltammetry (SWV) was used as the electrochemical technique for the detection of clothianidin (CLO), imidacloprid (IMD), thiamethoxam (TMX), and dinotefuran (DNT). The detection limits achieved were 823 nM for CLO, 384 nM for IMD, 338 nM for TMX, and 682 nM for DNT (Johnson et al. 2021). A plant-wearable sensor for detecting the organophosphorus pesticide methyl parathion was also developed using LIG modified with gold nanoparticles as the electrode. The electrode surface was modified with the organophosphorus hydrolase enzyme as a receptor resulting in the linear concentration range of 20 to 500 µM with a detection limit of 0.01 µM (Zhao et al. 2020).

Bisphenol A Bisphenol A (BPA) is a chemical used as a monomer to produce different plastics and resins which enters as waste into the aquatic environment, such as river sediment, and in the fish muscles. Once ingested into the human body through drinking water or food, this pollutant is known to cause carcinogenic and cardiovascular health impacts, making trace-level detection of BPA necessary. Cheng et al. (2016) demonstrated a sensor for detecting BPA based on LIG and aptamers. Aptamers are single-stranded DNA or RNA oligonucleotides (Yagati et al. 2020) that impart high selectivity and allow for low detection limits to electrochemical biosensors. The LIG electrodes are formed as interdigitated arrays on flexible polyimide sheets with aptamers immobilized on their surface. Due to the binding of aptamer with BPA, there is a change in the LIG interdigitated electrodes' capacitance, which is measured to quantify the BPA present. The binding of BPA and aptamer is aided by alternating current electro-osmotic (ACEO) effect. This novel strategy of combining laser-directed 3D graphene as a transducer with the ACEO effect leads to an ultra-trace limit of detection of 58.28 aM within a short response time of 20 s (Cheng et al. 2016). Thus, LIG shows great improvement over conventional graphene and other nanomaterial-based sensors. Another LIG-based sensor for BPA detection was demonstrated using molecularly imprinted polymer (MIP) as the receptor. Polypyrrole was used as a monomer for the preparation of MIP on LIG. The sensor depicted a linear range of 0.05 to 20 µM and a detection limit of 8 nM (Beduk et al. 2020).

Hydrazine Hydrazine (N_2H_4) is a chemical of various applications in military, aerospace, pharmaceutical, energy, and industrial applications of fuels and chemical synthesis. It is also a neurotoxin and hazardous to human health, even at low concentrations. A sensor is shown to detect hydrazine using LIG as electrode material in a three-electrode system-based sensor. The limit of detection achieved was 70 µM, along with high selectivity for hydrazine against glucose,

formaldehyde, and ascorbic acid. Higher selectivity is possibly due to hydrazine's high electron transfer rate to the sensing electrode than other analytes (Sharma et al. 2020).

LIG-based sensors of non-environmental applications

The diversity of conventional and emerging environmental contaminants makes it imperative to study different analytes and their responses to LIG-based biosensors. These biosensors have a major focus on non-environmental applications of food safety and clinical diagnosis. The surrounding matrix and possible interfering substances are different for these applications, viz., the surface or bulk medium of food products or blood or sweat serums for clinical diagnosis. However, these biosensors may give an insight into the possible combinations of bioreceptors with LIG for pollutants of similar chemical nature. Hence, these LIG-based biosensors for non-environmental applications have been discussed further.

Sensors for food safety

Trans-resveratrol Trans-resveratrol (TRA) is a polyphenolic nutrient that acts as a quality indicator for grape wines. A sensor using LIG is demonstrated by a novel approach of a double layer of Kapton and polyimide tape to detect this polyphenolic nutrient. This setup maintained the stability of LIG as well as produced excellent electrical properties. The sensor achieved a linear response range of 0.2 to 50 $\mu molL^{-1}$ with a detection limit of 0.16 $\mu molL^{-1}$ (Zhang et al. 2020a).

