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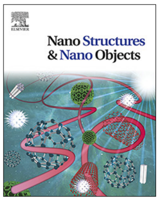

Recent advances in bioinspired sustainable sensing technologies

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ABSTRACT

Mimicking the innovative structure, design and multifunctional mechanisms of biological process based sustainable biosensor technology in living creatures is highly successful with multilingual features that draw in constantly more and more research efforts and extensive attention from the numerous species of diagnosis and clinical origins. This review will provide a comprehensive overview with respect to the innovative approach for identify, recognizing and showcasing the current advancements and key milestones accomplished in this field, aiming to provide an exclusive outlook for the bioderived materials and their most relevant applications to encourage readers to explore the uncharted realms of sustainable biointerfaces in the last 5 years.

Contents

1. Introduction

Nature has advanced potential to be a source of materials derived from biological samples with bio mimicking and bioinspired approaches. Nature-derived materials are appealing because of their sustainability, biodegradability, conformability, biorecognition, self-repair and stimuli response qualities. To provide ideal sustainability, the material/energy consumption rates should equal the rate of resource regeneration and the material should be biodegradable. The amazing efficiency of biological matter, such as their outstanding qualities based on weak ingredients, high performance per unit mass and various functions, in addition to mechanical capabilities, has been assumed to be related to their hierarchical organization. Especially, the self-repair

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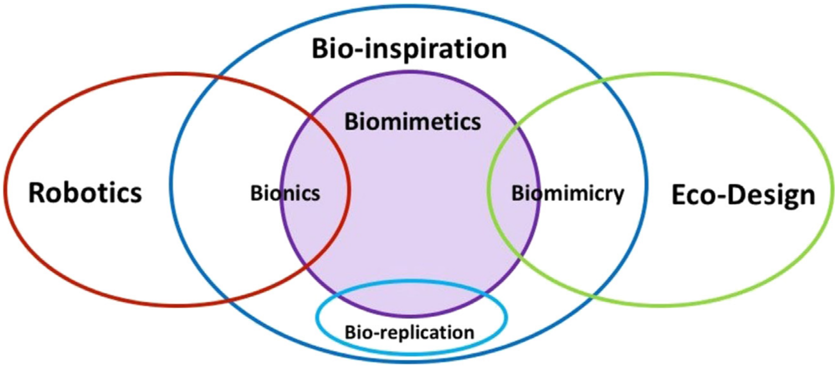


Fig. 1. Important concept boundaries in terms of bioinspiration reproduced from the open-access journal [9].

process occurs in living organisms due to a result of biological evolution. To provide self-repaired materials, biomimicking must be completed successfully. Technical applications usually need a broad multidisciplinary collaboration of basic scientists, designers, architects and engineers [1–4]. Here, the definition of common terms as should be highlighted.

Bioinspiration is an approach that recommends new topics for research by utilizing biological events to drive research in non-biological science and technology [5].

Bio-based materials, also known as biopolymers or bio-derived materials, are artificial materials generated entirely or partially from biological matter [6].

Biomimetic materials are defined as synthetic materials that can mimic the biological items that we encounter in our daily lives [7]. The ISO-standard defines biomimetics as "interdisciplinary cooperation of biology and technology or other fields of innovation to solve practical problems through functional analysis of biological systems, abstraction into models, and transfer into and application of these models to the solution of practical problems" [8].

All definitions are very similar and they are used in combination or instead of each other in the literature. There are also other terms such as bionic and bio-replication.

Bionics is defined as a "technical field that tries to imitate, augment or replace biological capabilities with electrical and/or mechanical counterparts". The direct reproduction of a biological structure to accomplish at least one specified capability is referred to as ***bio-replication*** [9]. These terms' similarities, differences and concurrences are represented with a Venn diagram in Fig. 1.

Plants, animals and microorganisms could be the inspirational source of biobased materials [10–12]. Plant or animal-based materials can be mainly exemplified as sericin, fibroin, chitin, chitosan, keratin, cellulose, agarose, alginate, collagen, melanin, carotenoid and indigo [2]. They are not limited only to these examples. Recent developments in nanotechnology, as well as the rapid expansion of additive manufacturing, have bolstered bioderived material research, opening up new avenues for incorporating natural traits in de novo bioinspired materials. Drawing inspiration from nature can be a viable method in the search for damage-tolerant materials that combine toughness and strength. For this purpose bamboo, tortoise shells, wood and wood-like materials, mollusk shell, mineralized tissue and hydroxyapatite could be good alternatives of nature [13].

Several types of bioderived/bioinspired materials are great candidates for biosensor development processes. They could open new windows for developing sensing applications in terms of superior properties, such as biocompatibility, conductivity, electroactivity, flexibility, biodegradability and implantability [14–17]. Except for traditional electrochemical or optic biosensors, they can be utilized for smart and novel technologies such as DNA machines, point of care diagnostics, micro/nanorobots, etc. [18].

Although different terminology is very common, to understand the tendency of bioinspired/bioderived materials in sensing technologies, Web of Science indexed article numbers can be used. Regarding the literature search from the Web of Science at 19 March 2022 with a keyword of "inspired biosensor", there are a total of 722 publications, 502 of them are research and 158 of them are review articles. In Fig. 2, last 10 years' data can be seen. Published articles have accelerated, especially after 2018.

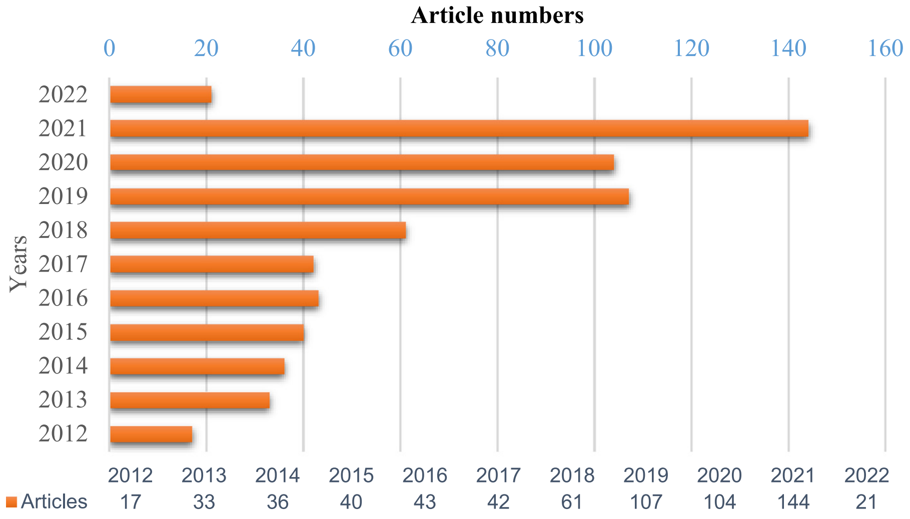
As per the understanding from Fig. 2, many studies on bio-inspired multifunctional intelligent materials have been conducted over the last several decades to build biosensing devices and enhance their overall performance with increased sensitivity, stability, linearity, repeatability, adaptability and selectivity. Two basic techniques drive current bioinspired/bioderived sensing technologies. To create smart bio-inspired sensors, the first technique is connected to structural design and the second is based on surface functional processes, or a mix of both [19]. This review aims to provide a comprehensive overview concerning the innovative approach for identifying, recognizing and showcasing the current advancements and key milestones accomplished in biosensing technologies based on bioinspired/bioderived materials, aiming to provide an exclusive outlook for their most relevant applications to encourage readers in the exploration into the uncharted realms of sustainable biological interfaces in last 5 years.

2. Classification of biosensor technology and their sustainable species

The analytical platforms that help in the conversion of biological stimuli into electrical signals that can be measured are called biosensors. Biosensors are categorized based on their sensing elements and transduction mode. They offer specificity, reusability, cost effectiveness, reliability and biocompatibility. There is a very wide range of biosensors that are being employed in multiple sectors, ranging from medical to environmental studies. Biosensors extend as a remarkable alternative to conventional methods which rely upon expensive and time-consuming strategies, as they combine physiochemical living characteristics with a real-time analytical approach to provide better solutions for a variety of applications. The following fields in Fig. 3 show the wide employment of biosensors for multiple applications.

