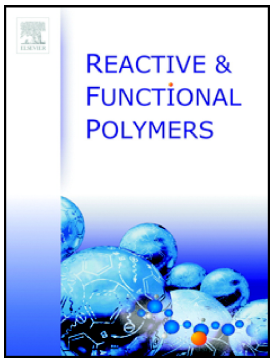

Recent developments in sensing devices based on polymeric systems

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Recent developments in sensing devices based on polymeric systems

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Abstract

This review is focused on the analysis of recent developments in the application of polymers in the detection and quantification of target species. The work begins with a description of the polymers that are employed as sensory materials, covering molecularly imprinted polymers or MIPs, hybrid polymers, acrylic polymers, conductive polymers, polymers with chiral motifs and also the use of polymeric arrays. After the description of the sensory polymers, the different target species which can be detected using sensory polymeric devices, including metallic cations and anionic species, gases, explosives, radionuclides and bacteria or the recent biomedical and biological applications is described. Finally, the sensory devices fabricated using smart polymers, including, for example, sensory devices based on Quartz Crystal Microbalances or the use of micro and nanoporous materials as substrates for sensory polymeric coatings is listed and reviewed. The work also details the different detection mechanisms based on the type of response of the sensory polymers, such as electrical, piezoelectric or fluorescence. In brief, the review details a review of the research work published in the last 10 years in this quickly evolving field, with special emphasis in the biomedical and biological applications, which have emerged recently raising great attention. To conclude, some perspectives and future challenges that must be overcome by this research field in the next years is exposed.

Keywords: Sensors; smart polymers; sensory polymers; detection; sensory devices

1. Introduction

The development of supramolecular chemistry by Pedersen, Cram, and Lehn in the 1960s brought forth the growth of a new research field called chemical sensors or chemosensors. These are molecules having receptor which provide information about the chemical composition of its environment through selective interaction with target molecules or analytes. In general, chemical sensors are usually organic or organometallic low-mass molecules, but they present some disadvantages which limit their applicability: They are generally water insoluble, exhibit moderate to low light and thermal stability, and tend to migrate when they are dispersed in physical supports.

However, the optimization of polymer-based chemistry has led to a completely new family of sensory materials and devices employing polymers that have the ability to respond reversible or irreversible to different stimulus in their environment. The sources that are able to cause such responses can be very different, for instance, temperature, pH, biological molecules, ionic strength or electric or magnetic fields. The response can take place in several ways such as modification of surface properties, changes in shape, solubility, color or fluorescence, and for this reason it is essential that the selective interaction that gives rise to the response relay on the recognition of the target species by the receptor motifs of the polymer, can be easily transduced to originate an measurable change. This kind of polymers are an emerging field due to their interesting properties and the fact that they can be employed in a wide range of applications such as medical devices and biomedical applications, drug delivery, tissue engineering as well as bio/sensors. Moreover, it is well known that polymeric sensors can be easily manufactured into different shapes such as micro/fibers, films, beads, coatings, wires, etc., and they can overcome the difficulties presented by the classical chemical sensors, then being used in many applications. [1-4]

In parallel, the society demands for new materials with specific applications, and in this sense, the role of sensory polymers and their use in detection devices has gained a lot of attention, due to their direct applicability to solve real-life problems. We can remark, for example, biological and biomedical applications (early detection and diagnosis of different diseases), the current terrorist menace, which requires the quick detection of harmful gases and explosives, or, for example, the detection of environmental pollutants in food-related applications (such as the presence of pesticides or heavy metallic cations). For all these reasons, the number of research papers and reviews published in this topic has increased greatly, is specially in the last two or three years.

In this context, a simple bibliographic research results in a collection of review papers dealing with this topic, from which we can extract a few review papers that analyze the sensory polymers and their applications. For example, the work of Chichosz *et al.*[5], outlooks different polymeric sensors to detect gases, temperature or moisture are analyzed, but only covering a thin range of applications. Ahmad *et al.*[6] report an interesting review devoted to the use of Molecularly Imprinted Polymers or MIPs in electrochemical and optical sensors. In this review, the use of electrochemical and optical sensing based on molecularly imprinted polymers (MIPs) is described mainly for biological and biomedical applications, as for example, point-of-care testing in real human samples.

Another review paper published recently was presented by Kassal *et al.*[7] This review describes the emerging research field of wireless chemical sensors and biosensors technology and applications, covering the period from 2006 to 2017, emphasizing the emerging interest of wearable chemical sensors in, for example, sport, health or fitness applications, and paying attention to the smartphone compatibility. The review covers the use of sensory polymers and devices (specially polymeric chips), in different devices to detect pH, temperature or environmental pollution, and focus its attention to the new wireless application and the use of

smartphones to the detection and quantification signals, but specifically focused on the field of wireless sensors and biosensors.

Environmental applications have also gained a lot of attention in the last years. Yan *et al.*[8] present a review detailing the use of optical sensors for the detection of pesticides, in which current optical sensing strategies are summarized, together with progress on sensitivity, reproducibility, selectivity and portability and also the advantages and limitations of these devices. Different sensory polymers are analyzed, such as molecularly imprinted polymers or the detection of pesticides using colorimetric changes in polymer-based nanocomposites.

The food-related applications are described extensively in the review presented by Pavase *et al.*[9] The paper presents an overview of food spoilage and conventional detection techniques, focusing attention specifically conjugated polymers and conjugated polymer nanocomposites and their applicability in these straight applications. The paper also details some detection mechanisms, such as colorimetric or naked eye detection, concluding with a future prospective of these sensors in the food packaging industry.

Finally, we include another recent review paper reported by Andre *et al*.[10] This work describes the theory, technology and applications of hybrid nanomaterials as volatile organic compounds sensors. In this work, the use of conducting polymers, combined to inorganic materials, such as carbon nanotubes or metal oxides to form hybrid materials is reviewed, then analyzing the improvement in different sensing characteristics (sensitivity and limit of detection).

All the review papers referenced above have been published in 2018, and although a more exhaustive research can be carried out, it is clear that review papers are mainly focused in targeted applications and/or specific type of sensors, then covering only a narrow research field of the extensive polymeric sensors investigation line. For this reason, we believe that a

review focused on sensory devices based on polymer materials, with a general and wider scope which covers from chemical to technological aspects would attract the attention of the scientific community.

Following this motivation, our group has recently presented a brief review paper in Chemosensors[11], which presents a description of the main sensory polymers and some of the sensory devices which can be employed using polymeric materials. This work can be considered as a first step to cover different and complementary aspects of the sensory devices based on polymers, from a wider perspective, in which we resume both the chemical aspects of the sensory polymers together with the sensory devices that are currently employed in different applications.

The work presented here is a further step, in which we intend to deepen each aspect of our previous work, describing in detail the different sensory polymer families, classifying and analyzing the spectrum of the target species (heavy metallic cations and anionic species, volatile organic compounds (VOCs), gases, explosives, biomedical and biological species, etc.), and finally, describing the sensory devices employed in terms of the sensory response. This last part will include devices based on piezoelectric responses, such as Quart Crystal Microbalances, electrical variation (sensory devices in which modified electrodes with polymer coatings are used) or the recent chips which are employed in biomedical applications for the detection of glucose and disease markers. This review includes more than 250 references, from classical papers analyzing the sensory polymers to more specialized works detailing specific sensory devices for a determined application, with special attention to the last advancements in the last 10 years.

The review is organized as follows: in section 2, a description of the main sensory polymers families will be given, with a brief description of the synthesis and chemistry to justify their use as sensory materials. In section 3, we will describe in detail the different target species, from

heavy metallic ions or volatile organic compounds which have been detected for decades, to the recently investigated biological molecules, such as glucose or aminoacids and also radionuclides and bacteria. Finally, in section 4, a description of the different sensory devices will be given, covering from classical modified electrodes to detect target species through electrical variation to new sensory devices based on biochips to detect traces or disease markers through color or fluorescence variation. To conclude, in section 5 some perspectives and challenges will be briefly exposed to give an idea of the main investigation lines involved in the next years.

2. Description of the main sensory polymer families

Sensory polymers are polymers that have the ability to show a response when they are in contact with a target species. The selective interaction that gives rise to the response relies on the recognition of the target species, or guest analyte, by the receptor motifs of the polymer, and it must be follow by a transduction process giving rise to an easily measurable change.[12,13] The most common polymeric materials or families employed as sensors are the following: molecularly imprinted polymers, polymeric nanocomposites and hybrid polymers, acrylic polymers, conjugated or conductive polymers and polymers with chiral motifs. Although many sensory devices are based on a unique polymer family, it is usual to find sensory devices in which different polymers are combined, giving rise to sensory arrays, which have gained a lot of attention in the last years to improve both selectivity and sensitivity. For this reason, we conclude the section by listing some references in which the reader will find interesting information about these combinations of sensory polymers. This section shows the most usual target species and their corresponding type of polymeric sensor, together with the description of the main sensory devices that have been developed in the last years. The description of the different polymers employed as sensory materials, including a brief description of their chemistry, synthesis and their importance as sensors is listed below. We will also include some

bibliographic references in which the reader will find additional information about the different materials.

Molecularly Imprinted Polymers (MIPs): The preparation of this kind of polymers is carried out by polymerization of monomers and target molecule, which is used as template. Then, this molecule is removed giving rise to spaces with specific shape that can act as specific receptor sites for the target. The use of MIPs in different applications has been described previously by different authors.[14,15] The chemistry of the MIPs, attention is drawn to a number of factors pertaining to the template molecule and the selection of suitable functional monomers, cross-linkers, solvents, initiators and general polymerization procedures. Functional monomers can be acidic (methacrylic acid), basic (4-vinylpyridine) or neutral (acrylamide), and they are responsible for the binding interactions in the imprinted binding sites, whereas cross-linkers are important in controlling the morphology of the polymer matrix, stabilizing the imprinted binding site and also imparts mechanical stability to the polymer matrix. Solvents bring all the components in the polymerization into one phase, and also they are responsible for creating the pores in macroporous polymers. The importance of MIPs as sensors lies in the high selectivity of these "plastic antibodies"[16] and that a great number of monomers can be used to detect specific target molecules, thus offering many sensory possibilities. A classical review concerning the synthesis and characterization of MIPs was presented by Cormark and Elorza in 2004,[17] and also more recent information can be found in the review published by Chen *et al.* in 2016.[18]

