Chapter 10

Advanced Functional Membranes for Sensor Technologies

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Abstract

Sensors are modern data acquisition devices intended for detecting a specific change in the surrounding area and responding with an output signal. Among them, biological sensors (or biosensors) are of great interest due to their capacity for detecting molecules that indicate changes in people's health. This chapter provides an overview of the polymeric membranes that have novel or advanced functions in applications for the development of lab-on-chip or sensor technologies. Important approaches in this matter include the improvement of membranes as supporting medium of recognition element of the sensor, advances in membrane composition for protecting the integrity of target molecules, or the option to filter undesired components. The chapter is divided into three sections: membrane fabrication, molecular probes and platforms for reading sensor devices.

Keywords

Active Sensor, Target Molecule, Transducer, Wearable Technology

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1. Introduction

Any device with the ability to detect and respond to a physical stimulus from the environment can be considered a sensor. The specific stimulus could be light, heat, movement, humidity, pressure, or other parameters that define the physicochemical state of the environment (system). The response is generally a signal that is transmitted on a user-readable screen.

In this sense, a device that responds to the presence of a particular chemical selectively and transforms the chemical quantity input into an output signal is known as a chemical sensor. That was defined by IUPAC in 1991 as a device that transforms chemical information [total sample concentration or specific component (analyte) concentration] into an analytically useful signal [1]. Chemical sensors consist basically of two components connected in series: a chemical recognition system (or receptor) and a physico-chemical transducer. The receptor is capable of selectively recognizing a certain analyte through a chemical interaction with it. The transducer is responsible for physically transforming the chemical signal, which is the result of the interaction between the receptor and the analyte, into another signal that is more easily quantifiable, measurable and manipulable. On the other hand, if the chemical sensor involves some kind of biological material as a receptor, then it is called a biosensor [2–4].

In the technological development of chemical sensors (including biosensors), membranes have played an important role, despite not being included in the IUPAC definition among the components of chemical sensors. A membrane should be understood as an interface acting as a selective barrier between two adjacent phases [5]. Its main function is to regulate the exchange of substances between the two adjacent phases, through the transport of matter within itself. The main advantage of the membrane technology is the selectivity of transport through the membrane, which depends on the chemical and physical structure. The regulation of the exchange of substances through the membranes does not require additives and can be carried out isothermally at low temperatures, which requires low energy consumption compared to other thermal separation processes. Hence, membranes are intended to be used as filters for concentrating and isolating analytes, but also, they have been used as platforms to immobilize, encapsulate or protect the receptors and bioreceptors, and they proved to be useful.

This chapter reviews recent literature on membranes specifically for their use in the development of sensor technologies. The chapter is divided in three sections, membrane fabrication, molecular probes and platforms for reading sensor devices.

2. About sensors

Sensors are common devices in modern data acquisition systems. Human beings use them everywhere on a daily basis, even if not noticing their presence, because many of them are smaller than a hair. They are devices or subsystems whose task is to detect a specific change in the environment and respond with a specific output signal. I.e., a sensor transforms a physical or chemical phenomenon into a measurable, easily interpreted, human-readable analogue or digital signal or transmitted for reading by another component or device, for further processing. A couple of examples of sensors that we use in everyday life, without having them in the mind as sensors are: (i) the microphone, which is a device that converts sound energy into an electrical signal, which can be amplified, broadcast or recorded; and (ii) a mercury thermometer, that is an old-fashioned sensor for measuring temperature. In a smartphone, there is an example of a system integrated by several sensors, it is a device that is not a sensor itself, but is loaded with several sensors such as; a fingerprint reader, an accelerometer, a magnetometer, or a proximity or ambient light sensor.

Human technology has progressed dramatically and many critical processes could not be possible without sensor data. Sensor technology is everywhere in our homes, hospitals and shopping centres. Sensors manufactured for applications in hotels, homes and smart rooms enhance artificial environments experience and imply a reduction of services costs. This reduction is achieved through intelligent energy management using automatic light and temperature controls by using thermostats and occupancy sensors [6–11].