2.1. Drug delivery

Modern drug delivery systems utilize microtubules, nanoparticles and responsive polymers to control the profile of drug release in patients that are too susceptible to diseases. These delivery



	2012	2013	2014	2015	2016	2017	2018	2019	2020	2021	2022
■ Articles	17	33	36	40	43	42	61	107	104	144	21

Fig. 2. WoS indexed articles related to inspired biosensors.

Fig. 3. Applications of bioderived sensors in several fields of specialization [20].

systems are focused on controlling the drug release and their degradation, along with their biocompatible nature. This design of drug delivery system provides increased efficiency, a therapeutic index, patient compliance, bioavailability, pharmacodynamics and pharmacokinetics, along with very low associated side effects [21,22]. A novel molecule, which is an oligonucleotide with the shape of hairpin, was recently discovered and termed as a molecular beacon [23,24]. Molecular beacons are labeled with both a fluorescent dye and a quencher. Due to their high specificity, they can be administered in a cell by different mechanisms for the probing of cancer biomarkers [25]. Another unique nanostructure based on DNA had a tetrahedral architecture and was utilized for drug loading application because of its remarkable mechanical characteristics and functionalities [26]. Another potential candidate for drug loading in theranostics application consists of the hollow nanoparticles of a Mn_3O_4 based biosensor, as this material is characterized with an interior void [27]. Even nanogel based biosensors are a great candidates for drug delivery as they

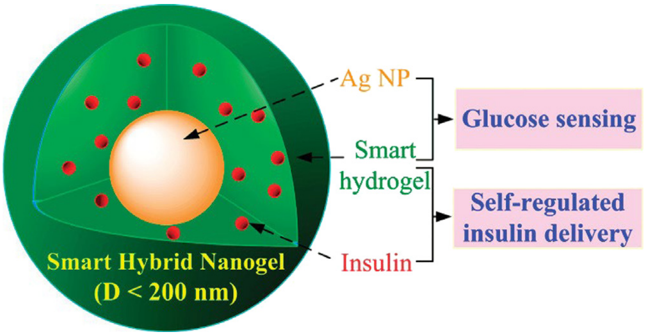


Fig. 4. Smart hybrid nanogels, shown schematically, can combine self-controlled insulin delivery and optical glucose detection at physiological pH and temperature in a single nano-object [30].

offer the advantages of both hydrogels and nanoparticles [28,29]. Nanogel delivery, that is used for drug loading (Fig. 4), is based on temperature, glucose concentration and pH [30].

For the therapy of diabetes patients, insulin delivery is paramount, for which glucose sensitive nanogels are utilized as they are highly sensitive towards glucose [31]. Micro-/nanomotors have emerged as potent, smart and self-propelled systems for drug delivery [32]. Biosensor based on MEMS and NEMS (nanoelectromechanical systems) have been developed as wearables and implantable devices for the intravenous delivery of drugs. The miniaturization and sensitivity of biosensors pose a great solution for the drug delivery [33–35].

2.2. Food

Food product management and its associated quality management require cost effective, real time and rapid techniques. These characteristics are offered by biosensors, in comparison to conventional spectroscopic methods [36]. Potentiometric alternating biosensors have been developed for the detection of *E. coli*, which is a potential pathogen in vegetables. The biosensor was based on a conjugate of urease and *E. coli* antibody. This sensor identifies the presence of bacteria by the detection of variations in pH due to ammonia generation [37]. A vital substrate and target in the fermentation industry (a large sector in the food industry) is sugar. Thus, a large number of biosensors have been developed to detect its level in a certain fermentation process [38]. A glucose oxidase-based screen-printed electrode biosensor for glucose was utilized to control the fermentation process in real time. This biosensor offered enhanced reproducibility and suitability towards large-scale production in a cost-effective way [39]. Another biosensing platform was designed in which the base electrode was not directly attached to the glucose oxidase enzyme, rather it measured the catalytic product formed by the enzymatic action, to facilitate on-line biosensing in the fermentation process [40]. Electrochemical impedance spectroscopy-based biosensors were developed and utilized on a large scale to aid food production in a sustainable manner. These biosensors utilized AC voltages of low amplitudes to measure the current in terms of frequency [41]. In gluten-based food, like wheat, rye and barley, an alcohol soluble fraction was derived, which is termed as a gliadin. These compounds can cause celiac disease, since they act as food allergens. Thus, gluten-free food is considered as the therapeutic measure for patients afflicted with celiac disease. Thus, a porous optical biosensor based on silicon was developed by employing a glutamine-binding protein derived from *Escherichia coli*, as this protein binds with the gliadin peptide, specifically, as found in wheat [42]. Another gliadin measuring biosensor was fabricated, which comes under the category of an electrochemical immunosensor. It shows a limit of detection of the order of µg/L, as presented in Fig. 5 [43].

For the rapid pre-screening of food for herbicides, biosensors offer a wide array of advantages, such as low limit of detection and cost effectiveness, along with high efficiency. Various biosensors which utilize a chemiluminescence based immunoassay, a molecular imprint-based chemo sensor and surface-enhanced Raman spectroscopy were fabricated for herbicide screening [44–46]. Biosensors have provided improved diagnosis in the food industry in terms of quality control, testing of materials with authentic and traceable results. Thus, they are ideal, reliable and cost-effective candidates to ensure food safety and quality.

2.3. Pharmaceuticals

A wide array of organic compounds which are effective at very small concentrations are categorized under pharmaceuticals. They have very wide usage in theranostics. Due to their uncontrolled excessive usage, they pose a new threat to human health and the environment. Thus, their detection becomes paramount in overcoming these problems on human health due to their usage. Biosensors offers a sensitive, effective, inexpensive, rapid, high-capacity real-time solution for this problem. Electrochemical detection of cocaine, one of the prominent pharmaceutical ingredients, has been done by a platinum nanoparticle based amperometric biosensor, which exhibited 1×10^{-5} LOD. This biosensor was an aptasensor which also utilized anticocaine aptamer functionalized gold support and platinum nanoparticles [47].

A composite based immunosensor was fabricated for the detection of progesterone, a component of contraceptive pills. This biosensor employed a composite of gold, graphite and Teflon functionalized with tyrosinase for the amperometric detection of progesterone [49]. Another hormone, cortisol, which is a metabolic regulator, was detected by an amperometric biosensor that employs a three-electrode system, in which one of them functionalizes AuNWs with cortisol specific antibodies [50]. For the detection of clozapine, which belongs to the dibenzodiazepine class of drugs, a biosensor based on magnetic nanoparticles was fabricated. Fig. 6 represents the biosensor amperometrically studies, the peroxidation of the drug, which may impart hazardous impact on human health [48].

Another biosensor based on microfluidics was developed to study the effect of an anticancer drug on the HL-60 cell line of leukemia. This biosensor was based on utilizing probes for the detection of apoptosis induced by the drug. The probes were made from annexin V-conjugated QDs [51]. For the automated detection of antibiotics, such as macrolides and penicillin etc. in milk samples, a parallel affinity sensor array-based biosensor was developed. This sensor was quite sensitive, effective and rapid in nature [52]. As biosensors are providing a portable, effective and continuous monitoring of pharmaceutical products in a cost-effective manner, their continuous development is required to develop enhanced flexible analytical systems for the pharmaceutical industry.