Polymeric nanocomposites and hybrid polymers

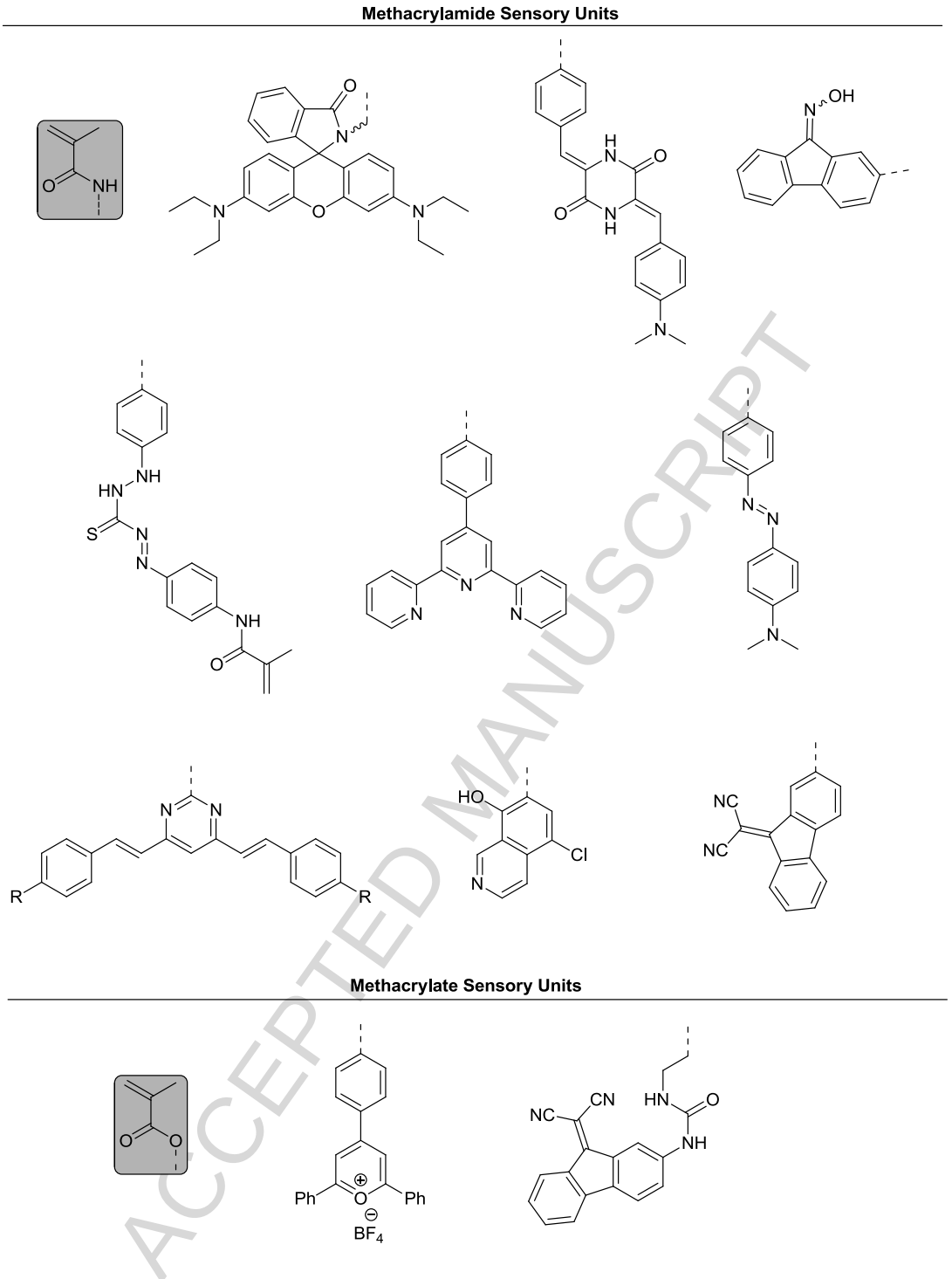
The preparation of polymeric nanocomposites starts from a mixture of a polymeric moiety with inorganic particles or units of small size (1-100 nm), which forms two different phases. On the other hand, hybrid polymers are also formed both by organic and inorganic moieties linked covalently to constitute one unique structure.[19] Polymer nanocomposites are obtained

through simple polymerization techniques in which inorganic particles, such as carbon nanotubes, graphene or metallic nanoparticles are added in-situ to the polymerization, then avoiding complicated synthesis processes. Then, the obtention of polymer nanocomposites is relatively simple, thus attracting great attention and consequently, offering a good number of sensory properties. An interesting review was presented in 2005 by Jordan *et al*. [20] and a more recent review was published by Lee *et al.* in 2018. [21]

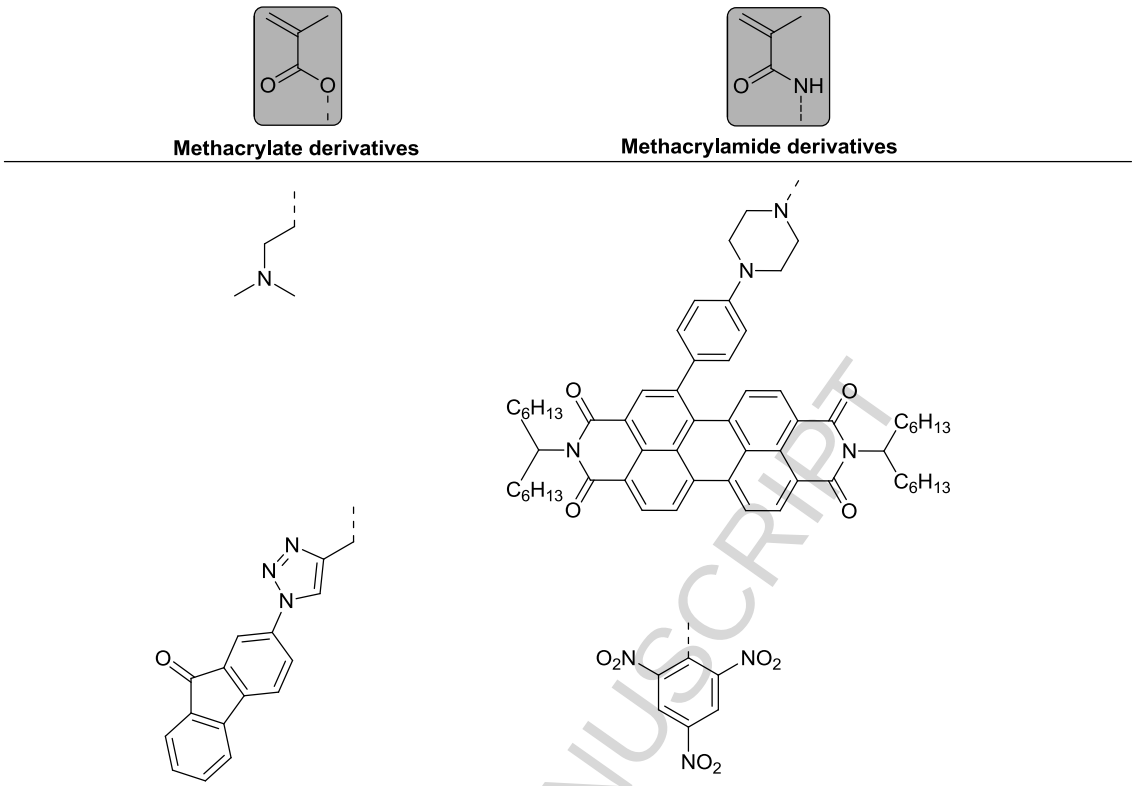
Concerning hybrid polymers, a great number of articles have been published in the last years showing different alternatives to obtain one-phase materials or hybrid polymers, including classical works in which block copolymers or elastomers are combined to metallic particles, such as molybdenum.[22,23] Precisely, this versatility offers different options to obtain sensory materials in which the polymer act as a matrix of inorganic sensory moieties.

Acrylic polymers

The main acrylic polymers found in sensory applications are polymer prepared with esters of acrylic or methacrylic acid, acrylamide derivatives, and copolymers thereof. Acrylic polymers are widely used as sensory materials due to their versatility, and the possibility of using a great variety of sensory units anchored chemically to their structure. A classical work was presented by Saunders in 1988[24] and a more recent work was presented by Ballard and Asua,[25] in which a description of the main acrylic polymers and derivatives (acrylic acid, methacrylic acid, esters of acrylic acid and of methacrylic acid, acrylonitrile, acrylamide or cyanoacrylates) is presented. Also, different acrylic copolymers are acrylic-ethylene, styrene-acrylonitrile, acrylonitrile-butadiene-styrene or acrylonitrile-butadiene copolymers. Concerning their synthesis, atom transfer radical polymerization is employed in most of the cases.[26] Focusing in sensory applications, In **Schemes 1** and **2** we present different monomers used to prepare polymers for charged and non-charged species.



Scheme 1. Acrylic sensory monomers used for preparing sensory polymers for the detection of charged species.

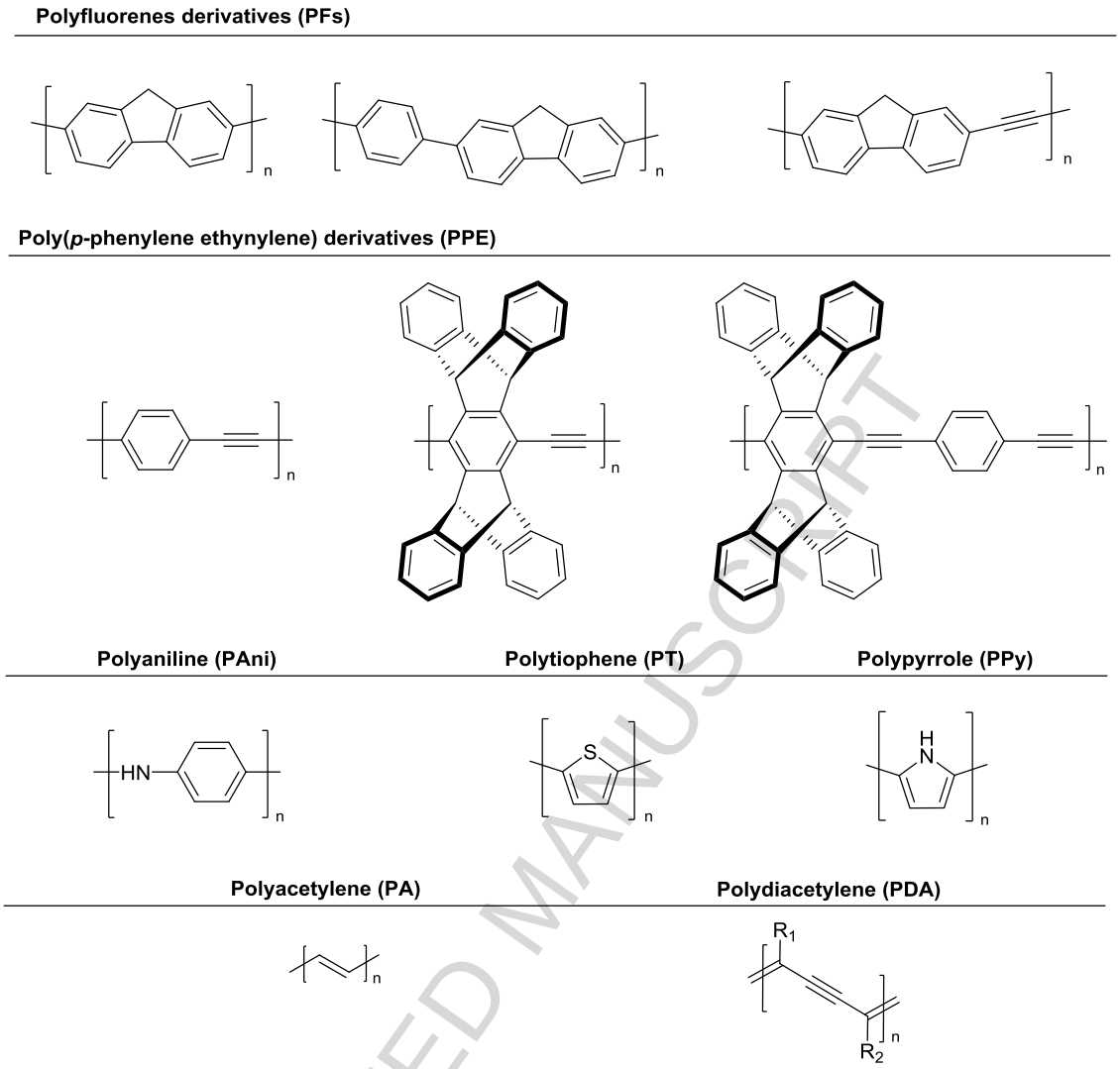


Scheme 2. Acrylic sensory monomers used for preparing sensory polymers for the detection of neutral species.