α-Naphthalene acetic acid A sensor for the detection of α-naphthalene acetic acid (NAA) was developed using a nanohybrid material along with LIG. NAA is a phyto-regulator originating from agricultural activities with traces found in agro-products. Since NAA can absorb pollutants from the environment, it is important from the environmental as well as food safety perspective. In this work by Zhu et al. (2021), flexible nanohybrid material is used as an electrode along with LIG. The nanohybrid is composed of phosphorene (BP)—a graphene-like 2D nanomaterial—and titanium carbide MXene (Ti_3C_2-MXene). The linear range achieved by the sensor is 0.02 to 40 µM, and the limit of detection is 1.6 nM. The novel approach here is the combination of nanomaterials that behave as enzymes to catalyze electrochemical reactions of the analyte. Such nanomaterials are termed as "nanozymes." This strategy combines phosphorene and Ti_3C_2-MXene via noncovalent interactions, preventing the agglomeration of each nanomaterial and improving their electro-catalytical performance (Zhu et al. 2021).

Multiple analytes in fast food A wearable sensor for simultaneous detection of chloramphenicol (CAP), clenbuterol (CLB), and ractopamine (RAC) was developed with the application of the flexibility property of LIG electrodes. The sensor enabled on-site detection of these residual antibiotics and lean meat powder constituents from food samples. Differential pulse voltammetry (DPV) was used as the electrochemical technique in this sensor. In laboratory samples, the detection limits were 2.70 µM for CAP, 1.29 µM for CLB, and 7.81 µM for RAC, and the linear ranges were 10–200 µM, 5–80 µM, and 25–250 µM, respectively. For real samples in pork, the limit of detections was 20 µM, 10 µM, and 30 µM, while in milk, those were 10 µM, 5 µM, and 25 µM for CAP, CLB, and RAC, respectively (Li and Bo 2022).

Biogenic amines Biogenic amines are important markers for food quality. High levels of biogenic amines indicate microbial metabolism and endogenous enzymatic activity. These can be causes of food poisoning at high levels. Nanocarbon-based biosensors with graphene, carbon nanotubes, etc. are found to be most promising in the case of biogenic amines detection for food quality. Vanegas et al. (2018) demonstrated a biosensor for the detection of biogenic amines with LIG as an electrode that was modified with copper microparticles and diamine oxidase. The sensor's detection limit is 11.6 µM, with an average histamine sensitivity of 23.3 µA/mM and a fast response time of 7.3 s. The generation of electrical signals in the electrochemical biosensor results from the oxidation of the biogenic amines ($R\text{-}CH_2\text{-}NH_2$). The amines are oxidized to produce aldehydes (R-CHO), ammonia, and hydrogen peroxide. The peroxide is further oxidized spontaneously to generate electrons, resulting in corresponding detection and quantification of the biogenic amines (Vanegas et al. 2018).

Sensors for biomedical applications

Glucose A biosensor for glucose detection in sweat was fabricated with chemical modification of the LIG used as electrode material. LIG modification was carried out using acetic acid to reduce the carboxyl and hydroxyl groups on the LIG surface. This led to additional C–C bonds, which ultimately showed higher conductivity and a lower glucose detection limit. The oxidation of glucose was carried out by the enzyme glucose oxidase (GO_x). The generated electrochemical signals led to glucose measurement in sweat with a detection limit of 300 nM (Yoon et al. 2020). Non-enzymatic sensors have been explored as alternatives to avoid the complexities and stability issues posed by the use of enzymes in biosensors. A glucose sensor was developed using copper nanoparticles anchored on LIG, which achieved a detection limit of 0.39 µM (Zhang et al. 2020b). Another sensor with pulse-deposited copper nano-cubes on LIG achieved a limit

of detection of 250 nM for glucose which is comparable to the enzyme-based sensors. The non-enzymatic approach provides the advantages of simple fabrication and fouling-free operation (Tehrani and Bavarian 2016).

Urea Biosensors for detection of urea using LIG are reported based on two approaches by Mamleyev et al. (2019). The urease enzyme used for urea detection was either directly deposited on the graphene surface or on a layer of chitosan pre-deposited before urease immobilization. The amount of adsorption, activity of urease, and useful lifespan were much higher in the chitosan-pre-deposited graphene surface. This is due to the hydroxyl and amino groups that facilitate the covalent and electrostatic immobilization of the enzyme. The urease enzyme catalyzed the hydrolysis of urea into carbon dioxide and ammonia gas which was detected using a simple pH change measurement or by using transducers. This sensor has application in monitoring urea for kidney functioning in the human body (Mamleyev et al. 2019).