2.4. Medical

For the diagnosis of diseases and the observation of physiological changes, biosensing is used internationally in health sciences and biomedical research. Sensitive measurements of physiological changes, such as temperature, pH, pressure, insulin and glucose levels, oxygen levels and ion concentrations in biological specimens, can be made using special fluorescence-based biosensing. A biosensor can monitor changes in the biological

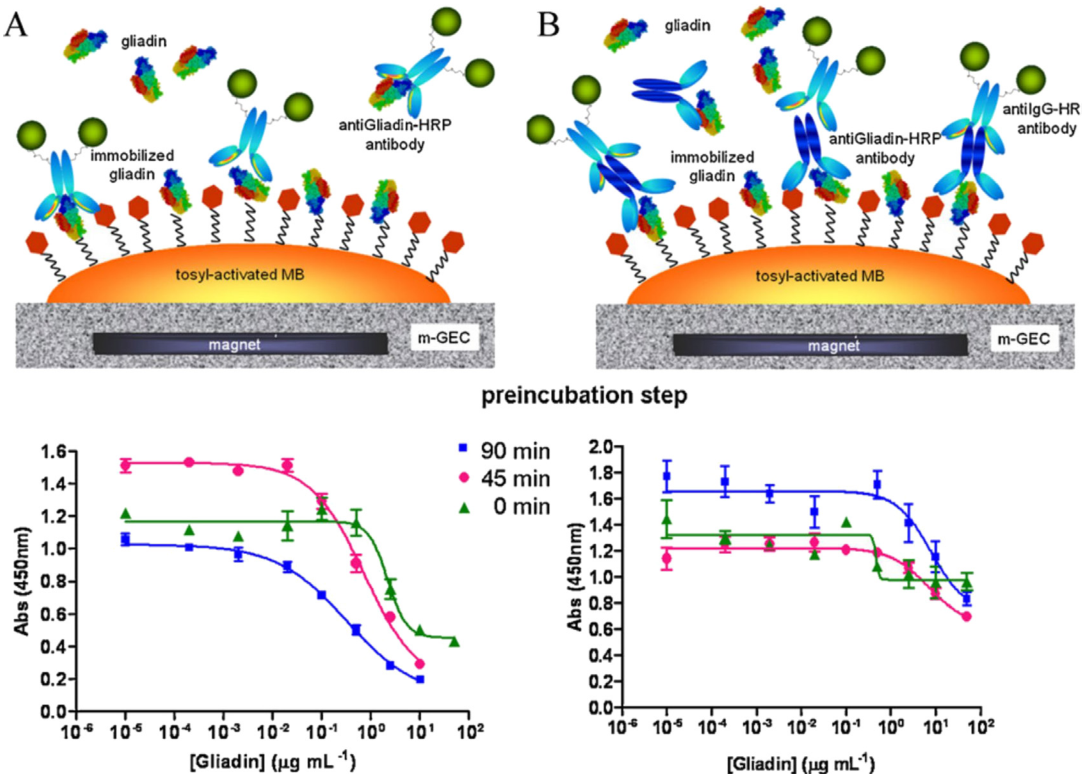


Fig. 5. Diagram showing the determination of gliadin using (A) direct and (B) indirect competitive magneto-immunoassays. Additionally, comparative outcomes from both immunological techniques that involved preincubating gliadin (from 0 to 90 min) with the antiGliadin primary antibody are displayed [43].

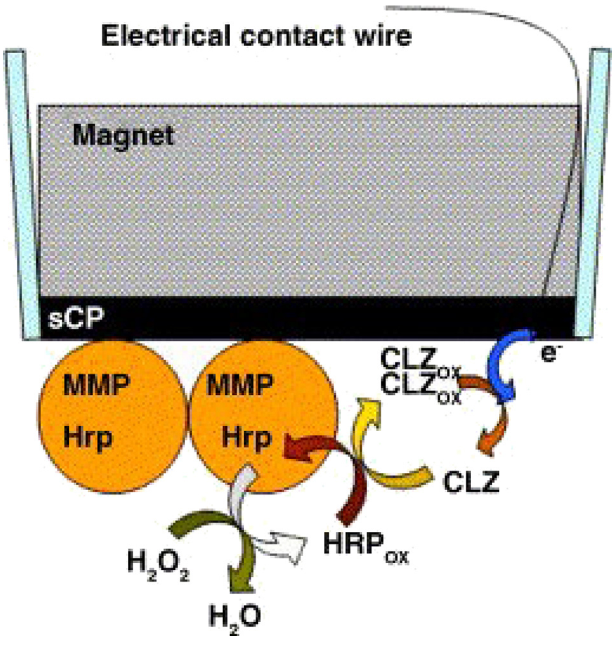


Fig. 6. Diagram showing the electroreduction of clozapine after it has been peroxided at horseradish peroxidase-immobilized magnetic microparticles (HRP-MMPs) (sCPE) [48].

system being studied by measuring the fluorescence intensity or wavelength, some of which may reach levels that are considered abnormal or unhealthy. The biosensors are typically examined in *ex vivo* cells and tissues or in biological fluids that have been collected, like blood or urine [53,54].

Khalid et al. reported using a silk fibroin coating on optical fibers for encasing fluorescent sensor molecules to overcome these restrictions and enable *in vivo* biosensing. An extremely thin layer of silk, a naturally generated biopolymer made completely of proteins and amino acids, was applied to the silica exposed core fiber (ECF) samples, as shown in Fig. 7. The fluorophore 5,6-carboxynapthofluorescein (CNF), which enables optical pH detection by a reliable ratiometric fluorescence technique, was doped into the silk [55].

2.5. Environmental monitoring

The monitoring of pollutants, toxicants and pathogens in the environment has become an immense need in today's world to overcome the harmful implications of environmental issues on human health. The chromatographic technique had served as the traditional method for the detection of environmental pollutants for a long time, but due to its time-consuming process and the expensive nature of the raw material and equipment, it was not well-suited. Thus, to overcome these problems, biosensors provide a cost-effective, easily operable, effective, rapid, sensitive and reproducible approach for a sustainable system to detect these pollutants and make a better environment for the survival of human beings. Pesticides, being the most used hazardous environmental pollutants, require sensitive detection as they impart multiple health threats. An enzymatic biosensor based on amperometric measurement utilizing a cysteamine self-assembled monolayer on a screen-printed electrode of gold was fabricated for the detection of Paraoxon, an organophosphorus insecticide. This biosensor, Fig. 8, is disposable and provides a LOD of 40 ppb.

Another biosensor was fabricated for real environmental samples from soil and water to detect acetamiprid. The sensor suited for soil sample detection of acetamiprid was an aptasensor based on colorimetric estimation and the sensor suited for water sample

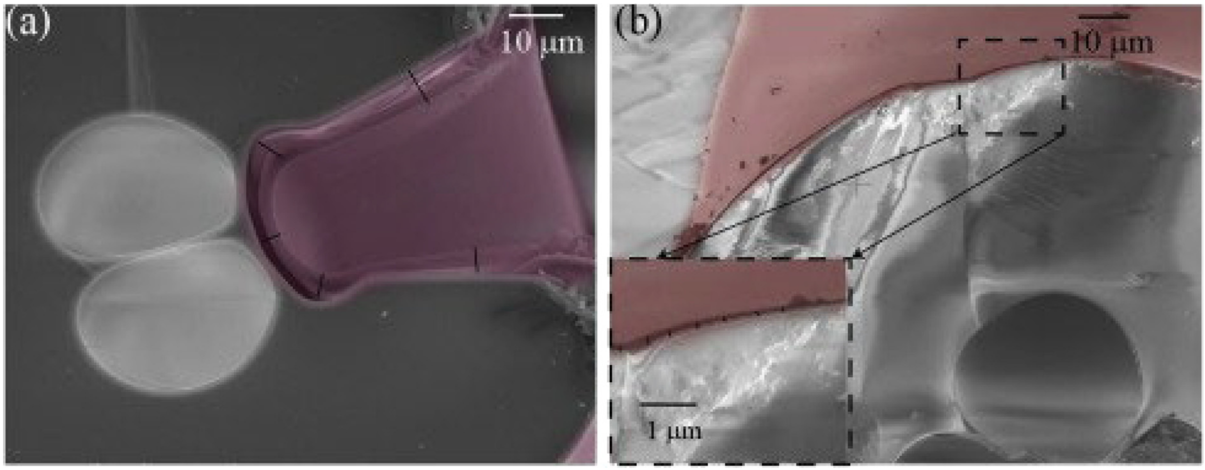


Fig. 7. (a) SEM picture of a silk solution coated ECF probe. The dark pink zone on the outside of the ECF shows a silk layer applied uniformly. (b) A comparatively thinner silk coating (pink) is visible on the fiber in the SEM image of an ECF coated with silk solution. The cross section of the uncoated fiber is shown in gray. An enlarged portion of the fiber used to gauge the coating thickness is visible in the inset inside the dotted rectangle [55]. . (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