Conjugated or conductive polymers (CPs)

Main polymer chains alternating simple and multiple bonds are insulating or semiconductor polymeric materials that can be easily converted to conductor materials by doping. They are luminescence and this property, along with the electrical signal derived from the (semi)conductivity, can be used as output signal in sensory devices. Doping consists, essentially, in raising the conductivity of certain organic polymers metallic levels by chemical or electrochemical 'π-doping' (oxidation), or 'n-doping' (reduction). This process is phenomenologically similar to the doping of a classical inorganic semiconductor in that very large increases in conductivity are observed when the material takes up very small amounts of certain chemical species, which are precisely the target molecule.[27] The main conducting

polymers are fluorene derivatives (PFs), polyaniline (PAni) or polypyrrole (PPy) (**Scheme 3**).[28] Their importance in the sensory research field lies in the fact that conduction is mostly associated to a change in an easy-detectable property variation, such as color or electrical conductivity, then allowing a simple detection and quantification process during the sensing effect. Also, conducting polymers can be co-polymerized in an easy way with acrylic polymers, anchoring the monomer of the conjugated polymer to the acrylic matrix through free radical polymerization processes.[29]

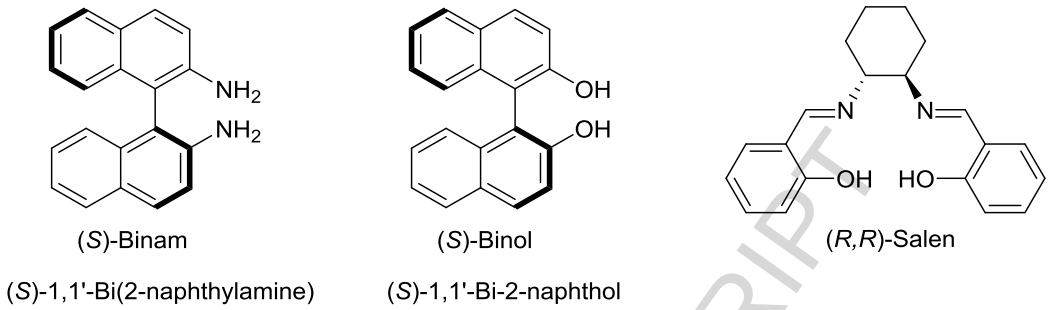


Scheme 3. Common CPs used as polymeric sensors

Polymers with chiral motifs

In the last decade, numerous chiral ligands have been introduced in the polymer main chain to achieve chiral recognition of important chiral species. Chirality confers optical response to the polymer, in presence of a target molecule. Then, the synthesis of polymers with chiral motifs is very interesting to obtain polymeric sensors in which optical properties vary when detecting the target specie. A classical review concerning the synthesis and properties of chiral polymers was published by Okamoto in 2000.[30] A description of the different ligands that are

currently been employed to obtain sensory polymers, mainly in luminescence and colorimetric sensors, can be found in several literature works.[31,32] Some of these chiral ligands, which are particularly interesting, are represented in Scheme 4.



(*S*)-Binam (*S*)-1,1'-Bi(2-naphthylamine)

(*S*)-Binol (*S*)-1,1'-Bi-2-naphthol

(*R,R*)-Salen

Scheme 4. Examples of chiral ligands used for preparing sensory polymers for chiral recognition.

3. Detection of target species

In this section, we will describe the different target species that can be detected using sensory polymeric materials. There are different groups of targets which will be currently being analyzed: In section 3.1, we will study the detection of metallic cations and anionic species; section 3.2 will be focused in the sensing processes of gases and volatile organic compounds (VOCs); in section 3.3 we will study a research line which has gained interest in the last years, related to the detection of explosives and chemical warfare agents (both in solution and in gas phase); section 3.4 will deal with the last developments in the field of biological and biomedical applications; the detection of humidity, herbicides and amine vapors will be briefly described in section 3.5, and finally, in section 3.6 we will group some recent research works focused in the detection of radionuclides and bacteria.

The sensing process is based on the change in different physical and chemical properties of the polymers, which vary in the presence of the target species. The polymer response allows us to classify the different sensors in several groups, as it is described as follows: First, resistive

sensors are based on changes in electrical resistance due to the surface reaction with different chemicals, detecting several target gases such as NH_3, H_2S or CO. In this sense, especially organic resistive sensors using conducting polymers (CPs) offer great potential to produce flexible, low-cost and lightweight devices with good mechanical properties and tunable electrical conductivity.[33-35] The resistive sensors are mainly employed in the detection of gases and VOCs, as we will detail in section 3.2.

Another typical sensory response is related to changes of colorimetric, luminescence (fluorescence or phosphorescence) properties of polymers when they interact with target species. In general, changing of color depends on selective chemical reactions (chemodosimeter approach, irreversible) or specific interactions based on feeble interactions, such as hydrogen bonds (chemosensor, reversible). The color change can be easily identified by naked-eye or using simple colorimetric techniques (RGB determination).[36] In CPs, the change of color depends on different external interactions (heat, redox processes or doping), and can be used to detect different VOCs.[37] In fluorescence sensors, the sensory behavior includes a receptor which recognizes the target and a fluorophore responsible of the fluorescence signal. In contrast to colorimetric sensors, the sensing must be quantified using fluorescence spectrometers, in terms of two different mechanisms. First, the turn-off or quenching of the luminescence, in which the interaction between the target and the sensory material deactivates the luminescence effect.[38] Also, the detection process can activate (turn-on) or deactivate (quenching) the luminescence of the sensory motif.[39-42] The optical response (colorimetric and luminescence) in sensory polymers is widely exploited in the detection of heavy metallic cations and anionic species, especially in aqueous solution, as detailed in next section.

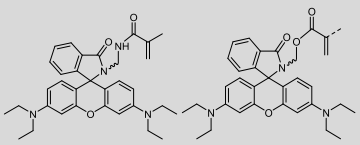
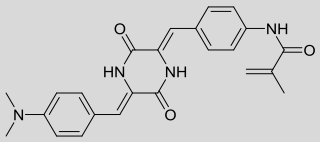
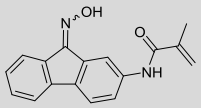
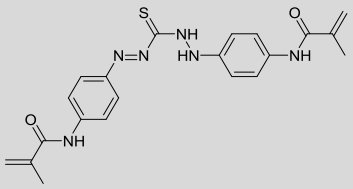
3.1 Detection of metallic cations and anionic species

Metallic cations and anionic species are detected employed sensory materials based on conducting, hybrid and acrylic polymers. For example, sensors based on PAni with species such as dithioaniline or methylene blue are used to detect Hg^{2+} through resistive measurements with detection limits in the range of ppb.[43,44] The detection of Hg^{2+} by turn-off fluorescence, using units or conjugated polymers is also described in different works[45,46] and the detection of Pd^{2+} and Pt^{4+} by UV-VIS or quenching fluorescence have been also deeply analyzed.[47,48] Cupper cations are detected employing different polymeric main structures such as polyacetylene, polypentiptycene ethynylene or polyfluorene derivatives using quenching fluorescence techniques, with detection limits in the range of ppm and ppb.[49-51] For the detection of Zn^{2+} distinct polymers containing *(R,R)*-salen-based moieties are used. These chiral ligands can show selective fluorescence response towards Zn^{2+} against other metallic cations.[52-55] Also, the detection of trivalent metal ions at low concentrations over monovalent and divalent metallic cations involves the synthesis of a chiral *(S)*-binaphthyldiamine-based fluorescent polymer sensor, which turns-on the fluorescence color at naked eye. Acrylic polymers have been extensively employed by Vallejos *et al.*[56] to detect mercury anions and concerning hybrid polymers. Li *et al.*[57] used D-π-A conjugated polymers based on benzo[c][1,2,5]selendiazole to also detect and quantify mercury ions.

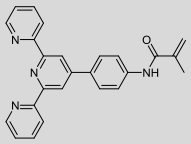
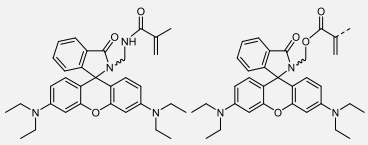
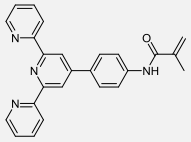
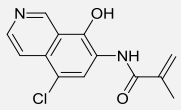
Anionic species, such as fluorides and cyanides, can be also detected using sensory polymeric materials. F^- anion is detected through the formation of coumarins in fluorinated poly(*p*-phenylene ethynylene) (PPE), observing a turn-on in the fluorescence signal,[58] and also from colorimetric changes detected by the naked eye in boron-dipyrromethene derivative polymers.[59] The detection of CN^- have been carried out using different polymers containing several backbone families, such as functionalized polyacetylene,[60] benzodiazaborole moieties[61] or polyfluorene with sulfate ions and diphenylmethylene malonitrile,[62,63] with detection limits in the range of ppb.

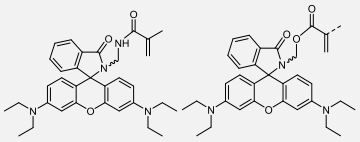
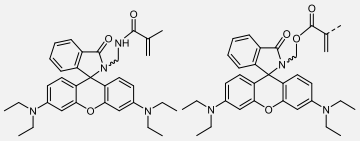
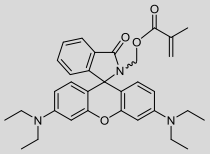
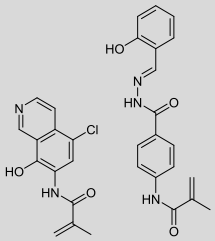
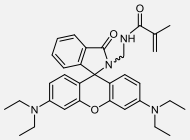
Tables 1 and **2** depict achievements published in the last years focused on the detection of metallic cations and anionic species, using the different polymers described previously in section 2. The table details the cation or anion detected, the type of polymer and the form in which is used during the sensing process, the composition of the sensory molecule and the type of physical/chemical response. In addition, when it is described in the original work, the sensitivity of the sensor is also showed.

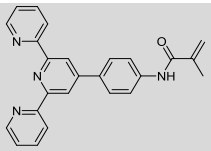
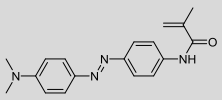
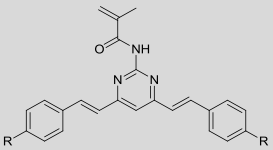
Table 1. Heavy metal and other cations detected using sensory polymers.

Target cation	Polymer type	Composition of sensory polymers or sensory monomers/sensory motifs of polymer chemosensors	Used as	Response	Sensitivity	Reference
Hg^{2+}	CP	PF + benzo[*c*][1,2,5]selenadiazole moieties	Solution	Fluorescence and UV/Vis variation	1.9 x 10^{-7} M	57
	Acrylic		Solution/film	Fluorescence and UV/Vis variation	10^{-6}-10^{-14} M	64, 65, 66
			Film	UV/Vis variation	10 ppb	56
			Film	UV/Vis variation	2.7 x 10^{-4} M	67
			Film	UV/Vis variation	1.6 ppb	68
	CPs	Main chain: Polyacetylene + benzo[2,1,3]thiadiazole	Solution	Fluorescence and UV/Vis variation	10^{-7} M	46

		PF derivative with dibutylaniline as binding unit	Solution	UV/Vis variation	20 mM	45
		PAni-Poly(2,2'-dithioaniline) or PAni-methylene blue	Film	Electrical	2.1 x 10^{-7} M	43,44
	Chiral	Main chain: Perylenyl + (*R,R*-salen) moieties	Solution	Fluorescence variation	7.28 x 10^{-7} M	69
		Main chain: PAni + *(S)*-Binol scaffold	Solution	Fluorescence variation	Not specified	70
	Epoxy	Poly-epoxy derivative with pendant triphenylamine units	Solid state /Solution	Fluorescence variation	Not specified	71
	Chiral	(*R,R*)-Salen + Polyacetylene, ethynylisoquinoline, 1,4-dioctyloxybenzene	Solution	Fluorescence variation	Not specified	52,53,54,55
Zn^{2+}	PEG	Acrylic quinoline	Solution	Fluorescence variation	3-14 nM	72
	Chiral array	*(R,R)*-Salen + *(S)*-Binol	Solution	Fluorescence variation	Not specified	73
	Chiral array	*(R,R)*-Salen + *(S)*-Binol	Solution	Fluorescence variation	Not specified	74
Cu^{2+}		PA + Imidazole pendant units	Film	Fluorescence variation	1.48 ppm	49
	CPs	Polypentypcene ethylene	Solution	UV/Vis variation	16.5 nM	50
		PF containing alkylsulfate chains	Soluti	UV/Vis	2.5	51