H_2O_2 A non-enzymatic flexible sensor was prepared using LIG decorated with silver nanoparticles for H_2O_2 detection. A linear range of 0.1 to 10 mM was shown by the sensor with a detection limit of 7.9 µM (Aparicio-Martínez et al. 2019). Another sensor for H_2O_2 detection was demonstrated that involved the use of platinum nanoparticles (PtNPs) loaded on LIG. This sensor achieved a further lower limit of detection of 0.1 µM (Zhang et al. 2019). A similar approach was used for sensing H_2O_2 but using bimetallic nanoparticles of copper and ruthenium which allowed led to a limit of detection of 1.8 µM (Thirumalai et al. 2021).

Dopamine Dopamine, a neurotransmitter, i.e., a chemical messenger in the body, is of interest for the diagnosis of diseases such as Alzheimer's and Parkinson's disease. A sensor for the detection of dopamine has been reported with the electrode material as LIG decorated with platinum nanoparticles which demonstrated a detection limit of 70 µM (Nayak et al. 2016). Another sensor was fabricated using LIG modified by PEDOT—poly(3,4-ethylenedioxythiophene). The limit of detection achieved by the sensor in the detection of dopamine is 0.33 µM. Selectivity of the sensor against interference by ascorbic acid (AA) and uric acid (UA) is achieved in this sensor (Xu et al. 2018). Further improvement in the detection of dopamine was achieved using LIG modified with Pt-Au nanoparticles. The sensor was fabricated on a polydimethylsiloxane (PDMS) substrate. The PDMS substrate imparted better stability to the sensor as compared to the polyimide substrate used in previously developed sensors. The limit of detection achieved using the Pt-Au combination nanoparticles is 75 nM (Hui et al. 2019). Furthermore, a comparative study on biosensors for dopamine detection using two types of LIG was done (Santos et al. 2021).

Thrombin Thrombin is a two-polypeptide chain, a glycosylated serine protease. It is an essential enzyme in human physiology responsible for blood clotting. It is derived from its precursor prothrombin and is a biomarker for different diseases such as pulmonary metastasis. As thrombin concentration increases, the risk of its associated disease increases. Yagati et al. demonstrated a biosensor with the use of aptamers specific to thrombin. This biosensor with LIG as capacitive transducer material showed a limit of detection of 0.12 pM in laboratory samples and 1.3 pM in serum (Yagati et al. 2020). Another biosensor with a similar approach of aptamer as bioreceptor was fabricated by Fenzl et al. (2017). The aptamer is bonded to the LIG surface using 1-pyrenebutyric acid for selective detection of thrombin. The limit of detection achieved is 1 pM in buffer and 5 pM in complex serum matrix (Fenzl et al. 2017).

Other sensors The miRNA or microRNA is a biomolecule of importance in biomedical application for diagnosing cancers and other diseases. A sensor for miRNA detection using LIG was demonstrated with a detection limit of 10 fM. In this case, sample processing is required since the miRNA has to be adsorbed into the graphene surface for detection. The separation of preeclampsia-specific miRNA hsa-miR-486-5p is carried out by magnetic separation, and detection is carried out by electrochemical interaction with the solution phase redox marker $[Fe(CN)_6]^{3-/4-}$ (Wan et al. 2020b). The development of an antibody-based sensor was done for the detection of carcinoembryonic antigen (CEA), an important tumor biomarker for colorectal cancer. The LIG material used in this sensor was doped with gold nanoparticles and further functionalized with -COOH groups for covalent binding of the antibody specific to CEA. The sensor displayed high sensitivity in CEA detection with a linear range of 0.01 to 100 ng/ml and a very low detection limit of 5 pg/ml (Wang et al. 2021). Paracetamol is a common medicine used as a painkiller for a variety of health issues. During the preparation of pharmaceutical drugs, it is important to monitor and control the components. Detection of paracetamol has been achieved using a LIG-based sensor and compared with conventional screen-printed carbon electrodes (SPCE). The LIG sensor showed highly improved electrochemical performance as compared to SPCE, with a linear range of 0.1 to 10 µM and a limit of detection of 31 nM (Ghanam et al. 2020). A point-of-use application sensor for hydration levels using LIG was developed to detect ammonium (NH_4^+) and potassium (K^+) ions in sweat. The use of ion-selective electrodes (ISEs) was done, leading to the detection limits of 30 µM and 100 µM for NH_4^+ and K^+, respectively (Kucherenko et al. 2020). The strategy of ion-selective electrodes with LIG has also been successfully applied for sensing of K^+ and H^+ ions in urine samples as a biomedical application as well as from a future environmental perspective.