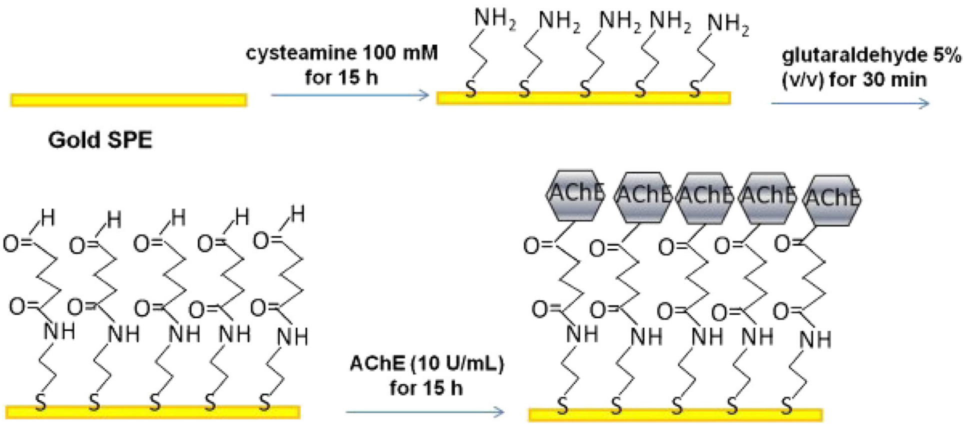


Fig. 8. Diagram showing how the AChE biosensor is assembled [56].

detection of acetamiprid was an aptasensor based on impedimetric estimation [57,58]. Mercury is a toxic and ubiquitous pollutant of the environment. A biosensor was developed for its detection and this was a DNA based optical sensor, which is portable, rapid and cost-effective in nature [59]. Another environmental toxin is brevotoxins, which can induce algal blooms in aquatic bodies, resulting in eutrophication. Its early detection is essential to maintain a healthy aquatic system. Thus, an aptasensor based on electrochemical measurement was fabricated for the detection of this potent neurotoxin, berevtoxin-2. The sensor employed a cysteamine self-assembled monolayer functionalized gold electrode for the sensitive detection of the toxin [60]. A biosensor which was based on an aptamer and gold nanoparticles was fabricated for the fluorescence measurement of biphenyl A in water samples, as it is an endocrine disrupting chemical [61]. Biosensors based on impedance measurements have been fabricated to detect norfluoxetine and BDE-47. These sensors are portable biosensors for environmental monitoring and exhibit 8.5 and 1.3 ng/mL, respectively as their limits of detection [62]. Thus, a wide variety of electrochemical and enzymatic sensors are utilized in the field of environmental monitoring.

3. Innovative methods for the fabrication of bio-nano interfacial interactions

The use of real and complicated biological samples at the point-of-care remains a major problem for researchers working on biosensor design and development [63]. As a result, developing methods for electrode fabrication, biomolecule immobilization, measurement instruments and related materials to be used as bioanalytical tools in different biosensors are critical [64].

Nanostructures add new functionality to biosensor development, allowing for a wider range of applications in clinical diagnostics, environmental monitoring and food safety testing. Amperometric biosensors based on nanomaterials have been shown to expand the capabilities of the present analytical methodologies by allowing for quick and precise analysis. Because of recent advancements with biofuel cells, the overarching objective of developing fully autonomous biosensor systems is now coming to fruition. However, significant biofuel cell and nanotechnology discoveries are still required to achieve this goal [64].

When creating an appropriate biosensing device, an improved and optimized nanomaterial deposition method is critical for the mechanical stability and fabrication reproducibility of the electrodes. Researchers have been utilizing increasingly advanced ways to improve not only sensing performance, but also stability, repeatability and mass production for decades. The deposition of nanomaterials onto a conductive electrode surface is an important step in the development of biosensors, as it improves the performance of the biosensors. To construct a successful matrix of nanomaterials that provides good contact between the material and electrode surface, various approaches have been tried. The goal of nanomaterial deposition is to increase the surface area of the biosensors to improve electroanalytical performance by allowing for more stable immobilization of enzymes in larger quantities and improving the catalytic or bioaffinity properties [65].

To make biosensors, most researchers have used separately produced nanomaterials. A traditional electrode fabrication approach, on the other hand, involves several steps, including combining with binders, covering the electrode surface and drying at higher temperatures [66,67]. Deposition of separately produced nanomaterials mixed with binders is undesirable for various reasons, including poor repeatability and stability, as well as the possibility that binders will block active catalytic sites, reducing the nanomaterials' electrocatalytic performance. In addition, coating and screen-printing these materials results in dense, inhomogeneous films [65].

Electrospinning is a quick and easy way to create nanofibrous structures and materials with high surface-to-volume ratios and desirable tailored characteristics. As a result, adding nanoscale building blocks (NBBs) into electrospun fibers, such as nanoparticles, graphene quantum dots, carbon nanotubes and graphene, has become one of the most popular study subjects in the field of biosensing. However, the dispersion behavior of NBBs in nanofibers, the limited surface area of nanofibers and the lack of immobilization sites for the tested biomolecules continue to hinder the manufactured biosensors' superior performances and applications [68].

Biosensing technologies are increasingly relying on inkjet printing, which is a modern fabrication technique for biosensors. A biosensor manufacturing strategy that is simple, rapid, flexible, high resolution, low cost, efficient for mass production and enhances the capabilities of devices beyond conventional manufacturing technologies has emerged as a result of parallel improvements in both ink chemistry and printers. Inkjet printing is currently a viable printing process as well as a cost-effective fabrication tool for biosensors, particularly point-of-care diagnostic biosensors. The wide range of inkjet printers available enables both the efficient and low-cost manufacturing of smart biosensors that require high-resolution deposition as well as efficient and low-cost fabrication. Although there have been numerous reports of partially inkjet printed biosensors, fully inkjet printed biosensors have also been realized, allowing for rapid mass production because the entire fabrication process may be completed in a single machine [69].

The current state of the art in employing electrical systems for biosensor manufacture has already resulted in sub-ppm detection levels for specific systems, as well as greater complexity in small molecule analyte discrimination. Current systems are also tackling topics like biomolecular sensing and overcoming the difficulties of electrical operation in biologically relevant media. Commercial applications will be dependent in the future on continual improvements in device stability, repeatability, sensitivity and specificity, as well as industrial advances of low-cost processing techniques [70].

4. Perspective on sustainable biomaterials and their applications

Utilization of sustainable biomaterials has emerged in recent years due to public awareness of sustainability and the advantages of biomaterials, such as biocompatibility, biodegradability, biorecognition and stimuli response. In this part of this article, some biomaterials will be given as examples with their application in biosensing.

Chemical sensing using optical fibers is often accomplished by attaching sensor molecules to the fiber, a procedure that uses chemicals that are not physiologically friendly or confined to thin monolayer coatings. To overcome these restrictions and allow *in vivo* biosensing, a silk fibroin coating of optical fibers for encapsulating fluorescent sensor molecules was first reported. A thin film of silk was applied to silica-exposed core fiber (ECF) sample, which is a naturally produced biopolymer made completely of proteins and amino acids. The fluorophore 5,6-carboxynapthofluorescein (CNF) was doped into the silk, allowing for a robust ratiometric fluorescence technique of optical pH measuring. The doped-silk layer's fluorescence signal was then connected into the ECF's core, allowing pH to be measured remotely. For demonstration of real-time pH sensing, a hypoxia model was used in a mouse. As the hypoxia progressed, a steady reduction in the subcutaneous pH in the mouse lumbar region was observed. This study was the first the research looking at the possibility of using a natural silk protein covering to perform fiber sensing within the body [71].

There is still much to learn about how manufactured nanoparticles interact with different types of biological entities and the environment. Several elements could impact a nanoparticle's overall toxicity and cellular uptake. Surface area, dose, chemical reactivity, size and shape, and charge distribution can all be regarded as contributing elements [72,73].

When a nanoparticle needs to enter or move through cells, tissues or organs, its size is important. Moreover, it influences toxicity, cellular absorption, penetration, half-time and therapeutic relevance. More toxicity exists in small particles than larger ones since small particles are accompanied with a larger surface. In addition, large sized NPs cannot escape the body's degradation system. Consideration should be given to a nanoparticle's cellular uptake, distribution and elimination when determining its "ideal" size for use as a medication drug delivery carrier or a diagnostic tool. As a result, there will be less toxicity and good efficacy [74,75].

High NP doses can result in a disproportionate decrease in cell viability due to the increase in the amount of reactive oxygen species (ROS) produced by cells, leading to an altered balance of cellular redox, as presented in Fig. 9 [76]. The shape, charge distribution, form and chemical reactivity (by surface functionalization) of nanoparticles are related to their functionality. These factors influence interactions between biological sources, such as targeting, stability, biocompatibility and toxicity. Surface functionalization is required to reduce non-specific cell adsorption and to keep nanoparticles from aggregating.