		Rodamine-based polymer	on	variation	µM	
	Acrylic		Solution	Fluorescence or UV/Vis variation	3.6 x 10^{-7} M	75, 76
			Film	UV/Vis variation	1.3 x 10^{-5} M	77
	Polyamide	*m*-phenylenediamine + aromatic diacid containing dipicolinic acid	Solution	UV/Vis variation	Not specified	78
	PEG	PEG+ Rhodamide	Coated filter paper	UV/Vis variation	2.5 x 10^{-8} M	79
			Solution/film	Fluorescence or UV/Vis variation	10^{-9}-10^{-14} M	80
	Acrylic		Film	Fluorescence variation	1.3 x 10^{-7} M	77
			Film	UV/Vis variation	142 ppb	81
Fe^{3+}	Epoxy	Epoxy derivative bearing 1-napthylamine units	Solution	Fluorescence variation	Not specified	82
	PS	PS derivative bearing dicyanomethylene groups	Solution	UV/Vis variation	Not specified	83
	Polyamide	*m*-phenylenediamine + aromatic diacid containing dipicolinic acid	Solution	Fluorescence variation	Not specified	78

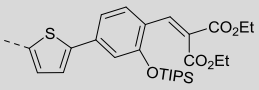
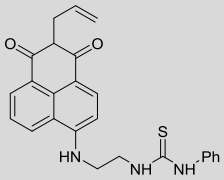
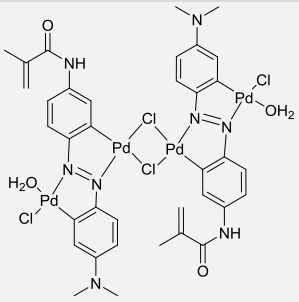
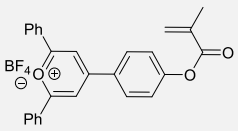
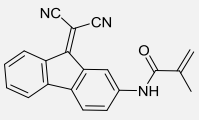
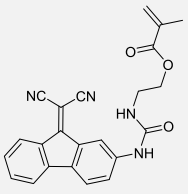
	Chiral	Polyamide + (*S*)-BINAM	Solution	Fluorescence variation	Not specified	84
	CP		Solution/film	Fluorescence and UV/Vis variation	10^{-6}-10^{-14} M	65,66
Pd^{2+}		Pyridine-based polymers	Solution	UV/Vis variation	1 ppm	47
Pd^{2+}, Pt^{4+}	CPs	2,6-dithienyl-4-phenylpyridine	Solution	UV/Vis variation	1 x 10^{-6} M	48
Cr^{3+}	Chiral	*(S)* – BINAM-based polymer	Solution	UV/Vis and fluorescence variation	Not specified	84
	CP		Solution/film	Fluorescence and UV/Vis variation	10^{-6}-10^{-14} M	65,66
Al^{3+}	Acrylic		Solution	UV/Vis and fluorescence variation	10^{-9}-10^{-14} M	66
			Film	Fluorescence variation	10 ppb	85, 86
Cr^{6+}	Acrylic		Film	UV/Vis variation	Not specified	80
	Polyamide	*m*-phenylenediamine + aromatic diacid containing dipicolinic acid	Solution	Fluorescent variation	Not specified	78

Co^{2+}, Sn^{2+}	Acrylic		Film	UV/Vis variation	6.4 x 10^{-8} M	77
Ag^{+}	PS	PS derivative bearing dicyanomethylene groups	Solution	UV/Vis variation	Not specified	83
U^{6+}	Resin	Poly[(4-methyl styrene)-co-(4-vinylbenzyl chloride)-co-(divinylbenzene)-co-(2-(1-napthyl)-4-vinyl-5-phenyloxazole)] bearing phosphoric groups	Solution	Fluorescence variation	Not specified	87
K^{+}, Na^{+}, Mg^{2+}, Cd^{2+}, Mn^{2+}	Array	Different CPs	Solution	Fluorescence variation	< 0.125 µM	74
H^{+}	Acrylic		Film and Fabric	UV/Vis variation	Not specified	88
			Film and Fabric	UV/Vis variation	Not specified	89
	Polyamide	*m*-phenylenediamine + aromatic diacid containing *N,N*-dimethylaminoazobenze	Film and Fabric	UV/Vis variation	Not specified	88

PA – Polyacetylene ; PF – Polyfluorene ; PS – Polystyrene ; PEG – Polyethylene glycol; BINAM - Binaphthyldiamin

Table 2. Anionic species detected using sensory polymers.

Target	Polymer type	Composition of sensory polymers or sensory monomers/sensory motifs of polymer	Used as	Response	Sensitivity	Reference

anion		chemosensors				e
F^-	PPE		Solution	Fluorescence variation	Not specified	58
	PF	BODIPY derivative	Solution	UV/Vis and fluorescence variation	Not specified	59
	Chiral motif	(*S*)-BINAM	Solution	UV/Vis and fluorescence variation	Not specified	90
	A vinyl styrene with sensory unit		Film	UV/Vis variation	Not specified	91
CN^-	Hybrid		Film/ fabric	UV/Vis variation	Not specified	92
	Acrylate		Film	UV/Vis variation	4×10^{-3} M	28
	Acrylamide	 	Film	UV/Vis variation	13-260 ppb	93, 94
	CP	PA + Imidazole pendant units	Film	UV/Vis variation	Not specified	49

Poly (Bendodiazo borole) with sulfonate groups	Solution	Fluorescence and UV/Vis variation	2 mM	61
PF bearing sulfate groups	Solution	Fluorescence variation	6 µM	62
PF with diphenylmethylene malonitrile	Solution	Fluorescence variation	0.5 µM	63

PPE – Poly(*p*-phenylene ethynylene) ; PF – Polyfluorene ; BODIPY – Boron dipyrromethene ; PT - Polythiophene

3.2 Detection of gases and volatile organic compounds (VOCs)

Another possible application of polymeric sensors is the detection of different non-harmful (e.g., CO_2) and harmful gases (e.g., NH_3 or chlorinated gases), volatile organic compounds, (mainly alcohols such as methanol, 2-propanol and 1-octanol), and also, the detection of H_2O molecules (humidity). In this case, the sensing response can be detected by means of different responses, including piezoelectric behavior,[95] variations of electrical impedance and resistance[96] and cyclic voltammetry when employing electrochemical measurements.[97] In addition, it is possible to design self-encoded polymer films based on resins spectroscopically active to detect other gases such as hexane, methylamine, methanol or acetonitrile nanotubes.[98]

In addition, fluorescence response is obtained using a branched PEI modified with tetraphenylethylene sensor when CO_2 is bubbling in an ethanol solution over other gases such as CO, SO_2, H_2S or other volatile organic compounds.[99] Humidity can be measured employing films prepared from the copolymerization of divinylbenzene and 4-vinylpyridinium salt derivative with a time response of only a few seconds.[100]

Table 3 details some works published in the last years focused on the detection and quantification of non-harmful gases and volatile organic compounds.

Table 3. Gases and Volatile Organic Compounds detected using sensory polymers.

Target species	Polymer type	Composition of sensory polymers or sensory monomers/sensory motifs of polymer chemosensors	Used as	Response	Sensitivity	Reference
NH_3	Nanocomposites	PEI +GO	Modified electrodes	Cyclic voltammetry	5 mM	101
		PPy + Pd nanoparticles	Film	Resistance variation	40 – 300 ppm	102
		PDDA/PSS + MoS_2/ZnO	Film	Resistance variation	12 ppb	103
	CP	PS-PVP nanomaterials	Fibres	Resistance variation	1-50 ppm	35
		PPy + Cobalt nanoparticles	Fibres	Impedance variation	Not specified	104
Humidity	Nanocomposites	PPy + FeO_3 or ZnO or TiO_2 nanoparticles	Pellet/Film	Impedance variation	0.1 % RH	105,106, 107
		PSS + ZnO nanoparticles	Film	Impedance variation	2 % RH	96
	Copolymer	Divinylbenzene and 4-vinylpyridinium salt derivative	Film	Impedance variation	1 % RH	100
CO_2	Graft copolymer	Poly(ethyleneterephtalate) with 1-(4-vinylbenzyl)-1H- imidazole	Membrane	Resistance variation	Not specified	108
	Branched copolymer	PEI	Branches	Resistance and fluorescence variation	Not specified	99,109
VOCs	Nanocomposites	PPy+MoO_3	Film	Resistance variation	Not specified	110,111
		PAni+SnO_2	Fibres	Cyclic voltammetry	500 ppm	97
		Polythiophene	Film	Piezoelectric	≈ ppm	95

	CP	PPE [1)]	Film	Fluorescence variation	Not specified	112
	Array	Styrene monomers derivatives	Film	FTIR and RAMAN spectra	Not specified	98
		Acrylic and and vinyl polymer	Chip	Resistance variation	1 – 1000 ppm	113
NO_x	Nanocomposites	PPy + ZnO	Nanoparticles	Cyclic voltammetry	1000 ppm	114
		PAni or PT + SnO_2	Nanoparticles	Cyclic voltammetry	≈ ppm	115,116
Aliphatic Isocyanates	CP	Pentiptycene and tetraphenylethylene moieties	Film	Fluorescence variation	≈ ppm	117
Chlorinated gases	Hybrid polymer gel	Co^{2+} cation ions + tetrazole derivative	Gel	Fluorescence variation	≈ ppb	118
DMMP	Array	Vinyl and condensation polymers	Fabric	Resistance variation	≈ ppm	119

PEI – Polyethylenimine ; GO – Graphene oxide ; PAAC – Poly(acrylic acid) ; PSS – Poly(sodium styrene sulfonate) ; PVP – Poly(vinyl pyrrolidone) ; RH – Relative humidity ; PT – Polythiophene ; DMMP - Dimethyl methylphosphonate ; PPE – Poly(*p*-phenylene ethynylene) ; [1)] with rotaxane motif

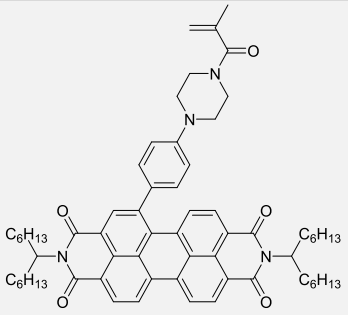
3.3 Detection of explosives and harmful substances

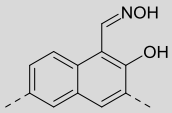
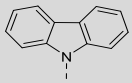
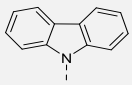
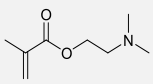
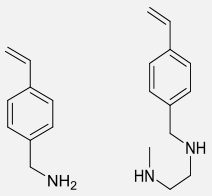
Detection of explosives and harmful substances, mainly in gas phase, is one of the applications of sensing polymers. A very interesting investigation line points out the use of

sensory materials prepared from CPs to detect vapors and explosives of different nature. A film-shaped polymeric sensor formed of pentiptycene and tetraphenylehylene moieties linked by acetylene have been designed to detect nitroaromatic explosives in aqueous and gas media at ppb level by quenching fluorescence.[120] Also, dinitrobenzene detection in liquid media can be carried out in few minutes with a microstructured optical fiber adhered to a poly(phenylenevinylene) derivative.[40] Hydrazine vapor can be detected using a film based in different poly(phenyl ethers) (PPEs), varying in their degree of electron density along the polymer chain.[121] As said before, CPs are widely used to detect harmful substances and explosives. For example, a recent research work[122] explores the use of CPs containing oxime derivatives as chromophores to detect organophosphorus agents. The use of acrylic polymers in the form of films to detect TNT vapors and in aqueous solution has been also analyzed deeply by Pablos *et al.*[123,124]

In **Table 4** we resume the most important research works concerning the detection and quantification of explosives and harmful substances using polymeric sensors.

Table 4. Explosives and harmful substances detected using sensory polymers.