This sensor showed a linear range of 0.3 to 150 mM for K^+ and 5.0 to 8.0 pH for H^+ concentration (Kucherenko et al. 2021). Conventional ISEs employ liquid contact electrolytes between the electrolyte and an ion-selective membrane creating problems such as evaporation and leaching of electrolyte. This could lead to unreliable signal strength as well as possible failure of the sensor. Solid-contact ISEs were employed in the above-mentioned two works to stabilize the sensor and prevent the leaching of the liquid electrolyte.

Potential for LIG-based electrochemical sensors for environmental application

As per literature, a diversified inference can be drawn related to the future of electrochemical detection of pollutants for environmental monitoring. Environmental analytes consist of a wide range of chemicals and biomolecules. These are present in complex matrices of natural streams, soil, air, etc., thus exposing human beings and other living organisms. Hence, for effective sensing of these analytes, categories of analytes with a specific approach for detection have to be established (Zhang et al. 2021b). Selectivity of detection is of utmost importance due to the complex nature of the matrices in which these analytes exist. Also, a variety of LIG-based sensors for non-environmental applications have been explored in the previous section. From these different possible combinations of nanomaterials, bioreceptors and mechanisms for detection can be understood. Similar combinations may be used for environmental pollutants depending upon their feasibility of electrochemical signal generation. Different types of bioreceptor, viz., aptamer, antibody, MIP, enzyme, or even bioreceptor-free sensing, can specifically target certain classes of pollutants. Hence, the approach of exploring these possibilities based on different classes of bioreceptors seems most relevant. A summary of combinations of nanomaterials and bioreceptors with LIG for existing biosensors and potential biosensors for environmental applications is depicted in Fig. 4.

Aptamer-based Aptamers have been used as biorecognition elements for highly selective detection of analytes such as bisphenol A and thrombin. The affinity of bisphenol A for aptamers depicts that the phenolic pollutants can be explored to show an affinity for specifically engineered aptamers. As thrombin being a protein biomolecule, the affinity of aptamers specifically engineered for biomolecule detection can be explored (Kong and Byun 2013). This may help in quantifying the organic matter present in waste samples. Due to the selectivity offered by aptamers, particular disease-causing pathogenic organisms or indicator organisms may also be detected with this approach. Aptamers are known to show a higher affinity for the microorganisms as compared to antibodies due to their smaller size (Yagati et al. 2020). Thus, they can be a viable option for improving the sensitivity of sensors for living and inactive microorganisms. Heavy metals like arsenic, lead, and copper have been shown to be detected using LIG-based aptasensors (Cui et al. 2016; Ensafi et al. 2018; Uda et al. 2021; Zhang et al. 2021a). The systematic evolution of ligands by exponential enrichment (SELEX) technique is commonly used for the aptamer selection for these sensors (Ellington and Szostak 1990; Tuerk and Gold 1990). Many modifications of the SELEX method have also been reported for the selection of a highly selective aptamer (Song et al. 2012). Further improvement in the performance of these sensors can be explored with the use of aptamers in combination with electrocatalyst metal nanoparticles.

Antibody-based Detection of pathogens or pathogen indicators like *E. coli* and *Salmonella enterica* has been conventionally achieved with antibodies specific to these microorganisms. Metal nanoparticles have shown improvement in the sensing performance as in the case of *E. coli*. Use of LIG for detection of other disease-causing pathogens can be potentially explored. Selectivity against a particular pathogen can be achieved with antibodies or other receptors like aptamers specific to that particular organism. The challenge for such a development would be the binding of antibodies on LIG surface. Efficient binding performance has to be explored for the effectiveness of such biosensors. Suitable binding molecules may also need to be explored for the same.