The surfaces of nanoparticles have a high level of chemical reactivity, allowing them to influence biological processes in living systems through interaction with proteins (denaturation, misfolding etc.), nucleic acids (DNA damage) and lipids (peroxidation). Different shapes and forms of the same NP have different cytotoxic effects; however the reason for these differences still remains to be enlighted [77–79]. What is more, alteration of the cell cycle differs depending on the type of NP. In addition, exposure of the same NP can have different effects on different cell lines. Therefore, a critical safe-design strategy for the development of nanoparticles' potential applications is based on chemical modification of the surface and structural type of the nanomaterial to reduce the hazardous effects of these tiny objects [80–82].

In the study of Abed and colleagues, they used a lignocellulosic material to develop a fiber based biosensor which is basically a natural sisal yarn containing a non-electro-conductive core impregnated with (polyvinyl alcohol) PVA polymer and coated by poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT: PSS) polymer as an electro-conductive sheath. First, the creation of this new sensor yarn was achieved and then it was introduced into various woven structures to monitor the mechanical behavior of the composite materials generated with these fiber

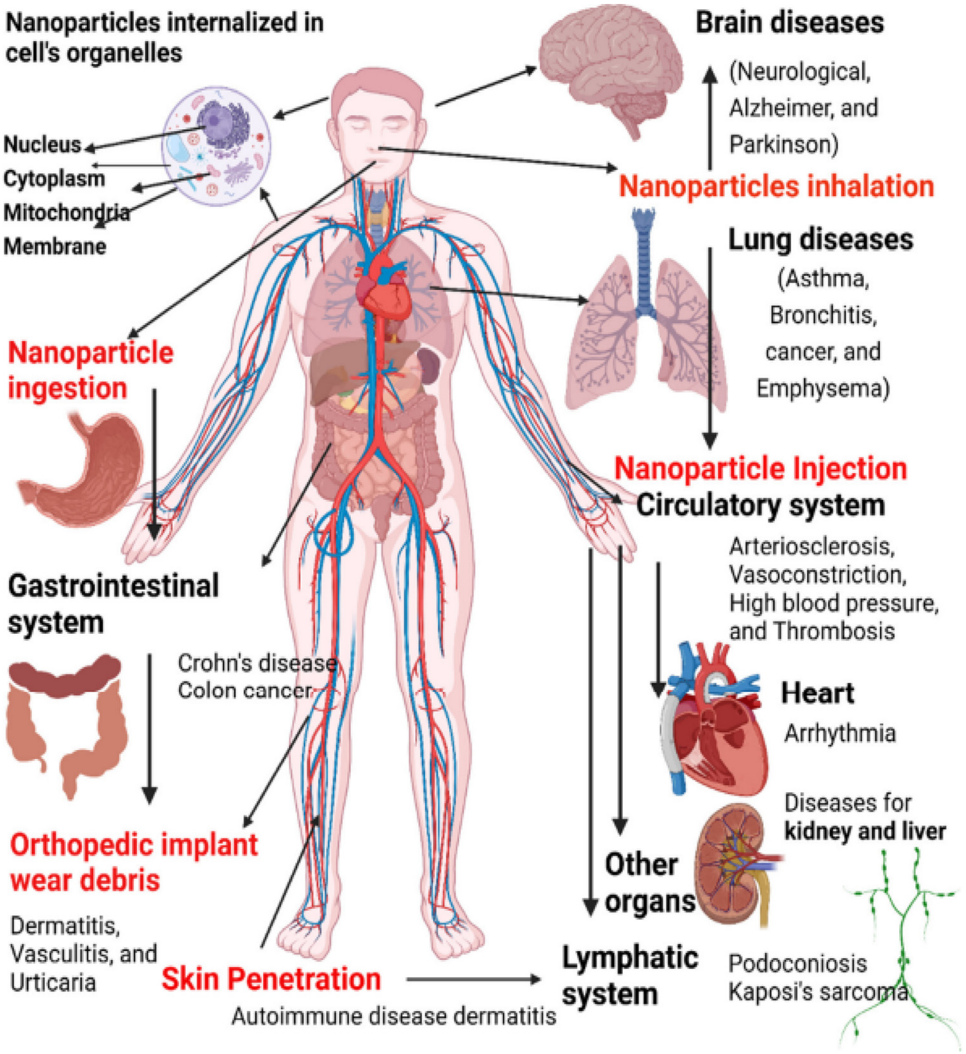


Fig. 9. Possible fate of nanoparticles after their entrance into the human body through different routes [76].

reinforcements. The amount of PEDOT: PSS coating layers on the sensor yarns was discovered to have a significant impact on their electromechanical behavior. On strain-sensor yarns with two and four PEDOT: PSS coating layers, several characterization approaches (e.g. piezo-resistive strain-sensor characteristics) were used and the dynamic strain-sensing behavior was further investigated using cyclic stretching-releasing experiments. Under cyclic settings, these strain-sensor yarns demonstrated precise and robust sensor responses, but it was stated that further investigation should be conducted to reveal the mechanism underlying the first results of these sisal fiber-based sensors [83].

Unicellular diatom microalgae are a promising natural resource of porous biosilica, emerging as a natural source which is extracted from either living unicellular diatom microalgae or diatomaceous earth. This material is biocompatible with an exceptional potential in the field of micro/nano-devices, drug delivery and so on [84].

In light of this information, Kamińska et al. developed a surface-enhanced Raman scattering (SERS) immunoassay for detection of interleukin 8 (IL-8) in blood plasma. This immunoassay is made of diatom biosilica with integrated gold nanoparticles (AuNPs). This assay has an enhanced detection limit (6.2 pg/ml) compared to ELISA (15.6 pg/ml) and the standard deviation of the assay is lower than 8%. This SERS immunoassay also has a good biological selectivity for IL-8 antigen detection and might be used as a helpful platform for the identification of other immune biomarkers in a clinical environment for medical diagnostics [85].

Bacterial biofilms are synthesized as well-defined, readily fiber networks. They can be engineered for various functional properties. They are biodegradable, non-toxic biomaterials. In 2017, researchers produced engineered bacterial biofilms to be used as conductive biomaterials, serving as a platform between electrodes. Different conductive peptide motifs were used to construct the bacterial biofilms to find the best candidates with the highest conductivity.

Genetically introduced conductive peptide motifs, including various tyrosine and tryptophan combinations, converted *E. coli* biofilm into a conductive form. The conductive biofilms can be produced on live cells for biosensing applications by integration of conductive peptide motifs [86].

Polyhydroxyalkanoates (PHAs) are biopolyoxoesters produced by various microorganisms under uneven growth conditions, such as scarce nutrients (N, P, S or O) and excess carbon. PHAs can be produced from industrial waste streams such as food, wine/beverage, dairy, olive-oil and biofuel industries, thus utilization of PHAs in different areas such as medical, packaging, adhesives and agriculture [87] contribute to a sustainable and circular economy. Moreover, PHAs are biodegradable and biocompatible. Sabarinathan et al. developed a fast detection assay with polyhydroxybutyrate (PHB — the main type of polyhydroxyalkanoates) isolated from *Pseudomonas plecoglossicida*. The detection is based on the comparison of attachment of breast cancer cells and normal epithelial cells onto PHB. The results revealed that breast cells adhered on the PHB sheet, while normal epithelial cells showed no adherence property. Surface functionalization of PHB to modify cells' adherence capability may pave the way for the development of different cancer biosensors [88]. This study is also a valuable example of a multidisciplinary study.

Enzymatic biosensors are frequently used in medical diagnostics as complementary instruments to traditional analytical methods thanks to their promising selectivity, high sensitivity and miniaturization/automation capabilities. Among the several enzyme immobilization procedures, cross-linking and covalent binding of enzymes enable the resulting biosensor's high stability. This immobilization procedure, which is obtained manually by drop-casting with a particular cross-linker, is not ideal for the precise functionalization of a single electrode from a microelectrode array.

Utilizing a mussel-inspired electro-cross-linking approach, My Savin and colleagues produced a nanohybrid enzymatic biosensor with great sensitivity using a cheap and large amount of tannic acid (which is a polyphenol found in a wide range of plants), native enzymes and gold salt. It was stated that since the principle behind the electro-cross-linking process is the catechol/amine reaction, this versatile reaction will let this procedure be used with any kind of enzyme. Moreover, development of enzymatic biosensors with longer storage conditions at room temperature (>2 weeks) with stable and high sensitivity are possible. They also mentioned that the electro-cross-linking process, which is based on the utilization of a cheap and abundant biobased compound, can also be used in designing enzymatic biofuel cells [89].