Target species	Polymer type	Composition of sensory polymers or sensory monomers/sensory motifs of polymer chemosensors	Used as	Response	Sensitivity	Reference
PETN/ RDX/TNT	Array / electronic nose	Mercaptobenzoic acid	Nanocomposite	Piezoelectric	≈ ppt	125
TATP	Acrylic		Film	UV/VIS variation	3.3 mM	126

DCP	Poly(arylene vinylene)		Film	UV/VIS variation	Not specified	122
Hydrazine	PPE	Pentiptycene moieties	Film	Fluorescence variation	100 ppb	121
DNT	Polyacetylene	Pentiptycene and tetraphenylethylene moieties	Film	Fluorescence variation	≈ ppb	120
	Poly(phenylene vinylene)	Poly[2-methoxy-5-(2-ethylhexyloxy)-1,4-phenylenevinylene]	Fibres	UV/Vis variation	6.3 ppm	40
	Polysiloxane or polyphenylsilane		Film	Fluorescence variation	Not specified	127
	Polysiloxane or polyphenylsilane		Film	Fluorescence variation	Not specified	127
	Nanocomposites	PPV + Silicon microcavities PPV + GO	Thin film	Photoluminescence variation	1.3 x 10^{-3} M	128, 129
TNT	Acrylic		Film/fabric	UV/VIS variation	10^{-3}-10^{-6} M	123, 130
	Array	Polythiophene derivatives and poly[phenylenevinylene] derivatives	Film	Fluorescence and colorimetric variation	1 – 100 µM	131, 132
	Vinyl-based polymers		Film/fabric	UV/VIS variation	10^{-4}-10^{-6} M	123, 124

Array / electronic nose	Mercaptobenzoic acid	Nanocompos ite	Piezoelectric	≈ ppt	125

PETN - Pentaerythritol tetranitrate ; RDX – 1,3,5-trinitro-1,3,5-triazinane ; PPE – Poly(phenyl ether) ; TATP – Triacetone triperoxide ; DCP – Diethyl chlorophospate ; TNT – 2,4,6-trinitrotoluene

3.4 Detection of biomolecules

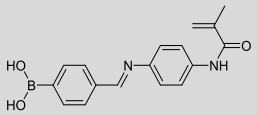
Several biomolecules can be detected employing sensors based on conjugated polymers. Sensors systems formed of amine or carboxylic acid-terminated polydiacetylene films or wire-shaped have been used to determine the presence of cyclodextrin and streptividin.[133] Dopamine can be also detected using poly(4-vinylpyridine) films.[134] Other species such as aminoacids and neurotransmitters of X-linked inhibitors of apoptosis protein are detected by polyacetylenes (PDA).[135] Folic acid can be detected using a sensor based on MIP technique with the acid as template. The MIP is formed by electropolymerization of the thermosensitive amine-terminated poly(*N*-isopropylacrylamide) with *o*-phenylenediamine and *N,N'*-methylenebisacrylamide as cross-linker giving rise to an on-off-switchable sensor which changes are measured employing cyclic voltammetry and differential pulse voltammetry.[136] Poly(hydroxyethyl methacrylate-*N*-methacryloyl-(*L*)-tryptophan methyl ester) has been employed to prepare a nanofilm with myoglobin as template on the gold surface of a surface plasmon resonance sensor. The sensor shows high selectivity and a sensitivity of 26.3 ng/mL for myoglobin, distinguishing among other species such as lysozyme, cytochrome and bovine serum albumin. The analyte has been detected and measured also in real samples from myocardial infarction patients.[137] Testosterone is also possible to detect by its ability to bind to a MIP film prepared from acrylic acid, pentaerythritol triacrylate as cross-linker and the steroid as template through micropatterning by interference photolithography method.[138] Melamine

in milk samples can be detected with a detection limit of 1.55×10^{-4} M with a fluorescent MIP chemosensor based on a rhodamine derivative.[139]

Table 5 details the literature review concerning the detection and quantification of biomolecules using sensory polymers.

Table 5. Biomolecules detected using sensory polymers.

Target biomolecule	Polymer type	Composition of sensory polymers or sensory monomers/sensory motifs of polymer chemosensors	Used as	Response	Sensitivity	Reference
Drugs						
Fluconazol	Nanocomposite	PVC/Cyclodextin derivative Surface + Fe_3O_4 nanoparticles	Electrode	Voltammetry	10^{-11} M	140
Phenorbital	MIP/Nanocomposite	Methacrylic acid + nickel nanoparticles	Electrode	DPV	≈ nM	141
Chlortetracycline	MIP	*o*-phenylenediamine	Electrode	CV / DPV	Not specified	142
Oxytetracycline	MIP	*o*-phenylenediamine	Electrode	CV / DPV	3.3×10^{-10} M	143
Tetracycline	MIP	Acrylic acid and acrylamide	Film	Bragg Diffraction	Not specified	144
Hormones						
(*D*)-Thyroxine	MIP	Acrylic derivative	Nanowires	DPASV	10^{-3} ng/mL	145
Testosterone	MIP	Methacrylic acid + Pentaerythritol triacrylate (cross-linker)	Film	Bragg Diffraction	Not speci	138

					fied	
Neurotransmitters						
Epinephrine	Nanocomposite	*N*-hydroxyphenylmaleimide + MWCNTs	Electrode	DPASV	0.002 ng/mL	146
	Nanocomposite	*o*-phenylendiamine + DNA and gold nanoparticles	Electrode	DPV	≈ nM	147
Dopamine	Acrylic		Film	Fluorescence variation	$4x10^{-4}$ M	148
	Hybrid polymer	Poly(4-vinylpyridine) and zinc dendritic porphyrin	Film	Fluorescence variation	Not specified	134
Sugars/Saccharides						
	Nanocomposite	PPy + MWCNTs	Film	Amperometry	Not specified	149
Glucose	Nanocomposite	Poly(vinyl alcohol) derivative + SWCNTs	Hydrogel	Raman spectra variation	Not specified	150
	Nanocomposite	Polyacrylic acid + Cu Nanoparticles + graphene	Electrode	Amperometry	0.08 µM	151
	CP	PPy	Film	Cyclic voltametry	0.5 mM	152
Glucose, Fructose, Sucrose	Acrylic	Polyacrylamide + Poly(*N*-acryloyl-*m*-aminophenylboronic acid)	Gel	UV/Vis variation	≈ mM	153, 154, 155,

						156
Cyclodextrin	CP	PDA	Membranes/Chips	Fluorescence variation	1 ng/mL	133
(*D,L*)-Arabitol	MIP	Poly(2,2'-bithiophene-5-boronic acid)	Film	Piezoelectric effect	≈ µM	157
Aminoacids						
(L)-Methionine	MIP	PolyBenzidine with carboxylated CNTs	Fibres	DPV	10^{-3} ng/mL	158
(D, L)-Phenylalanine	MIP	Carboxy derivatized bis(bithiophene)	Film	DPV	13 µM	159
(L)-N-Buthoxycarbonyl-alanine	Chiral/Hybrid	5,5'-((2,5-dioctyloxy-1,4-phenylene)bis- (ethyne-2,1-diyl)bis(2-hydroxy-3-(piperidin-1-ylmethyl)-benzaldehyde + *S*-BINAM	Solution	Fluorescence variation	Not specified	160
(D)-Phenylalaninol	Chiral	5,5'-((2,5-dibutoxy-1,4-phenylene)bis(ethyne-2,1-diyl))bis(2-hydroxy-3-(piperidin-1-ylmethyl) benzaldehyde + *S*-BINAM	Solution	Fluorescence and UV/Vis variation	Not specified	161
(L)-Phenylalaninol	Chiral	PAni + *S*-BINOL	Solution	Fluorescence variation	Not specified	70
Alaninol	Chiral	2,5-diiodo-1,4-dioctyloxybenzene + *S*-BINAM	Solution	Fluorescence variation	Not specified	162
Metabolites						
(L)-Lactic and *(L)*-Mandelic acids	Chiral	5,5'-((2,5-dibutoxy-1,4-phenylene)bis(ethyne-2,1-diyl))bis(2-hydroxy-3-(piperidin-1-ylmethyl) benzaldehyde + (*R,R*)-Salen	Solution	Fluorescence and UV/Vis variation	Not specified	163
(L)-Malic acid	MIP	4-gluconamido phenyl methacrylate	Coating	Piezoeletric effect	0.2 ng/m	164

					L	
Folic acid	MIP	*o*-phenylenediamine, Amine-terminated poly(*N*-isopropylacrylamide) and (*N,N'*-methylenebisacrylamide)	Electrode	CV/DPV	0.9 µM	136
Other biomolecules						
Myoglobin	MIP	Poly(hydroxyethylmethacrylate-*N*-methacryloyl-(*L*)-tryptophan methyl ester	Film	Optical reflectivity	26 ng/mL	137
Streptavidin	CP	PDA-Biotin	Membrane	Raman spectra variation	1 ng/mL	133
Cholesterol	Nanocomposite	PAni + MWCNTs	Electrode	Cyclic voltammetry	1.3-13 mM	165
Creatinine	Nanocomposite	creatinine amidohydrolase, amidinohydrolase and sarcosine oxidase + PANI + Carboxylated MWCNTs	Electrode	CV	0.1 µM	166
Adenine	Hybrid polymer	Polydimethylsiloxane with Zn acyl hydrazine complex	Film	Luminescence variation	90 ppb	167
Melamine	MIP	Combination of rhodamine derivative and methacrylic acid	Solution	Fluorescence variation	10^{-4} mM	139

DVP - Differential pulse voltammetry ; CV - Cyclic voltammetry ; DPASV - Differential pulse anodic stripping voltammetry ; PDA - Polydiacetylene ; MWCNT – Multiwalled carbon nanotubes

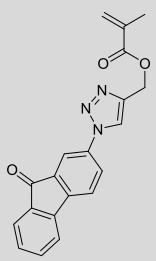
3.5 Detection of humidity, herbicides and amine vapors

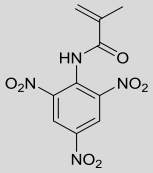
In this section, we list some other interesting works focused in three different applications which are also of growing interest. First, the detection of humidity, essentially through the employment of highly-sensitive hydrophilic polymers. Secondly, the detection of emerging pollutants, such as herbicides, and finally, the detection of amine vapors. For example,

herbicides and insecticides can be sensed in the environment, avoiding the laboratory tests, and allowing performing in-situ experiments. In this context, the herbicide alachlor is recognized by a MIP sensor designed from 2-acrylamide-6-methoxybenzothiazole, through the quenching of the fluorescence, detecting traces in corn seed samples against other competing molecules such as cyanazine and cypermethrin with a detection limit of 0.5 µM.[168] Another example of herbicide detection consists in using allyl fluorescein as functional monomer to prepare fluorescent MIP microspheres-shaped for the optical detection of ultratraces of fenvalerate and λ-cyhalotrin in water samples with a sensitivity of 0.068 µM and 0.0037 nM, respectively.[169,170] Other applications include the sensing of humidity,[171-173] or the detection of amines using fluorescent hybrid polymers.[174]

Table 6 lists some of the recent research works focused on the detection of H_2O molecules, herbicides and amines using polymeric sensors.

Table 6. Detection of humidity, herbicides and amines using sensory polymers.

Target species	Polymer type	Composition of sensory polymers or sensory monomers/sensory motifs of polymer chemosensors	Used as	Response	Sensitivity	Reference
H_2O	Nanocomposite	PU+ CNTS	Film	Resistivity variation	Not specified	171
	Nanocomposite	PU + rGO	Film	Resistivity variation	Not specified	172
	Acrylic		Film	UV/Vis variation	10^{-3}-10^{-5} M	173
Herbici	MIP	Allyl fluorescence	Nanop	Fluorescenc	0.068	169

des			articles	e variation	µM	
	MIP	Polysilane	Nanoparticles	Fluorescence variation	0.0037 nM	170
	Nanocomposite	Methacrylic acid, *N,N'*-(methylene)-bisacrylamide + NiO nanoparticles	Film	Cyclic Voltammetry	0.5 µM	168
Amines	Hybrid	Polyfluorene/Pd tetraphenylporphyrine	Film	Fluorescence variation	Not specified	174
	Acrylic		Film/fabric	UV/Vis variation	Not specified	175

PU - Polyurethane ; CNTS – Carbon nanotubes ; rGO – Graphene oxide

3.6 Detection of radionuclides and bacteria

Radionuclides can be detected thanks to sensory polymers. For example, uranium traces can be quantified using functionalized copolymers,[176] a second paper published by Losno *et al.*[177], presenting an interesting experimental design to adjust the morphology of a polymer poly(EDMA-*co*-AMA) to tailor impregnated polymers specific for radionuclides separation in microsystems, and finally, a third paper in which an ion imprinted polymer based sensor is employed for monitoring toxic uranium in environmental samples, reaching sensibilities around $2x10^{-8}$ M.[178]

The second group of polymeric sensors deals with the detection of bacteria using polymer nanocomposites. Different works have been published in this research field. In this review we present three papers published in the last years that will be an starting point for the readers. Arshak *et al.*[179] presents the characterization of different polymer nanocomposite sensors for the quantification of bacterial cultures, mainly food pathogens such as Salmonella

or Bacilus Cereus. Another interesting paper was presented by Suma *et al.* in 2017,[180] reporting the optoelectronic sensors based on poly vinyl alcohol and silver nano colloids, employed in the detection of the E.Coli bacteria by means of electro voltammetry measurements. Finally, we present another paper published by Mei *et al.*[181], in which polymeric-Ag nanocomposites are used as effective antimicrobial sensors against different Gram-positive and Gram-negative bacteria.