MIP-based Molecularly imprinted polymer technology (MIP) has also gathered attention recently to improve the selectivity of sensors. For example, MIP has been used successfully for selective detection of the antibiotic chloramphenicol in one study. Another research of MIP-based biosensors using LIG has been applied for dual detection of amoxicillin and ascorbic acid. As in the case of aptamers, this method of detection of analytes is highly selective due to patterned sites created for the analyte. Graphene-based sensors with conventional reduced graphene oxide form and MIP as a bioreceptor have been employed for pollutants such as 4-nitrophenol (Zeng et al. 2014) and ractopamine (Li et al. 2018). LIG may be able to improve further the analytical performance in the detection of these analytes.

Enzyme-based Detection of certain pollutants like urea is carried out through enzymatic actions specific to the chemical, viz., urease in this particular case. This is another method of selective detection of the analyte. Enzymes are also used to detect glucose, hydrogen peroxide, and dopamine biomarker. A similar oxidation phenomenon by enzymatic action can be explored for organic pollutants such as

phenolic compounds, parabens, and polycyclic aromatic hydrocarbons (PAH) for their selective detection using this approach. The challenge is to find suitable enzymes that may be able to oxidize or reduce the organic compounds. Enzyme inhibition actions have been extensively used in the electrochemical detection of pollutants using graphene in the form of reduced graphene oxide. The inhibition of the acetylcholinesterase enzyme has been a popular technique for the detection of organophosphorus pesticides (Itsoponpan et al. 2021). A similar approach can be employed where pollutants inhibit the action of a particular enzyme. For example, heavy metal arsenic is known to inhibit the action of cholesterol oxidase (Duoc et al. 2020). These pollutants may be organic or inorganic in nature, as per the existing literature on graphene-based enzymatic electrochemical biosensors. LIG as an electrode material can help in the improvement of analytical features for such sensors.

Bioreceptor-free sensors Non-enzymatic glucose sensors and hydrogen peroxide sensors involve the use of metal nanoparticles. These nanoparticles catalyze the electrochemical oxidation/reduction of the analyte. Nanoparticles such as Ag, Au, Pt, and Cu have shown such electroanalytical behavior along with LIG. The use of LIG without the use of receptors has also been done for the detection of heavy metals, as in the case of lead (Pb^{2+}) (Grossman et al. 2020). Such a type of electrochemical sensing requires the application of a suitable electrochemical technique for a particular analyte. As in the case of lead, the anodic stripping voltammetry technique has been employed, which can be favorable for metal analytes due to the inherent electroplating mechanism used. Similar bioreceptor-free approaches have also been used for non-metallic pollutants with reduced graphene oxide as the electrode. The bioreceptor-free approach has been used in the sensing of miRNA (Wan et al. 2020b), hydrazine (Sharma et al. 2020), trans-resveratrol (Zhang et al. 2020a), and α-naphthalene acetic acid (Zhu et al. 2021) without using a receptor and transduction layer separately. This offers a distinct advantage of a simple and fast fabrication process due to the elimination of complex chemical steps in immobilizing the bioreceptor. The analytes are directly adsorbed onto the LIG or LIG modified with other nanomaterials. The techniques used for electrochemical signal generation include differential pulse voltammetry (DPV), cyclic voltammetry (CV), linear sweep voltammetry (LSV), etc. miRNAs are nucleic acids of biomedical application, hydrazine is a chemical of hazardous nature, while TRA is a polyphenolic compound and NAA is a phyto-regulator. Thus, the application of the bioreceptor-free technique in LIG-based sensors can be widespread. The essential requirement for such electrochemical sensors is in an environmental context to identify the correct electrochemical technique and suitable conditions in the sample matrix, such as pH, for generating the signal. The selection of proper precursor LIG materials, viz., polyimide (PI) and polyethersulfone (PES), is important in order to generate proper electrochemical signals