5. Current progress, solutions and future challenges

Technological development in the synthesis of advanced materials with desired structure and features, and employing these materials and their unparalleled characteristics in sensing and measuring [90–94], among other applications, has been a turning point in numerous fields of science. Similarly, sensors, and more especially biosensors, have benefited from the unique features of nanomaterials, such as high specific surface area, abundant binding sites, biocompatibility, physical and chemical stability, and properties such as catalytic and conductivity [95–101]. The unprecedented features of nanostructures in nanomaterials has opened new scope to employ them as: (i) electrode materials [102–104], (ii) carriers [105], (iii) separators or collectors based on electrochemical reactions or magnetic properties [106–108] and (iv) catalysts [109].

Throughout history, nature and its sophisticated engineering models have inspired mankind. Dry adhesion or sticky feet in Geckos, the super hydrophobicity of the lotus leaf and structural coloration in Morpho butterfly's wings are among the well-known examples of mechanical and optical features that rely on the presence of ordered micro- and nanoscale structures, and a perfect balance between order over long and short distances [110,111]. Learning from nature and benefiting the synergy achieved by integration of biomaterials with abiotic counterparts has been a promising approach to tackle the emerging challenges in various field of science [110,112–114]. Having high potential, nature-inspired solutions and materials have found their way into sensing techniques. In this regard, mimicking tactile hairs or bristles in insects as mechanosensory organelles and the design of ultrasensitive pressure and strain sensors [115] using the porous 3D structure of diatoms in algae [116–118], chitin-based contact lenses combined with a graphene field-effect transistor [119] and sensors based on the photonic structures of Morpho butterfly wings [120] can be mentioned. Selectivity (detection of a particular target analyte in a sample containing alike molecules and unwanted contaminants), sensitivity (the lowest amount of analyte that can be detected), linearity (consistency of measurements over the entire range of measurements), response time, reproducibility (similar result for one sample) and stability (vulnerability of the sensor to the internal and external disturbances) are the main characteristics of a biosensor. Biomaterials as well as bio-solutions, have been employed to improve these main characteristics. These solutions can be employed in sensing, transducing and signal processing parts of a sensor or biosensor and in Fig. 10 some of these bio-solutions have been depicted graphically.

Sieving according to particle size as well as electrostatic and steric hinderance [121–123] has been employed to increase the selectivity of biosensors. Using porous matrices as a membrane at the sensing part of the sensors has been adopted to improve the selectivity of the biosensors by rejecting the untargeted analytes. However, limitations, like passing molecules with a similar size to that of the target analyte through the membrane and a decrease in the sensitivity of the sensor resulting from reduced permeability of the analyte, have restricted this approach [124]. In this regard, selective permeation, inspired by glucose uptake via a glucose transporter in the cell membrane, would be a well-suited solution [125]. Glucose transporter 1 (a uniporter protein which facilitates the transport of glucose across the plasma membrane) is the most common type in mammalian cells. Red blood cells constantly uptake glucose from blood plasma and keep the glucose concentration at about 5 mM [126]. Glucose transportation via GLUT1 is selective (distinguishes glucose from other saccharides) and fast enough. The cell membrane of red blood cells contains a high number of these specified transporters and can be used as a permselective barrier for glucose detection and measuring. In a recent study, Kim et al. developed an enzymatic glucose sensor by coating red blood cells on the surface of an electrode and cyclic voltammetry was adopted to validate the performance of the coated electrode [121]. The optimized sensor showed an improved sensing range (1–15 mM), detection limit (0.66 mM) and sensitivity (2.978 μA mM^{-1}). Uric acid (UA) transporter 1 (URAT1) in the kidney cell membrane was employed to fabricate a UA biosensor for selective and accurate detection of UA in human serum. The detection range (0–1000 μM) and accuracy of the biosensor coated with kidney cell membranes to detect uric acid without interference of antioxidants in human blood suggested that it would allow the diagnosis of early stages of gout [127].

Since the emergence of biosensors in 1962 by Clark and Lyons, they have been employed in the detection and measurement of a vast range of analytes. The early biosensing procedure relied on the consumption of oxygen by the enzyme catalyzed oxidation of β-D-glucose to β-D-glucono-δ-lactone [128,129]. Nowadays, commercial blood glucose sensors, as an indispensable part for diabetic people's life, can accurately detect the blood glucose concentration in the range of 1.1–33.3 mM in less than 30 s. The major challenge in enzymatic biosensors is to preserve the activity of the device to achieve long term repeatability [130]. Two possible approaches to tackle this challenge have been: (1) using (nano)materials capable of catalyzing the same reaction as enzymes (developing non-enzymatic biosensors) [131–133], (2) selecting the most compatible support or matrix to immobilize the enzymes. Despite the catalytic properties of nanomaterials, they cannot be as selective as enzymes [134]. Natural polymers like cellulose, chitosan, agarose and proteins have been employed as a matrix to envelop and immobilize the desired enzyme for biosensor application [135–139]. Furthermore, virus-like particles (VLPs) have been used as a nanotemplate due to their cavities. Besides their structural cavities, a series of amino acids (e.g., lysine,

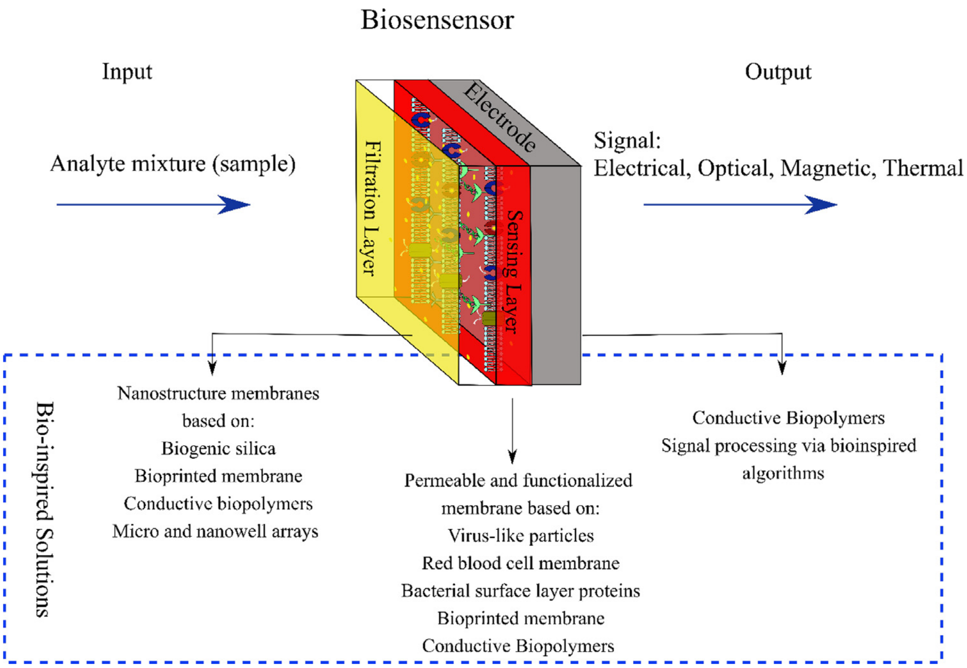


Fig. 10. Schematic illustration about the parts of a biosensor with different kinds of bio-inspired solutions.

cysteine, tyrosine and histidine) located on the protein shell of the viral particles are applicable to fluorescent and medical imaging dyes, proteins or small molecule therapeutics and sensors [140]. Thanks to nanotechnology, nowadays, multifunctional nanoparticles and nanoscale building blocks have been incorporated into VLPs to shape enzyme loaded viral scaffolds [141]. Fig. 11 shows some morphologies and the potential of hybrid VLPs for biosensing applications. A high surface density of the loaded enzyme improved reusability and VLP-assisted sensors have exhibited higher analysis rates compared to layouts without viral adapters [142]. Zang et al. developed a capillary microfluidics-integrated sensor system for label-free biosensing. In this sensor genetically engineered Tobacco mosaic virus (TMV) was utilized as a nanoreceptor for antibody sensing. A dense layer of receptor monolayer formed on the impedance sensor using VLP. The VLP-functionalized impedance sensor was able to detect 55 pM target analytes within 5 min. Moreover, bacterial surface (S-) layer proteins can provide a crystalline two-dimensional protein lattice which can be used on the surfaces and interfaces of the sensors [143].