Today, there are many types of human-made sensors because we have put our skills and imagination into the design of an overwhelming variety of sensors; therefore, a classification of them is useful, as is shown in Fig. 1.

In various research reports sensors are classified in various ways, including a review of the different categories of sensors[12–14]. Sensors were first classified as active and passive; this group is included in the category "Based on collecting data technique" in Fig. 1. Active sensors need external excitation energy for data acquisition, while passive sensors have no need for external energy to provide an output response [15,16].

Another category used is "Based on the Application", which considers the technological field or the activity of its applicability, e.g.; automotive, domestic application, telecommunication, space, agriculture, manufacturing, for daily life, or quality of life and health. However, the most used category is "Based on the detection principle used", where

sensors are grouped into physical; chemical; thermal; and biological (biosensors, in abbreviated form).

Figure 1 Sensors Classification

Physical sensors are characterized for measuring a physical property such as force, pressure, acceleration, flow rate, mass, volume, or density. I.e., microelectromechanical systems (MEMS) are physical sensors used in the biomedical field. Whereas chemical sensors are defined as devices that chemically interact with a target molecule or analyte. According to the IUPAC, they also have two parts; the recognition element, in charge of identifying the analyte; and a transducer that transforms chemical information into an analytically useful signal (analyte concentration or/and total composition analysis)[17].

Biosensors are also defined by the IUPAC in a similar way to chemical sensors, with the main difference in the nature of the recognition element or probe; which must be formed by a biomolecule such as an antibody, antigen, DNA fragment, enzymatic, or polysaccharide. Therefore, biosensors specialize in the detection of biomolecules of interest in the diagnosis of diseases [1,3,4,17–19]. Chemical sensors and biosensors, both are used to obtain information about the activity or concentration of the relevant chemical species in the gas or liquid phase, with importance for the control of environmental contamination, food analysis, drug analysis, control organophosphate compound assays, or for clinical diagnostic purposes.

For the physical sensors in some cases the membranes are an important part of these devices, as in the case of the sensors based on Quartz Crystal Microbalance (QCM). But it has been chemical sensors and biosensors that have made the best use of the sales that membrane technology offers to sensor development. So we will focus on QCM-type sensors, biosensors and chemical sensors.

2.1 The importance of membranes as sensor components

As mentioned in the introduction, chemical sensors have basic elements such as a receptor and a transducer. The receptor interacts with the analyte (target molecule) to transform chemical information into chemical energy and transfer it to the transducer. The transducer transforms the chemical energy into an energy of another physicochemical nature. This process is shown in Fig. 2, considering a chemical sensor interacting with a sample with *n*-components. Each component has a different chemical identity and only one of them is the molecule of interest for sensing, which corresponds to the analyte or target molecule. The analyte must approach the probes or "recognition elements" which are chemical molecules capable of recognizing the target. These probes can be synthesized in the laboratory or extracted from a natural source. The interaction between the analyte and the probe can be chemical or physical. If the interaction is chemical, it can be destructive as a chemical reaction, or non-destructive as a non-covalent interaction between the analyte and the recognition element. This process will generate specific chemical information from the interaction between analyte functional groups electrons and probe electrons. In this sense, biosensors are chemical sensors (they are a subgroup) whose chemical probes are biomolecules such as nucleic acid fragments, enzymes, proteins, or polysaccharides; according to the IUPAC definition [4,5,17,18].

The chemical energy related to the interaction between the probe and the analyte must be transformed into the energy of easy handling to generate a useful analytical signal. The transducer will convert chemical energy into light energy, fluorescent energy, acoustic energy, calorific energy, electrochemical energy, etc. Therefore, a user-friendly signal will

be produced at the transducer, as illustrated in Fig. 2. This signal must be processed later by a properly designed electronic system. Hence, Fig. 2 is the general representation of chemical sensors and biosensors depending on the nature of the probe, but in which the role of the membranes is emphasized. In Fig. 2, a membrane has the function of being the supporting medium of the recognition element, which must be immobilized and ready to work. In the case of biosensors, the biomolecules generally used are sensitive to the oxygen in the atmosphere and to other chemical compounds that may be present in the sample. Therefore, probes based on biomolecules must be protected from possible chemical attacks or unwanted interactions, and here the membranes have another great role; to encapsulate the probes to protect them.