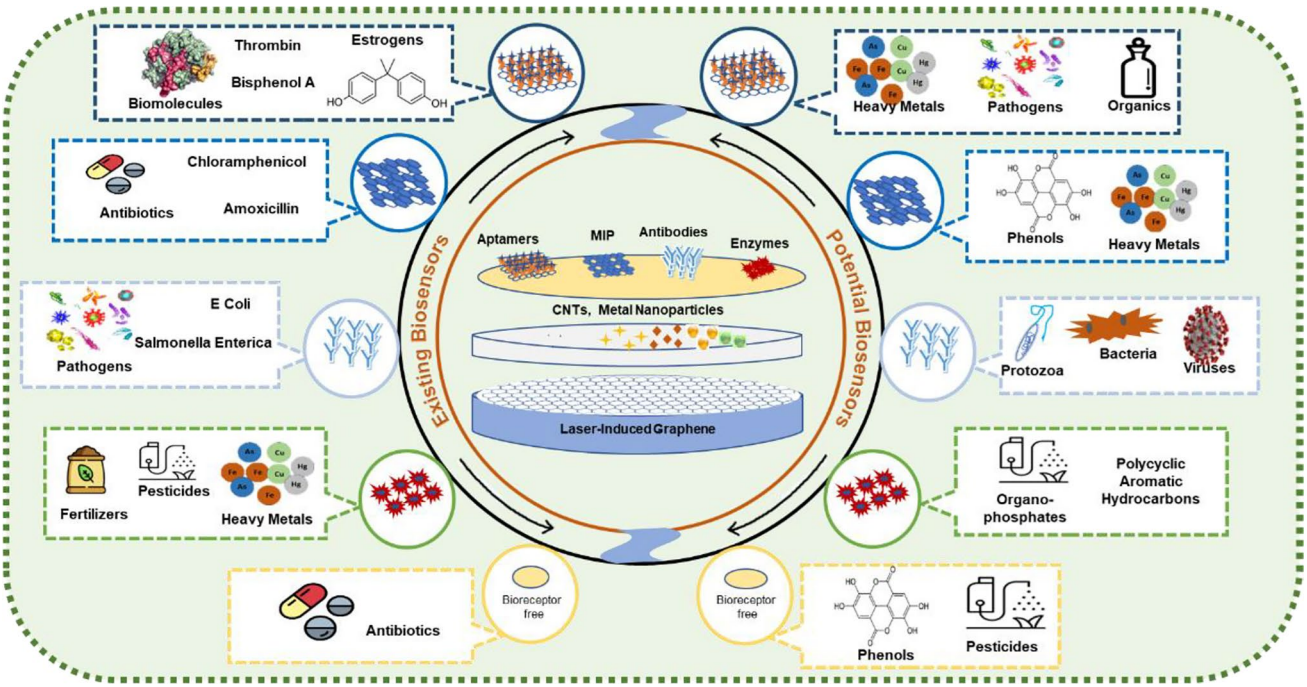

Fig. 4 Combinations of LIG, other nanomaterials, and bioreceptors for existing biosensors and potential biosensor development for various environmental analyte

(Grossman et al. 2020). Based on the nature of the species to be detected, the inherent residual elements/functional groups may enhance or reduce the signal.

LIG-based sensors—strengths, weaknesses, opportunities, and threats analysis

It is important to evaluate the different aspects related to long-term technical and financial viability for the effective incorporation of LIG-based sensors as a mainstream option for electrochemical sensing (Wan et al. 2020a). The feasibility of widespread use of biosensors has various factors, including the cost of precursor chemicals and raw materials, number of preparation steps, intermediate chemicals/materials required, post-fabrication treatment, energy requirement, the time required for fabrication, cost of instrument for fabrication and/or man-hours required for fabrication per unit, lifespan or durability of the sensor, and disposal cost. Apart from these direct costs, the scale of production, marketability, and profitability are important factors that can impact the financial viability of biosensors. Strengths, weaknesses, opportunities, and threats (SWOT) analysis has been done for LIG-based sensors to wholistically project their potential and challenges in incorporating these sensors into the mainstream market (Fig. 5).