Conducting polymers can not only provide a direct electrical readout for the presence of analytes, but also they can serve as both sensing elements and transducers at the same time. As a result, polymers like polypyrrole, polycarbazole, polythiophene and poly(3,4-ethylenedioxythiophene) provide the opportunity to miniaturize and simplify sensor designs [145–147]. Moreover, a robust mechanical property, multi functionality for binding sites, biocompatibility and biodegradability of the biopolymers have made an apt choice for fabrication of a variety of flexible electronics, like wearable biosensors [148,149] and skin patchable sensors [150]. Cellulose, silk fibroin and chitin-based polymers have shown desired properties so as to be used in biocompatible flexible point-of-care devices [151,152]. Bioprinting techniques, as an emerging state-of-art method, have received noticeable attention among researchers in different fields, including biomanufacturing, living biosensors, bioremediation and fundamental microbiology [153]. Using bioprinting techniques, three-dimensional patterns of proteins or other biomolecules/biopolymers can be fabricated. Thickness, conductivity and physiochemical properties of the printed film can be adjusted to shape the engineered biofilm required for the sensing and transducing part of the biosensors [145–158].

Analysis of thermal transport through functional interfaces is another novel method which has been employed in sensing [159–161]. Specific changes at the solid–liquid interface can lead to a change in the thermal resistance of the liquid and the solid surface itself [159]. Microelectromechanical (MEMS) thermal sensors have been fruitful for monitoring metabolic applications on the basis of temperature detection [162]. Linear range and high sensitivity, low measurement time and parallel measurements of multiple samples have been reported as the advantages of MEMS thermal sensors [163]. Thermal stimuli, including local heating and temperature gradients across cells, have a strong potential on the formation of morphological and cytoskeletal changes [164]. In other words, temperature gradients can not only initiate cellular processes, but they can also effectively control their spatiotemporal dynamics [165,166]. The sensor measures the thermal resistance at the solid–liquid interface and has numerous advantages over many traditional diagnostic methods [160,167]. For instance, Yongabi, D., et al. used a very mild temperature gradient as a stimulus to trigger the cell response. They found that under specific conditions, synchronized glycolytic-type oscillations are observed during detachment of mammalian and yeast-cell ensembles, providing additional cell-specific signatures [164]. In another study, McClements, J., et al. [168] developed a thermal assay with molecularly imprinted polymer nanoparticles electrografted onto screen-printed electrodes to accurately quantify SARS-CoV-2 antigens. They found that heat transfer-based measurements were superior to commercial rapid antigen tests for both the alpha (∼9.9 fg mL^{-1}) and delta (∼6.1 fg mL^{-1}) variants of the spike protein.

Chronic diseases like diabetes threaten human health and lives, and regulated and continuous glucose monitoring (CGM) has been an indispensable feature of diabetic patients. To date, CGM relies on invasive lancet approaches, which can be accompanied by skin irritation and bacterial infections [169,170]. The existence of some biomarkers in externally excreted body fluids (e.g. tears, saliva, urine and sweat) provides the opportunity

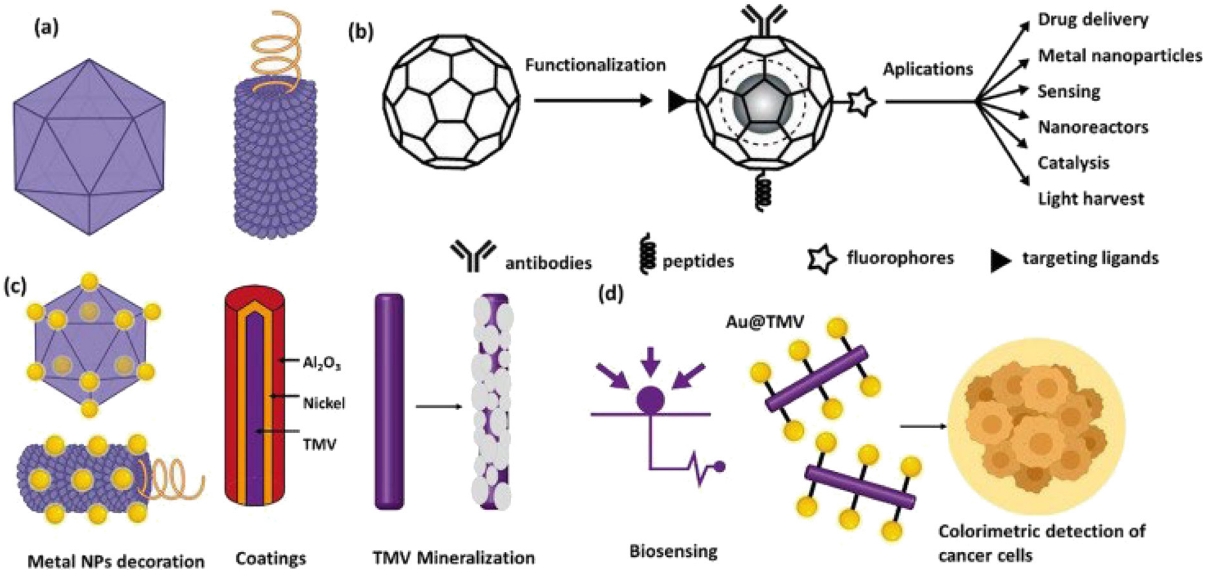


Fig. 11. (a) Hybrid-VLPs with different morphologies; (b) Hybrid-VLPs for several applications. Copyrights MDPI 2020 [137]; (c) Strategies for the obtention of hybrid VLPs systems; (d) Hybrid VLP (Au@TMV) template for the colorimetric detection of cancer cells [144].

for non-invasive diagnostic sensors to be used as an alternative to blood-based diagnoses [171]. In this regard, an ultrathin (∼3 μm) nanostructured skin-like biosensor [172], with high sensitivity (130.4 μA/mM), and a hydrogel-based touch sensor pad for detection of sweat components [171] have been created.

Bioinspired devices uses different types of membranes that depends on the pH at which they operates. Proton exchange membranes are used in fuel cells and other electrochemical cells. Chitin is the mostly commonly used natural proton exchange membrane. This membrane has the role to transport a proton. Other bio-inspired and bio-derived materials are presented in Fig. 12.

The numerous publications in recent years, more than 29,000 studies since 2016 (based on the web of science website's statistics with biosensor as a keyword), indicate the urge of academy and industry to wield biosensing in non-biological applications. However, most of the available biosensing assays require controlled and clean conditions for precise measuring, which are unattainable in analyzing real non-biological samples, like water, as well as food material. As result, despite the invariable growing market of biosensors (over $18 billion for point-of-care devices), no conclusive records can be found regarding the real potential and market of biosensors for non-biological applications. Therefore, efficient mass production of reliable biosensors, as a prerequisite for product commercialization, is still highly desired. Multifunctional and robust biosensors for non-biological samples are among the missing parts of the biosensing roadmap and further investigation of natural phenomena and mechanisms could be a leading clue to find these invaluable missing parts. Despite the progress in enhancing the signal-to-noise ratio of the biosensor's response in recent years, sensitivity has been a perpetual challenge in this field. Concentration of a great deal of practical cancer and cardiovascular biomarkers in biological samples is far below the detection limit of current methods. Additionally, despite the relatively high concentrations of protein or nucleic acid biomarker in cerebrospinal fluid (CSF), their concentrations in blood, saliva or other biofluids are extremely minute to be detected by currents biosensors [174]. Bioinspired methods with improved sensitivity, like micro- or nanowell arrays, for single-molecule measurements can allow the detection of these biomarkers directly from the blood via minimally invasive sampling [175–179].

A minute amount of sample containing different molecules (target and non-target analytes), where their concentrations can be interdependent, necessitates detection and measuring methods capable of simultaneous analysis of more than one analyte. Currently assays only detect one analyte at a time and microfluidic assisted label-free biosensors can be useful to address the limitation of multiple analysis at a time. In label-free biosensors, the physical properties of the targeted analyte, such as Raman scattering, refractive index and second harmonic generation, are used to trace the target molecule(s) [180].