Figure 2 Elements and components of a typical chemistry sensor or biosensor

A third role of the membranes also has been represented in Fig. 2, by placing the different components of the sample at different distances from the probes. This is intended to represent the importance of membranes to filter or separate uninteresting components from the target or analyte.

3. Membranes for sensor technologies

The IUPAC defines membranes as structures that have lateral dimensions significantly greater than their thickness, through which mass transfer can occur under a variety of driving forces. They are continuous layers composed of a semi-permeable material, with controlled permeability. Therefore, the membranes can separate the components of the test or sample solution [20,21].

Biomedical applications of membranes are extensive, such as the semi-permeable membranes that only allow target molecules to pass through them. As well as platforms or support structures for other layers, to immobilization, and/or to encapsulation of sensing probes as illustrated in Fig. 3. They are also used as a support structure for proteins, DNA, and RNA within techniques such as western, southern, and northern blots, or indirect epifluorescence (DEFT). For this reason, device developers integrate membranes into sensors (particularly biosensors). Detection of pathogens or diseases (through biomarkers) has been achieved using some of these sensors. A classic example is the glucometer or glucose sensor, which can have more than two different types of membranes.

Figure 3 Applications and uses of membranes in sensing technology

Given the variety of applications and the different important roles that membranes must fulfil, their design requires a high control of various parameters, which can be grouped into

two major focus areas. On one hand, the physical parameters such as surface area and roughness, porosity (pore size and distribution), and membrane thickness. On the other hand, chemical parameters or characteristics such as chemical composition, functional groups (type and their distribution), chemical affinity, swelling, and chemisorption properties.

The physical parameters of the membrane are decisive for filtration and separation applications; while the chemical parameters are essential for structural support. For the chemical sensor design, both sets of parameters are important.

Chemical properties can be studied taking advantage of the pragmatic division that has been used for years, for the study of chemical compounds; i.e., membranes can be classified as organic, inorganic, and hybrid (composite). Some examples of the materials used in the manufacture of those membranes are mentioned in Table 1.

The main function of inorganic membranes has been as a support structure, by increasing the sensor surface area, to carry out a capillary action, or as mediators of fluid transport by capillary action. As mentioned in Table 1, glass membranes also are good examples of inorganic membranes that have been reported [38–40] for biosensor applications. In this table, more examples of inorganic membranes are given; nevertheless, the reports of organic membranes in sensing applications are much more numerous.

Organic membranes are used primarily as a support structure to immobilize sensing probes and as a filter medium. However, they are also used to microencapsulate and protect sensing probes or biorecognition elements such as enzymes. But they have also been used as a functional part of the recognition element, such as fluorescent polymeric membranes that are both support and chemosensor [41–43].

Another main difference between organic membranes and inorganic ones is the great variety of refilled materials for organic membranes. Cellulose derivatives, as seen in Table 1, are one of the first materials reported for the manufacture of organic membranes used in the development of sensors, again, the glucometer is a good example of this. In addition to the materials already mentioned (Table 1), polyethersulfone, polydimethylsiloxane, polycarbonate, polyvinyl chloride, polyamine, polyvinylidene fluoride, nylon, 2-hydroxyethyl methacrylate, polyacrylamide, polylactic acid, among others, are also used with that purpose.

Table 1 Examples of materials for membranes manufacturing

Type of membrane	Material	References
Inorganic	Aluminum or aluminum oxide	[22–25]
	Gold	[26–28]
	Silver	[29–31]
	Titanium oxide	[32–34]
	Silicon nitride	[35–37]
	Glass membranes	[38–40]
Organic	Cellulose nitrate, nitrocellulose, cellulose acetate	[2–4]
	Polyurethane	[1][2–4]
Hybrid (composite)	Titanium Carbide ($Ti_3C_2T_x$) modified with Ag nanoparticles	[31]

Sometimes it is desirable to have both organic and inorganic membrane properties. Hence, it is not unusual to find hybrid membranes for sensing applications, which are composed of organic and inorganic components that effectively merge. This type of membrane can be of two types; (i) Organic membranes covered by a thin layer of inorganic material, usually, a conductor such as gold [31,40–45]; and (ii) Membranes synthesized by sol-gel methods to obtain structures of inorganic nanoparticles with polymer chains[13,26,30,46]. Finally, composite membranes are multiple layers of membranes that are stacked together side-to-side or vertically to comprise a whole sensor[32–38,47].