Strengths The conventional techniques of graphene fabrication, such as chemical vapor deposition (CVD), exfoliation, and chemical reduction, are highly chemically intensive (Novoselov et al. 2012; Wan et al. 2020a). In contrast, the laser induction process for graphene fabrication requires no chemical treatment (Stefano et al. 2021). Therefore, LIG offers a simple, production-friendly alternative with a smaller footprint in terms of chemical consumption for electrochemical sensors. Furthermore, the conventional chemical-based methods of graphene synthesis involve direct human effort in fabrication steps that substantially increase the time required, and the requirement of skilled resources for complex chemical fabrication is also expensive. In comparison, the instrument-operated fabrication of LIG is fast and gives a more stable as well as the consistent product.

Weaknesses LIG fabrication is devoid of chemically intensive processes, but the initial capital investment may be higher due to instrumentation cost as compared to instrument-free techniques such as exfoliation and reduction. However, this cost may be easily traded off by the chemical-free approach for LIG, which saves material and manpower costs as well as time. A comprehensive evaluation of life cycle cost is required to establish the viability of LIG against conventional methods. Another potential weakness in the usability of LIG-based sensors pertains to the lifespan of the biosensor, which depends upon the strength and stability of the electrode material (Kaidarova and Kosel 2021). The number of times the sensor can be reliably used ultimately defines its cost per usage. Due to the porous structure of LIG, the electrode surfaces are more susceptible to scraping and damage. Efforts are required to make LIG sensors more robust and durable so that performance reliability over cost is increased.

Opportunities In one of the studies on the market size and cost evaluation of biosensors, they are classified as single-use, intermittently used, and continuous use biosensors (Sadana 2005). In this study, the cost vs. data rate is found to be very high for single-use sensors. LIG-based sensors have the potential to contribute to single-use type disposable sensors due to their low fabrication cost and easy usage with miniaturized technology (Tehrani and Bavarian 2016). The advantage of LIG providing better electrochemical performance as well as an automated, scalable fabrication process may lead to significantly reduced costs while maintaining the reliability of data from the sensors. Although biosensors themselves are intended for rapid detection of analytes, the time required in production, supply, and usage at the final point of use is also an important factor in its financial viability. A rapid and industry scalable process for production, as in the case of LIG, provides the advantage of reduced time and hence better financial viability in this aspect. The improved sensitivity and selectivity of sensors due to the porous nature of graphene in LIG can allow for trace-level selective sensing of pollutants (Cheng et al. 2016; Nasraoui et al. 2021). Thus, LIG-based electrochemical sensors have the potential to be integrated with environmental systems and improve their performances significantly.

Threats Fouling of sensors, especially in the case of receptor-based sensors, is a major issue for electrochemical sensors (Lin and Li 2020; Russo et al. 2021). But this issue is relevant to all the types of electrochemical biosensors as it concerns the type of analyte to be detected and other species in the sample matrix that can deposit on the electrode surface. Strategies for LIG-based biosensor development with minimal fouling due to the receptor and deposition of analytes are essential to avoid this threat in the progress of these biosensors. The disposal of electronic wastes post usage has been a long-standing issue of concern for environmental sustainability. The leaching of heavy metals and other residues in used graphene-based sensors poses environmental risks that need to be addressed for a sustainable implementation of LIG-based electrochemical sensor application (Tao et al. 2021). Efforts are required to recycle the used materials and mechanisms to channel these wastes through proper waste recycling streams. This issue pertains to all types of biosensors, whether single-use or multi-use, such as carbon-based and glassy carbon-based biosensors apart from LIG. A life cycle impact assessment of these different biosensor alternatives can give further clarity on the sustainability parameter