Table 7 lists some of the recent research works investigating the detection of radionuclides and bacteria.

Target species	Polymer type	Composition of sensory polymers or sensory monomers/sensory motifs of polymer chemosensors	Used as	Response	Sensitivity	Reference
Radionuclides	Functionalized copolymer	Poly(4-methylstyrene)-*co*-(4-vinylbenzyl chloride)	Solid resin	Online flow-cell detector	0.8 mMol/g	176
	Acrylic	Methacrylate based copolymer	Porous resin	Chromatography	75 mg Uranium/g resin	177
	MIP	Polyvinyl chloride matrix	Electrode	Resistivity variation	10^{-8} M	178
Bacteria	Nanocomposite	Polyaniline and polyethylene adipate	Electrode	Cyclic voltammetry	Not specified	179
		Poly vinyl alcohol	Electrode	Cyclic voltammetry	Not specified	180
		(Dimethylamino)ethyl methacrylate based	Film	Fluorescenc	0.5 µg/mL	181

copolymers	e variation

4. Micro and nano sensory devices based on smart polymers

Sensory polymers are being currently used in different devices, which essentially are focused on detecting different physical properties (electrical conductivity, optical properties, piezoelectricity, etc.) which change due to the sensing effect. The quantification of these properties is related directly to the amount (sensitivity) and the type (selectivity) of the target species. In this second part of this review, we detail the different types of devices in which smart polymers are employed as sensory materials. The devices can use different polymeric-based materials. First, the use of fibers and nanofibers takes advantage of the enhanced surface area. Other devices employ membranes and thin films, in which the main advantage is their easy processability. The third group analyzed is the sensory arrays, extensively used in electronic noses, in which the combination of different polymers increases the selectivity and sensitivity of the sensory device. The fourth group described is based on quartz crystal micro balances (QCMs), in which the piezoelectric effect leads to an easy processable electrical signal, then simplifying the quantification process. Next, sensory devices based on microfluidic devices have gained a lot of interest in the last years due to the possibility of operate on microscale and nanoscale liquids for sensing purposes, with lower economic costs and also processing advantages. Finally, the last group of devices employ the analysis of electrochemical properties of sensing polymers throughout the use of different modified-electrodes in which the polymer is deposited, a well-known technique extensively used since the 80's. We will also include a brief description of other devices with increasing interest, such as holographic sensors and sensory chips for biological and biomedical applications.

4.1 Fibers and nanofibers

These type of sensory polymers have gained a lot of interest due to the combination of high surface area, large porosity and interconnected porous structure. Sensors based on these nanomaterials present high selectivity and sensitivity, reversibility and also fast response time. Concerning sensory applications and devices, the nanofiber-based sensor materials present a large surface-area volume ratio, thus improving greatly the sensory characteristics compared to other polymeric sensors. Nowadays, there are four main nanofiber-based sensors, classified as a function of the detection mechanism: surface acoustic wave (SAW) sensors, resistive sensors, optical sensors and amperometric sensors. A very interesting review which resumes the main fabrication process and applications of sensory devices based on nanofibers was presented by Wang *et al.*[182]

In a SAW device, the sensing mechanism is related to the analysis of the change in the velocity of an acoustic wave on a piezoelectric substrate surface, which is caused due to the adsorption process of different analytes. Resistive sensors are based on changes in electrical resistance due to the surface reaction with different chemicals. Using this mechanism, ultrasensitive sensors can be prepared to detect several gases such as NH_3, H_2S or CO.

Optical sensors use the change in colorimetric, fluorescence or luminescence properties of smart polymers in presence of different analytes. The mechanisms of detection and quantification have been already exposed in previous section.

There are different applications of sensory devices based on nanofibers which are currently being investigated. For example, nanofibers have been used in amperometric biosensors. Especially glucose sensors have been developed in the diagnosis and treatment of diabetes, due to the high sensitivity and selectivity to glucose and glucose oxidase. Other analytes that can be detected with these devices are fructose, methanol, hydrazine, or different biotoxines, such as thyroxine and methionine.

4.2 Films and membranes

Due to their easy processability, polymers can be transformed into films, coatings or even finished materials with specific shapes. For this reason, a great number of sensory devices are prepared using these materials. In general, polymeric films are employed as a substrate in which the sensory monomer, or macromolecules with receptor motifs, is dispersed or anchored, generally in small proportions (up to 5 %. wt.). Then, polymerization processes, such as radical polymerization, are employed to prepare the sensory film, which is after employed in the detection, through different mechanisms (colorimetry, fluorescence, etc.). Films can be easily modulated in terms of flexibility, hydrophilicity or transparency, obtaining easy manageable sensory materials. Concerning the use of polymer coatings or thin films, the sensory device consists in a solid substrate in which a thin layer of the sensory polymer, or a polymer solution containing the sensory polymer, is deposited. The thickness of the coating layer is between 1 and 10 µm. The deposition takes place usually using spraying or sputtering techniques of the polymer solution using air brushes. Then, sensing procedure takes place analyzing the physical properties of the coatings in presence of different target species. Following this idea, an important research line concerns the use of conducting polymeric thin films in electrodes, to evaluate their sensing properties measuring their chemoresistive characteristics when they are exposed to different analytes. A detailed resume of the state-of-art concerning the use of polymeric thin films of sensory polymers can be found in several works published by Fittgerald and Mosbach.[98,183]

4.3 Sensory arrays

Another group of sensory devices employ the use of sensor arrays, which is especially interesting to analyse volatile analytes. A typical sensor array includes a number of cross-reactive sensors, which react individually to different analytes in various degrees. Using this disposition, it is possible to obtain, for example, the sensory response of an array to

spectrophotometer responses, generated at different wavelengths.[98] ***Although they cannot be considered as a proper and differenced polymer family, sensor arrays combine polymers of the previous families. Frequently, the response of only one type of polymer is not selective or sensitive enough for the application and it is necessary to combine some polymers to create one effective sensor. The combination of different polymers, increase the se***nsitivity ***and selectivity of the sensing procedure. As said before, sensory arrays are formed by a combination of different polymer families listed above, so it is normal to find in the literature sensory arrays based, for example, on conducting polymers or MIPs.*** [184,185]

Sensory arrays are finding interesting applications in many research fields. For example, in biomedicine and biological research lines, sensory chips arrays are used to detect different proteins and biomolecules to prevent the apparition of diseases, also using sensory polymer-nancomposite materials.[186] Also, the detection of explosives and hazardous substances is one of the key targets in this type of sensory devices, and it is possible to detect and quantify different species in water, including TNT, tetryl or nitroamides.

Recently, a specific type of sensory arrays has been developed. The electronic noses are devices based on smart polymers originally intended to detect odors and flavors. The technical and commercial developments of these devices have increased greatly in the last decade, as it has been described in the review published by Arshak *et al.*,[187] which describes the operating principles and fabrication methods of each sensor type and their main applications. However, an specific application which has attracted a lot of interest in the last years is the characterization of olive oil and wines.[188] Other recent applications that have emerged in the last decade are the analysis of food quality and safety (detecting traces of gases produced from rotten food), the sensing of metallic cations or the discrimination of different azo-compounds in water.

4.4 QCMs

Another type of sensory device which has been extensively used is the QCMs. In general, the use of QCMs in the polymeric sensors research provides more potential and presents many advantages which allow the elimination of the sophisticated preparation of samples and expensive off-site analysis. The sensory effect is produced due to the mass change of the polymer when the analyte is absorbed. The mass change produces a variation in the resonance frequency of the piezoelectric crystal, which can be easily measured and quantified. This electrical signal as output of the sensory effect allows enhancing sensitivity, shortening the response time, stabilizes reproducibility, etc. Thus, the use of appropriate coating materials on QCM sensor surfaces have played an important role on determining their final properties and applications. Thus, polymer coated QCM sensors were employed in the detection of organic vapors and harmful gases. In this approach, the QCM balance is coated with different sensory polymers which react against several volatile organic compounds (VOCs), then varying the resonance frequency of the quartz crystal.[189-191] New developments include, for example, the use of wireless resonant sensor as chemical detectors for environmental applications.[192] The biomedical and biological applications of QCM-based sensors have also attracted interest in the last years, such as in the immobilization of antibodies and DNA, and the detection of biological hormones.[193] Finally, it is important to remark the polymer coated QCM resonator devices are also employed currently in the detection of heavy metal ions (copper, cadmium or gold cations). This research line has been developed since 2010, with a great number of published works.[194-196]

4.5 Polymeric sensors based on microfluidic devices

The research and development of microfluidic and nanofluidic technologies, which began between 2000-2002, have also emerged in the last decade as a very interesting fabrication technique to obtain micro and nano sensory devices in which smart polymers are widely used. With this technology it is possible to operate on microscale and nanoscale liquids for sensing

purposes, with lower economic costs and also processing advantages.[197] Although the detection methods that can be carried out in these devices can be optical, electrochemical and mass spectrometry, electrochemical measurements enhance the selectivity and sensitivity. This method is based on the electrical modulation of the analyte species that undergo under redox reactions, and they are usually employed for the detection of the electroactive species, through cyclic voltammetry measurements.

Focusing in the applications of micro and nanofluidic devices, numerous works related to biological and biomedical sensing have been published in the last 10 years. For example, the detection of glucose using biocompatible chips,[155] the detection of different proteins[154] and biomolecules.[198] A review of the last developments in the biological and biomedical applications of the sensory systems integrated in microfluidic devices have been published recently, including the fluid body testing throughout the use of sensory chips.[199]

Other interesting applications follow the classical research lines exposed previously. For example, the detection of heavy metal ions, specially Cu^{2+} and Hg^{2+} is crucial for environmental and safety applications.[200] Also, and due to the importance of the analysis and control of environmental pollution analysis, especially in-situ measurements, this investigation line has gained interest, as it is detailed in the review recently published in this topic.[201]

4.6 Polymeric sensors based on modified electrodes

The next group pf sensory devices are related to the use of modified electrodes, in which analytical methods allows the utilization of electrochemical measurements (cyclic voltammetry) to determine the sensing effect by means of electrical signals. The characteristics of non-modified electrodes are improved through the chemical modification of the electrodes, a technique developed in the early 80's. The modification of these electrodes can be done in several ways and using different materials, as it has been related in the review

published in 2017 by Naveen et *al.*[202] Mainly, metallic or graphene electrodes are coated with sensory polymers (conducting polymers or polymer nanocomposites), then measuring the electrochemical properties of the modified device, correlating the voltammetry results directly with the detection and quantification properties of the sensor.

The applications of the modified electrodes have been basically described previously for the other sensory devices. The detection of heavy metal ions is a topic of interest using electrochemical sensors embedded in electrodes. Several cations and ions, such mercury or lead, can be detected and quantified using electrochemical sensors based on smart polymers at graphene modified electrodes.[43,44,203] Biological applications, mainly protein detection, has been also studied using these devices. For example, the electrochemical biosensing of glucose and lactic acid is reported using poly(ethylene) glycol diacrylate, fabricated by photolithography, as working electrodes.[204] A very interesting review focused on the DNA immobilization and detection using electrochemical measurements in modified electrodes have been reported recently.[205] Also different medications are detected using these sensory devices, such as anticancer drugs[206] or phenobarbital.[207] The detection of VOCs, such as phenol, ethanol or methanol is also analyzed using these sensory devices, and another interesting application of these sensory devices is the quantification of humidity, which is mainly carried out using conducting polymers.[106] At this point, it is important to remark that a recent review of the applications and characteristics of electrochemical polymeric sensory devices has been published in 2016, with special emphasis in biological and biomedical applications, which is one of the most promising research field nowadays.[208]

4.7 Emerging polymeric sensory devices

In this part of the section we include other specific sensory devices based on polymers, but with still incipient research that will surely growth in the next years: Sensory chips, sensory devices based on microporous and nanoporous materials, the use of piezo-resistive quartz

microcantilevers, sensors based on quartz tuning forks (QTFs), holographic sensors, thermo-responsive based sensory polymer devices, optical polymeric tweezers and finally polymeric switches.