4. Wearable membranes sensor

The prospects for sensor technology point towards its integration with wearable technology and flexible electronics; with the aim of developing portable sensors and biosensors, as less invasive as possible and with continuous data collection, to monitor user activities. Challenges in this area include standardization, device connectivity, interaction between multiple devices, compatibility between the gadgets and the analysis software. On the other hand, the growth of technology has allowed portable devices to become an everyday and versatile tool. Portable devices currently have several integrated sensors with which they acquire the data necessary to carry out the tasks for which they were created. Statistical

data is even collected to study our habits and preferences with the intention of improving the user experience[48–50].

Wearable or portable devices make use of wireless strategies for non-invasive, real-time monitoring in many applications. One of these applications that have gained special relevance and interest is the personalized monitoring of parameters related to health and fitness. These portable devices can be designed as common wearables; i.e., smartwatches, bracelets, hearing aids, electronic or optical tattoos, sports glasses, screens, subcutaneous sensors, electronic footwear and textiles. In this context, there is a keen interest in membrane development that improves these technologies. To achieve the efficient applicability of membrane technology (continuous, efficient and without losing its general properties), the membranes must meet specific requirements in terms of body adaptation and biocompatibility. Traditionally, the common approach to developing sensing technology was miniaturized electronics mounted on rigid platforms [13,40,49,51–53].

However, they have a low body adaptation capacity due to their low resistance to deformation and, therefore, flexible materials and membranes come into play in this new trend of developing wearables. To achieve the desired mechanical stability, the electrodes and, in general, the electronics associated with the sensors and biosensors must be flexible, therefore, they require the introduction of characteristics such as the ability to stretch to withstand mechanical deformations and the associated mechanical stress during use. Furthermore, these membranes must be non-toxic and biocompatible to minimize the risk of epithelialization or allergic reactions. In this context, the use of enzymes [54–57], hydrogels [6,41,58–60], and conductive polymers such as polyaniline (PANI) [61–64], polypyrrole (PPy) [65–67], polythiophene (PT) [68–70], or polyethylene dioxiide thiophene (PEDOT) [43,71–74] has been reported for the development of bioelectrodes due to their biocompatibility, conductivity, and the presence of tunable functionalities. However, its application in the development of biosensors is limited by different reasons and challenges to overcome, such as its relative fragility; its decrease in properties against variations in the pH of the medium, possible degradation in toxic or reactive compounds; and in some cases the use of synthesis processes and complex manufacturing procedures that are difficult to implement outside the laboratory scale [70–76].

Conclusions

The chapter describes the importance of membranes for sensing technological applications, including specialized selectivity, encapsulation features, and structural supporting properties. Materials for manufacturing membranes are used depending on the purpose of the specific sensor or the selected target molecule. Polymeric materials had been combined with inorganic materials for obtaining a greater efficiency in the sensor design. Physical

parameters (surface area and roughness, porosity, and membrane thickness) and chemical parameters (chemical composition and affinity, swelling, and chemisorption properties) determine membrane performance in sensing techniques. The prospects for sensor technology point towards its integration with wearable technology and flexible electronics, aiming to develop less invasive biosensors with continuous data collection to monitor user activities and sending those data to a health professional on a real-time basis. The development of membrane technology is crucial to extend the technological frontier for these sensing/biosensing devices and platforms. It is the polymeric membranes that will help take the step to the front of the wearable and portable biosensors, which promise a great advance in the monitoring of biometric parameters to improve the quality of life of the modern human.

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