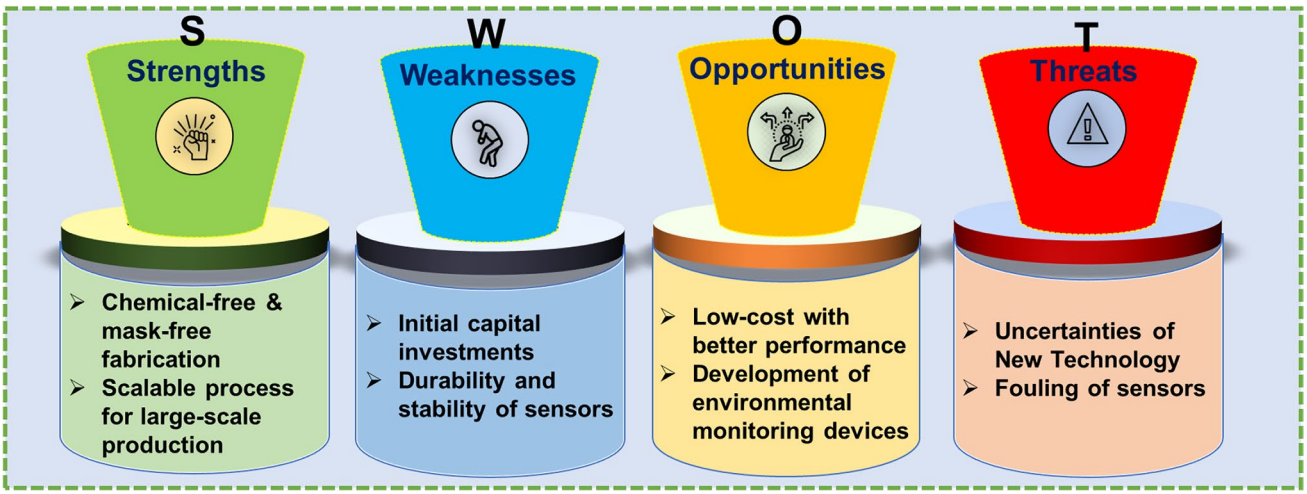

Fig. 5 Strengths, weaknesses, opportunities, and threats in incorporating LIG-based electrochemical biosensors for environmental applications

for LIG compared to other biosensors (Moro et al. 2019). Toxicology studies are also crucial for incorporating LIG as a mainstream electrochemical biosensor alternative. Graphene family nanomaterials are known to exhibit some in vivo and in vitro toxic effects due to the morphology, presence of functional groups, and other physicochemical characteristics (Tonelli et al. 2015). Laser induction of graphene is a relatively new technology with uncertainties which is also a threat to the upscaling of LIG-based biosensors. Detailed studies are required to establish the impact of LIG on exposure to human health and the environment, to establish the reliability of this technology.

Conclusion

A wide variety of LIG-based sensors have been analyzed in this study. From the reviewed literature, it can be inferred that the advent of LIG has shown potential for the improvement of electrochemical sensors. To impart selectivity of the biosensors, the use of receptors like aptamers, molecularly imprinted polymers (MIP), enzymes, and antibodies is important. At the same time, even bioreceptor-free sensors can be designed, which provide a simple and easily scalable solution. Since most proposed sensors are laboratory scale, the performance of the sensors in complex matrices is of concern moving forward. These matrices are a common phenomenon occurring in nature, and hence, selectivity is a major requirement in sensor development. Nanoparticles and their combinations have also shown improvements in sensor performances via increasing the catalytic activity on the surface of the LIG electrodes. This is attributed to the nanomaterials' electrochemical activity, which simplifies the fabrication and imparts stability to the sensors.

Based on the classes of analytes reviewed, examples of biosensor development have been studied for the analytes of importance for environmental monitoring. These include antibiotics like chloramphenicol, amoxicillin, phyto-regulators like α-naphthalene acetic acid, pathogens, estrogens, ions, and heavy metals. A major focus on biomedical as well as food safety monitoring sensors is observed through the class of analytes. An organized approach to the development of sensors for environmental monitoring can also be defined through the sensing mechanisms and materials used for these different classes of analytes. This approach majorly revolves around different classes of bioreceptors to be used for specific classes of pollutants. Since the type of analytes for environmental monitoring is also diversified, the sensor developments for different analytes also require special approaches. Scope for the common multifunctional sensors is also derived due to the requirement for detecting multiple pollutants and quality markers in a single sample. LIG provides a simple, low-cost, performance-improving option for graphene-based sensors and biosensors. The active research for LIG-based biosensors will lead to low-cost and effective solutions for environmental sensing. The strengths, weaknesses, opportunities, and threats analysis also highlights the different aspects that are important to address to incorporate LIG-based electrochemical sensors as a viable option for environmental applications.

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Declarations

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Competing interests The authors declare no competing interests.

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