As said before, due to their easy processability and sensing characteristics, smart polymers are ideal candidates to obtain handleable chips that can be used directly on human skin to carry out in-situ biological analysis, such as the detection of different proteins, the immobilization of biological markers of different types of cancer or simply the temperature monitoring. In this research field, the number of published paper is increased greatly in the last years, especially in the biological and biomedical applications.[209] Although a list of published papers will be detailed at the end of the section, it is important to remark the review published in 2015 by Chuan-Guang *et al.*, devoted to the analysis and detection of azobenzene derivatives using polymeric sensing chips.[210]

Using micro and nanoporous materials in sensory devices presents several advantages over traditional sensors, in which polymers are deposited on a flat substrate. For example, the nanoporous surface increases the interfacial area, which originates higher sensitivity. Different approximations have been carried out to detect several analytes using nanoporous materials, including the detection of nitroaromatic vapours,[128] and the immobilization and quantification of lipids using nanoporous silicon substrates.[211]

Another innovative method to detect different analytes is the use of piezo-resistive microcantilevers. This procedure, which has some similarities with the QCMs, has been demonstrated to result in an ultra-sensitive detection of trace-level vapors. The sensing mechanism is based in the selective molecular adsorption on a cantilever surface, which changes the surface stress and consequently, the resistance of the piezo-resistive layer. The development of sensory polymers and the coating of these microcantilevers have expanded the range of sensitivity and selectivity of these devices. Especially interesting is the detection

of traces of different explosives vapors such as TNT, RDX and pentaerythritol, research field in which some works have been published in 2014.[212]

In a similar way, the piezoelectric effect of quartz is also employed for sensing purposes in the previously mentioned devices denominated QTFs. In this device, two prongs connected at one end make a resonator whose resonance frequency is defined by the properties of the material from which it is made and by its geometry. The use of a piezoelectric substrate allows the mechanical excitation of the tuning fork to be replaced by an electrical excitation. This effect makes the QTFs suitable in gas sensor applications, by modifying QTFs with smart polymers.[213,214]

In this part of the review, we also include a brief description of holographic sensors, which are being explored recently. Essentially, and holographic sensor is a device that comprises a hologram embedded in a smart polymer for detection purposes. The detection takes place throughout a chemical interaction of the smart polymer with the analyte, then changing different optical properties of the holographic reflection, such as refractive index. The main advantage of these devices is that they can be read for a far distance at naked eye, because the transducer element is light that is refracted and reflected differently in presence of the target molecules, so they can be used in non-contact applications. A few applications of these novel devices are now being analyzed, focused, for example, on the real-time detection of several divalent metal ions.[215] A complete revision of different holographic molecularly imprinted polymers and its application in chemical sensing was published in 2011. The investigation also includes an example of steroid testosterone detection and quantification using smart polymeric films in hologram sensory devices.[138]

4.8 Use of smart polymers

In this last part of the section, we introduce the use of smart polymers in sensory devices. We will focus our analysis in the thermo-responsive and optically active polymers. For example, the thermo-responsive polymeric sensors based on volume phase transition are described in the review published by Zhang *et al*. [216], which could be a good starting point for the non-experience readers. Another novel and interesting publication in the same field was reported in 2018 by Ding *et al.*[217] devoted to the determination of the intracellular temperature using thermo-responsive polymers. Both papers are focused in the temperature determination from 20 to 80 °C, through the UV/Vis variation measurement.

Additionally, we include two more research works, focused on the optical tweezers and switches. First, Rodrigues *et al.*[218] reported in 2018, a very interesting and recent biomedical application of polymer fiber tweezers, which are used to single cell trapping and identification. Finally, a very comprehensive work focused in biological applications of smart polymers membranes with on/off switching control can be found in the work published in 2009 by Csetneki *et al.* [219]

After this brief resume of the state of the art, a detailed revision of the research works focused on the fabrication and analysis of detection properties of micro and nano sensory devices based on smart polymers is presented in **Tables 8-16**. In the case of a review article, it is conveniently pointed out in the table entry.

Table 8. Detection and quantification of analytes using fibers and nanofibers based on smart polymers (additional and detailed information can be found in reference 182).

Polymer type	Target species	Type of response	Detection time	Sensitivity	Reference
Polyamides	Acidity in water	Colorimetric – UV/Vis	5 min	Pks between 2.7 and -6.5	220

Polyethylene oxide	Isopropanol, nitrobenzene, toluene, H_2O_2	SAW	5 min	30 ppm	221
Polyaniline/poly(vinyl butyral)	Humidity	SAW	1 s	0.5 % RH	222
Polyaniline	Humidity	Electrical	6 s	1 % RH	33
	NH_3	Electrical	100 s	0.5 ppm	34
Polypyrrole	NH_3	Electrical	15 s	1-50 ppm	35
Poyamide 6	Formaldehyde	Colorimetric	30 min	50 ppb	36
Polydiacetylenes	$CHCl_3$, THF, ethyl acetate, hexane	Colorimetric	30 min	Not specified	37
Aromatic polyamides	$HClO_4$	Colorimetric - UV/Vis	5 min	10^{-2} – 3 M	88
Pyridylazo-2-naphthol-poly(acrylic acid)	Ni^{2+}	Fluorescence	Not specified	0.07 µM	38
Poly(methyl) methacrylate	Cu^{2+}	Fluorescence	Not specified	20 µM	39
Poly[2-methoxy-5-(2-ethylhexyloxy) -1,4-phenylenevinylene]	1,4-dinitrobenzene	Fluorescence	5 min	6.3 ppm	40
Polydimethylsiloxane	Ru^{2+}	Luminescence	Not specified	10 ppm	41

CP	Pd^{2+}	Luminescence	Not specified	1 µM	47
Polyamide	Glucose	Electrical	30 s	6 µM	223
Poly(vinylidene fluoride) and poly(aminophenylboronic acid)	Glucose, fructose, hydrazine and NO_2	Electrical	6 s	15 mM	224
1,3-diacriloylurea	Thyroxine	Electrical	30 s	1 ng/mL	145
Polybenzidine	Methionine	Electrical	60 s	1 ng/mL	158

Table 9. Detection and quantification of analytes using films and coatings based on smart polymers (additional and detailed information can be found in references 98 and 183).

Polymer type	**Target species**	**Type of response**	**Detection time**	**Sensitivity**	**Reference**
Pentiptycene/tetraphenylethylene /acetylene-based polymer	Picric acid and dinitrotoluene	Fluorescence	10 s	4 µM	120
	Isocyanates	Fluorescence	10 s	5 ppb	117
Thiophene-based polymers	TNT	Fluorescence	6 s	1 ppb	131
Acrylic polymers	Cyanide	Colorimetric	20 – 80 s	13 ppb	94
	TNT	Colorimetric	10 min	10 µM	123
	Cyanide	UV/Vis	5 min	0.01 M	28

Silicon-based polymers	TNT and DNT	Fluorescence	60 s	10 ppb	127
Polyfluorene-based polymer	Amines	Fluorescence	10 s	Not specified	174
2-hydroxyethyl acrylate-based polymers	Fe^{3+}	Colorimetric	5 min	50 ppb	81
	Hg^{2+}	Colorimetric	5 min	1 ppm	225
	Al^{3+}	Fluorescence	Not specified	25 ppb	85
	Al^{3+}	Fluorescence	20 min	1.5 ppb	86
Acrylic polymers with pyridine motifs	Fe^{3+}, Co^{2+}, Cu^{2+} and Sn^{2+}	Colorimetric	Not specified	10^{-7}-10^{-5} M	77
2-hydroxyethyl acrylate-based polymers with fluorenone and 1,2,3-triazole motifs	Water concentration in organic solvents / Humidity	UV/Vis	Not specified	0.007 % of H_2O and 20 % RH	173
2-hydroxyethyl acrylate-based polymers with fluorenone motifs	Hg^{2+}	Fluorescence	Not specified	10^{-3} M	67
1-vinyl-2-pyrrolydone/methyl methacrylate-based polymers	Hg^{2+}	Colorimetric	2 min	1 nM	68
Polyphenylenesulfide	O_2, CO_2, N_2	Phosphorescence	Not specified	1 µM	226

Polyethylene terephthalate and PDEAEMA	CO_2, N_2	Electrochemical	Not specified	Not specified	109
Acrylamide-based polymers	Glucose and sugar	Optical density	90 min	1 – 60 mM	153
Tetrakis(pentafluorophenyl)porpholacton and poly(trimethylsilylpropyne)	O_2 gas pressure	Luminescence	Not specified	Up to 21 KPa	227
Poly(thiophene) and PEDOT	SO_2 and O_2	Electrochemical	Not specified	0.8 % molar	228
Poly(diallyldimethylammonium chloride) and PSS	NH_3	Electrical	50 s	0.5 – 100 ppm	103
Poly(methyl methacrylate)	Ethanol, methanol	UV/Vis	10 min	1000 ppm	229
Polypyrrole and polyaniline	Ethanol	Electrical	50 s	25 ppm	230
Poly(hydroxyethyl methacrylate-*N*-methacryloyl-*L*-tryptophan methyl ester)	Myoglobin	Optical reflectivity	200 s	87 µM	137
o-phenylenediamine	Dopamine	Electrochemical	Not specified	6 nM	147
4-gluconamidophenyl methacrylate	(l)-methionine and malic acid	Piezoelectrical	Not specified	1 µM	158
o-phenylenediamine	Chlortetracycline	Electrochemical	Not specified	10 µM	142

PDEAMA – Poly 2-(diethyamino)ethyl methacrylate ; PSS - Poly(sodium styrene sulfonate)

Table 10. Detection and quantification of analytes using sensory arrays based on smart polymers (additional and detailed information can be found in references 186-188 and 231).

Polymer type	Target species	Type of response	Detection time	Sensitivity	Reference
Poly(methyl methacrylate)	Lipides	Electrochemical	Not specified	Not specified	211
Poly(phenylenevinylene) and fluorene-based polymers	Explosives	Fluorescence	100 s	50 µM	132
4-mercaptobenzoic acid, 6-mercaptonicotonic acid and 2-mercaptonicotonic acid	TNT, RDX and PETN	Piezoelectric	20 – 100 s	5 – 50 ppm	125
Poly(ethylene-co-vinyl acetate) and poly(dimethyl siloxane)	Toluene, isooctane, ethanol and dimethyl-methyl-phosphonate	Electrical	200 min	100 ppm	119
Polystyrene-co-malic acid, PVC and PVP	Ammonia, acetic acid, acetone and ethanol	Colorimetric	Not specified	500 ppm	232
Polypyrrole / Polyaniline / PEDOT	Olive oil aroma	Electrical	60 s	Not specified	233
Different polymers up to 17	Benzene derivatives and fungi bacteria	Electrical	300 – 600 s	0.1 atm of partial pressure	113
3-Methylthiophene, bis(trimethylsilyl)acetylene-based polymers	Na^{+},K^{+},Mg^{2+},Ca^{2+},Mn^{2+}, Zn^{2+},Cd^{2+}	Fluorescence	Not specified	0.125 µM	74

Poly(methacrylic acid)	Amaranth, tartrazine, sunset yellow, orange II, chrysoidin and rhodamine	UV/Vis	Not specified	0.02 mM	234

Table 11. Detection and quantification of analytes using quartz crystal microbalances based on smart polymers.