Although numerous studies have been reported on the high potential of bioinspired approaches in various fields of application, there are still some obstacles need to be addressed for practical and commercial usage of these approaches in sensing and measurement.

(1) There is a lack of holistic cognition of the real mechanism behind the bio-element or behavior which is going to be used or mimicked. For instance, salicylic acid is critical to regulating many aspects of plant growth, development and defense. However, It has become increasingly apparent that salicylic acid is part of an intricate network that involves many other plant hormones [181].

(2) Sensing and more especially biosensing output signals (e.g. gene expression, enzyme activity, light, fluorescence or electron release) can enable us to shape a two-way communication between human and biological processes. However, active read out (sensing) due to language barriers (lack of sensitive and selective interaction mechanism between the abiotic counterparts of sensor and biological elements) is still a matter of concern in biomimicking and using bioderived nanostructures [182]. In this regard, machine learning (ML), artificial intelligence (AI) and bioinspired algorithms [183–185] have been reported as beneficial aids in signal sorting, noise reducing and finding patterns, and these consequently increase reliability, accuracy, sensitivity and selectivity [186–189]. In other words, in respect of two-way communication, ML and AI can help us in deciphering the bilanguage of bioprocesses. Table 1 shows the different types of bioinspired sensors.

(3) No conclusive data exists on the cost-efficiency of using biological elements (e.g. viruses, biogenic silica, red blood cells, among others) in commercial biosensors.

(4) There is a lack of physical and chemical stability assessment during extraction, fabrication and service.

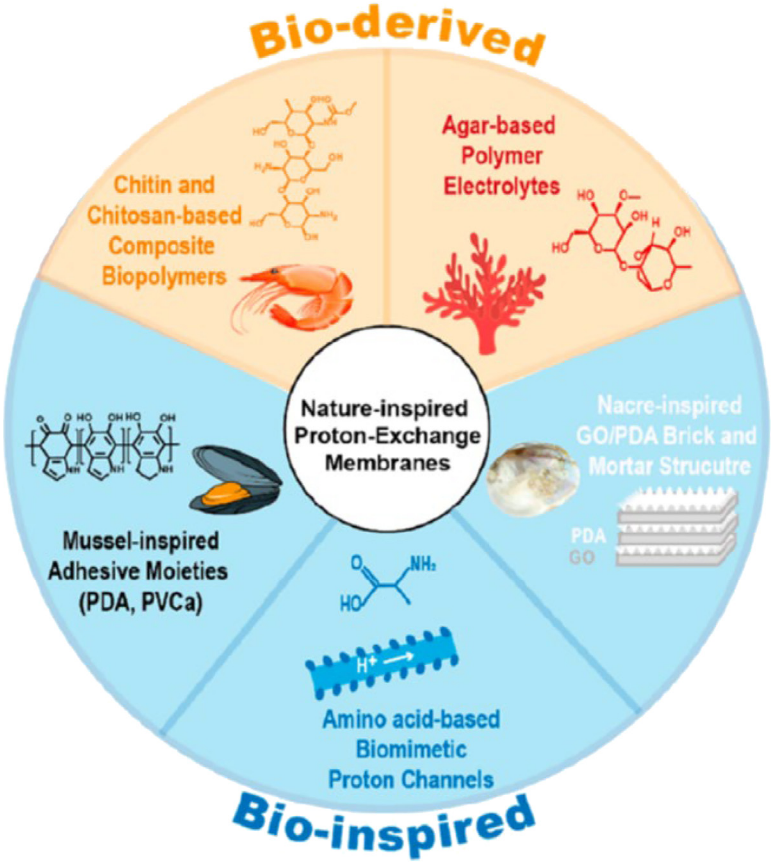


Fig. 12. Bioinspired and bioderived materials for proton exchange materials [173].

Table 1
Different types of bioinspired sensors.

Types features	Piezoresistive	Capacitive	Triboelectric	Piezoelectric
Bionic strategy	Bionic materials [190] Bionic structures [191]	Bionic materials [192] Bionic structures [193]	Functional bionic [194] Bionic materials [195] Bionic structures [196]	Functional bionic [197] Bionic structures [198]
Bionic source	Plant surface [190] Human Skin [199] Stereocilia [191]	Human skin [192] Komochi Kondu [200] Spongia [193] Plant surface [201] Microhair [202,203]	Plant surface [195] Ion channels [196] Eardrum [204] Auditory system [205] Olfactory epithelium [194]	Human skin [206] Hair [198] Fingertip [197] Corti cells [207]
Sensitivity	120 kPa^{-1} [206] $\sim 10^{3}$ kPa^{-1} [192] 83.9 kPa^{-1} [199] 1.5 Ω μm^{-1} [208] 30 mV (m S^{-1}) [198]	0.293 kPa^{-1} [201] 0.171 kPa^{-1} [209] 0.63 kPa [210] 0.815 kPa^{-1} [211] 0.56 kPa^{-1} [212]	127.22 mV kPa^{-1} [213] 51 mV Pa^{-1} [209] 110 mV dB^{-1} [214]	2.21×10^{-3} V Pa^{-1} [200]
Response time	$<$16.7 ms [215] 30 ms [190]	162 ms [200] 38 ms [201]	$<$6 ms [204]	
Measuring range	0.88 Pa–32 kPa [190] 0.02–30 kPa [199]	0–250 kPa [200] 0–90 kPa [193] 0-50 N [201]	0.1–3.2 kHz [204] 100–5000 Hz [205]	1–500 Hz [198] 100–1600 Hz [216]
Durability	$>$1000 cycles [190] $>$28 000 cycles [202]	$>$1000 cycles [192] $>$10 000 cycles [200] $>$10 000 cycles [193] $>$3000 cycles [216]	$>$1000 cycles [194] $>$5000 cycles [195] $>$50 000 cycles [196] $>$40 000 cycles [204]	
Functions	Flow sensing [191] Tactile sensing [191] Pressure sensing [190,217]	Tactile sensing [193] Pressure sensing [216]	Tactile sensing [195] Acoustic sensing [205] Pressure sensing [204] Gas sensing [194]	Pressure sensing [206] Temperature sensing [206] Acoustic sensing [198] Flow sensing [198] Tactile sensing [197]

6. Conclusions

Today, sustainability has become an indispensible requisite in all aspects of human life. In this regard, sustainable energy, lifestyle and measuring can be mentioned among others. Through the history of the Earth, nature always has had its sustainable approaches to address its emerging needs. Mocking up nature, sensing technologies have benefited from sustainable bioinspired solutions and materials throughout the history of biosensor development. In this review article, several examples of bioderived/bioinspired materials employed in biosensors in last five years, have been mentioned. Biomaterials and bioinspired solutions could open new windows on developing sensing applications in terms of superior features, such as biocompatibility, conductivity, electroactivity, flexibility, biodegradability, implantability and so on. In numerous studies published in recent years on biosensors, the combination of technological development with bio-solution has resulted in a quantum leap in the sensitivity of detection and measuring methods. In this regard, a deep-learning program for determining the 3D shapes of proteins can be mentioned. Despite the significant improvement in sensing technology in recent years, there is still room in furtherance of their ability by deeper cognition of the real mechanism behind the bio-element or bio-behavior.

Abbreviations:

- ISO-standard: Internationally recognized standards
- DNA: Deoxyribonucleic acid
- MEMS: Microelectromechanical sensors
- NEMS: Nano electrochemical sensors
- AC: Alternating current
- E. coli: Escherichia coli
- LOD: Limit of detection
- AuNWs: Gold nanowalls
- ECF: Exposed core fiber
- CNF: 5,6-Carboxynapthofluorescein
- SEM: Scanning electron microscope
- NBBs: Nanoscale building blocks
- NP: Nanoparticles
- PVA: Polyvinyl alcohol
- PEDOT-PSS: Poly(3,4-ethylenedioxythiophene)polystyrene sulfonate
- SERS: Surface-enhanced Raman scattering
- AuNPs: Gold nanoparticles
- PHAs: Polyhydroxyalkanoates
- PHB: Polyhydroxybutyrate
- URAT1: Uric acid transporter 1
- VLPs: Virus-like particles
- CGM: Continuous glucose monitoring
- CSF: Cerebrospinal fluid
- ML: Machine learning
- AI: Artificial intelligence

Declaration of competing interest

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

Data availability

Data will be made available on request

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