Polymer type	Target species	Type of response	Detection time	Sensitivity	Reference
Propylene-butyl	Toluene, xylene, diethyether, chloroform and acetone	Piezoelectrical	30 s	250 ppm	189,190, 191
Polydimethylsiloxane	Benzene, toluene, ethylbenzene, and xylenes	Piezoelectrical	2 min	4 ppm	235
Poly(3-hexyl thiophene)	Alcoholic volatile vapors associated with Salmonell	Piezoelectrical	2 min	4.35 ppm	95

	a typhimurium				
Polytetrafluoroethylene	Volatile alcohols	Piezoelectrical	Not specified	Not specified	236
Poly(4-vinylpyridine), poly(methyl methacrylate) and poly(4-tert-butylstyrene)	Hexane, acetone and ethanol	Piezoelectrical	30 s	Not specified	192
Polyethylene and poly(ethylene-co-vinylalcohol)	Synthetic air and essential oils	Piezoelectrical	10 min	375 ppb	237
N-*N*-dimethylaminoethyl methacrylate	Indole-3-acetic acid	Piezoelectrical	Not specified	10 nM	193
Cyclodextrin-modified poly(*l*-lysine)	Bisphenol A	Piezoelectrical	5 min	400 µM	238
Polyamidoamine and 2,2'-bis(acrylamido)acetic acid	Cu^{2+}, Pb^{2+}, Cr^{3+}, Cd^{2+}	Piezoelectrical	8 min	0.01 – 1000 ppm	194
N-vinylimidazole-based polymers	Cu^{2+}	Piezoelectrical	15 min	10 ppm	195
Poly(*N,N*-dimethylacrylamide)	Au^{3+}	Piezoelectrical	60 s	0.1 ppm	196

Table 12. Detection and quantification of analytes using microfluidic devices based on smart polymers (additional and detailed information can be found in references 197, 199, 201 and 239).

Polymer type	**Target species**	**Type of response**	**Detection time**	**Sensitivity**	**Reference**

Polydiacetylene	Cyclodextrin	UV/Vis	Not specified	10 mM	133
Poly(vinylpyrrolidone), poly(ethylene glycol) and teflon	Escherichia Coli	Fluorescence	Not specified	1 mM	240
Poly(*N*-isopropylacrylamide)	Biological analytes	Electrical	Not specified	Not specified	241
Polyethylene	C-reactive protein	Fluorescence	Not specified	0.3 ng/mL	242
3-aminopropyltriethoxysilane	Cu^{2+}, Hg^{2+}	Fluorescence	Not specified	0.04 µg/L	200
2-methyltetrahydrofuran	Hg^{2+}	Fluorescence	Not specified	500 µM	45

Table 13. Detection and quantification of analytes using modified electrodes based on smart polymers (additional and detailed information can be found in references 202, 205 and 208).

Polymer type	**Target species**	**Type of response**	**Detection time**	**Sensitivity**	**Reference**
Methacrylic acid monomer	Carvedilol	Electrochemical	Not specified	16 µmol/l	243
Poly(2,2-dithiodianiline) and PAni	Hg^{2+}	Electrochemical	Not specified	56 ppb	43,44
Allylurea monomer	Pd^{2+}	Electrochemical	Not specified	6 nM/l	203
Poly(4-vinypyridine) monomer	Ag^{+}	Electrochemical	Not specified	50 nM	244

o-phenylenediamine monomer	In^{3+}	Electrochemical	Not specified	4.7 nM	245
Acrylamide monomer	Cefixime	Electrochemical	Not specified	0.001 µM	246
Poly(ethylene) glycol diacrylate	Glucose and lactic acid	Electrochemical	Not specified	1 µM	204
Polyaniline	Glucose	Electrochemical	Not specified	3 µM	247
β-cyclodextrin polymer powder	Mitoxantrone	Electrochemical	Not specified	$3 \cdot 10^{-8}$ M	206
Methacrylic acid monomer and ethylenglycol maleic rosinate acrylate monomer	Phenobarbital	Electrochemical	Not specified	$8.2 \cdot 10^{-9}$ M/l	207
Methylsilsesquioxane oligomer	Procainamide	Electrochemical	Not specified	1.3 nM	248
4-aminobenzoic acid monomer	Naloxone	Electrochemical	Not specified	0.2 µM	249
Polypyrrole/ZnO hybrid polymer	Humidity	Electrical	100 s	11 % RH	106
Poly(3,4-ethylenedioxythiophene) polystyrene-sulfonate	Humidity	Electrical	Not specified	0 - 90 % RH	250
Poly(2,5-dimethoxyaniline phenanthrene sulphonic acid)	Phenol	Electroche	Not	$7 \cdot 10^{-5}$ -	251

		mical	specifi ed	$1.5 \cdot 10^{-2}$ M	
Polyvinylpyrrolidone	Methanol , ethanol, H_2O	Electrical	10 s	1360 ppm	252

Table 14. Detection and quantification of analytes using chips and biochips based on smart polymers (additional and detailed information can be found in reference 210).

Polymer type	Target species	Type of response	Detection time	Sensitivity	Reference
Poly(*N*-isopropylacrylamide)	Anti-biotin antibody	Surface Plasmon Resonance	100 s	10^{-13} mol/cm^2	209
Polydiacetylene	Temperature	Fluorescence	Not specified	Not specified	253
KMPR® photoresist polymer	Temperature	Photoresistance	Not specified	Up to 160 °C	254

Table 15. Detection and quantification of analytes using micro and nanoporous materials based on smart polymers.

Polymer type	Target species	Type of response	Detecti on time	Sensitivi ty	Refere nce
Poly(2-methoxy-5-(2- ethylhexyloxy)-*p*-phenylenevinylene)	TNT	Fluorescenc e	60 s	7 ppb	128
Poly(2-methoxy-5-(2-ethylhexyloxy)-*p*-phenylenevinylene) and polyfluorene	TNT, RDX	Fluorescenc e	10 s	1 ppb	255
Polypyrrole	Glucose	Electroche mical	Not specifie d	0.5 mM	152
Carbazole-based polymers	TNT	Fluorescenc e	20 s	5 – 100 ppb	256

2-acrylamide-6-methoxybenzothiazole	Trinitrophenol	Fluorescence	3 min	43 nM	257

Table 16. Detection and quantification of analytes using microcantilevers, quartz tuning forks, holographic sensors, thermo-responsive polymers, polymeric tweezers and polymeric switches.

Polymer type	Target species	Type of response	Detection time	Sensitivity	Reference
Polyaniline	Humidity and soil moisture	Piezoelectric	8 - 60 s	1 % RH and 0.1 % in soil moisture	212 [1)]
4-mercaptobenzoic acid, 6-mercaptonicotonic acid and 2-mercaptonicotonic acid	TNT, RDX and PETN vapors	Piezoelectric	100 s	10^{-9} – 10^{-5} Torr	125 [1)]
Polyaniline and poly(vinyl butyral)	Humidity	Piezoelectric	1 s	0.5 % RH	222 [1)]
Poly(vinyl pyrrolidone), polyaniline and poly(vinyl) alcohol	Ethanol, methanol, chloroform and acetone	Piezoelectric	13 min	Not specified	213 [2)]
Polyaniline and polystyrene	Methanol and ethanol	Piezoelectric	1 – 3 min	Not specified	214 [2)]
Hydroxyethyl methacrylate –co-iminodiacetic acid metacrylate	Ca^{2+}, Mg^{2+}, Ni^{2+}, Co^{2+} and Zn^{2+}	Refraction index	30 s	10 – 40 mM	215 [3)]
2-hydroxyethyl acrylate and tri(ethylene glycol) methyl ether acrylate	Temperature	UV/Vis variation	Not specifi	Not specified	216 [4)]

			ed		
N-isopropylacrylamide and *N*-isopropylmethacrylamide	Temperature	UV/Vis variation	Not specified	Not specified	217 [4)]
Pentaerythritol triacrylate	Yeast Cells immobilization	Optical	Not specified	≈ 1 pN of trapping force	218 [5)]
Poly (*N*-isopropyacrylamide)	Bovine serum albumin permeability	UV/Vis variation	5 min	Not specified	219 [6)]

[1)] Microcantilevers ; [2)] Quartz tuning forks ; [3)] Holographic sensors ; [4)] Thermo-responsive polymers ; [5)] Polymeric tweezers ; [6)] Polymeric switches

5. Conclusions and perspectives

As it has been described in the present review, the fabrication of micro and nano sensory devices using polymers has grown greatly in the last decade. This work lists more than 230 research papers, only covering specific topics of the sensory devices based on polymers. This number indicates the importance of this research field, in which the number of papers published in 2017 was higher than 3000 (data taken from Scopus website in September, 2018).

First, the main sensory polymers have been described, combining classical acrylic polymers, which have been used in the last two decades, with novel synthetized polymers with complex chiral motifs, specially designed for specific detection applications. Current investigations combine the use of simple and commodity polymers for sensing processes, but it is clear that the detection of new target species (such as RNA biomarkers, nucleotides or viruses and bacteria) require the use of novel polymers which are specially synthetized, then improving also the polymerization processes associated to sensory polymers.

Concerning the target species or analytes, sensory polymers have been used in many different detection processes. First, the detection of heavy metallic cations and anionic species has been investigated since the 90's, specially the detection of Hg(II), Fe(III) and Cu(II). However, in the last years, the detection of novel species, such as F^- and CN^- has become of great importance. In parallel, the detection of explosives of hazardous gases is, nowadays, one of the key research lines, due to the increasing security problems and the terrorist menace. Then, the detection target species such as TNT or amine vapors is continuously under investigation, with the addition of new analytes such as TATP or RDX, which can now be detected in the range of ppm. These classical detection lines live together with the emerging and promising application field is related to biological and biomedical applications. This research field has gained most of the attention in the last years, focusing the efforts in the use of biochips to detect and immobilize proteins associated to different diseases to improve the diagnosis methods, together with the detection of glucose and sugar-derivates for the treatment of diabetes. Then, the detection of different drugs, neurotransmitters and hormones has developed greatly, lowering the detection limits to the range of nM. Moreover, the specific requirements of biomedical applications have also improved the development of new biocompatible polymers with sensory properties. Finally, we have also mentioned some research works concerning the detection of emerging pollutants related to the environmental protection, such as herbicides and pesticides, which can be now detected in the range of μM.

Detecting new analytes has entailed the design and construction of different devices in which the sensory polymers play an essential role. Classical devices include modified electrodes, in which electrochemical processes are easily measured and quantified, or sensory arrays, employed to combine different polymers to increase the selectivity and the applicability range of the device. Nowadays, it is interesting to remark the use of new microfluidic devices and specially the development of sensory devices based on the piezoelectric effect, employing quartz crystal microbalances in which the high sensitivity (up to

picograms), can be easily transformed into a simple electrical signal. Other devices take advantage of the colorimetric and UV/Vis variation, which are also easily quantifiable, leading to enhance the sensitivity and selectivity of the sensory devices.

However, there are still some difficulties and challenges that must be overcome. We can list, for example:

1 – First, it is needed to increase the number of target species than can be detected, especially in the biomedical and biological applications, along with the selectivity of the sensory devices. Then, it will be possible to detect and diagnose different diseases, then expanding the applicability of these sensory devices. Also, the development of new biopolymers with sensory properties must be a priority in this investigation line.

2 - Secondly, it is compulsory to reduce the detection or response time, which is a critical factor in the detection of dangerous and hazardous substances, such as heavy metallic cations or explosives. Nowadays, detection time cannot be reduced down around 30 seconds in the detection of chlorine or TNT gases, which is still a relatively high value.

3 - Third, it is interesting to analyze and improve the transduction process from the chemical interactions between the receptor and the target to a measurable property (electrical, mechanical or optical), which will be reflected in an improvement of the detection characteristics. In this sense, the use of devices based on microfluidic fabrication techniques and also the devices based on QCMs have reduced the detection time in the last years in many different sensing processes.

4 - Finally, the development of the research lines must be carried out considering also the industrial applications, then improving the production methods in order to obtain and commercialize cheap and portable sensory devices that can be used for non-specialized

personal, thus reducing the distance between fundamental research and life applications, responding to the actual society demand.

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