

# E-Tongues/Noses Based on Conducting Polymers and Composite Materials

Subjects: Materials Science, Biomaterials

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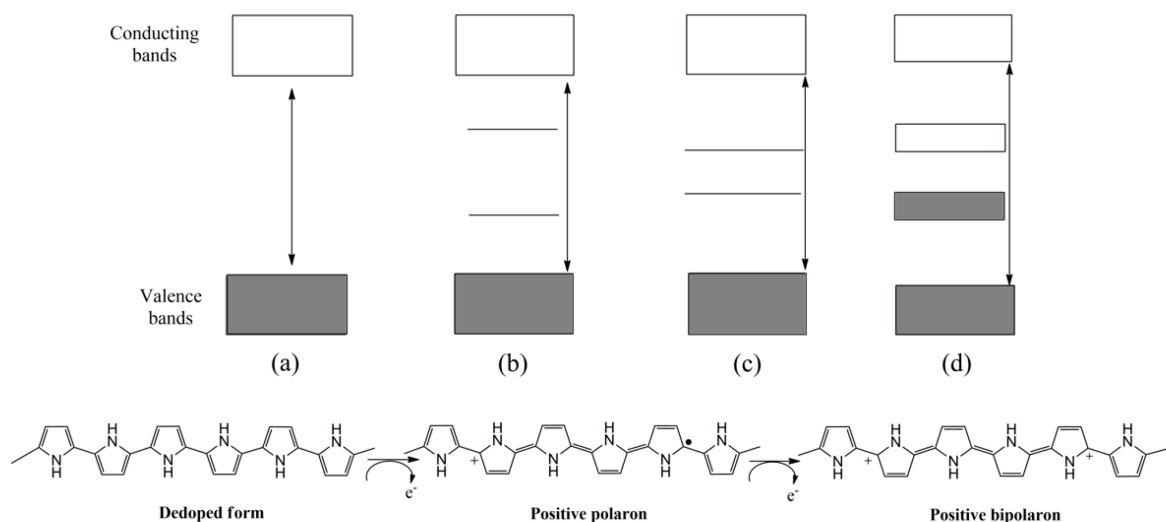
Intrinsic conducting polymers (CPs) have excellent electrochemical characteristics, such as tailored electrical conductivity by electronic doping, high environmental stability, and biocompatibility. This entry intend to overview the use of conducting polymers (CPs), extensively studied due to their high versatility and electrical properties, as chemical sensor arrays in electronic tongues and noses. Their performance in terms of sensitivity and other parameters will be studied based on the characteristic features of common conducting polymers, such as electrical conductivity and nanostructured morphology. Furthermore, the application of electronic devices in commercial prototypes will also be included here.

Keywords: conducting polymers ; electronic tongues ; electronic noses ; chemometric ; discrimination ; analysis

## 1. Introduction

In the last decades, intrinsic conducting polymers (CPs) have attracted wide attention due to their excellent electrochemical characteristics, such as tailored electrical conductivity by electronic doping, high environmental stability, and biocompatibility. Their electrical and optical properties establish them as excellent candidates for chemical sensing applications targeting the detection/determination of diverse analytes of interest. In this sense, electrochemical sensors<sup>[1][2][3]</sup>, gas sensing devices<sup>[4]</sup>, and optical sensors<sup>[5][6]</sup> have been proposed as providing good analytical features in terms of figures of merit (e.g., sensitivity, limits of detection and quantitation, repeatability, reproducibility, linear range, and robustness). In addition, the modulation of instrumental conditions during the electrodeposition process<sup>[7]</sup>, spin coating<sup>[8]</sup>, or sequential solution polymerization technique<sup>[9][10]</sup> allows precise control over the thickness and morphology of the resulting conducting coating. This advantage is particularly important in the development of electrochemical and gas sensors<sup>[11][12][13]</sup>.

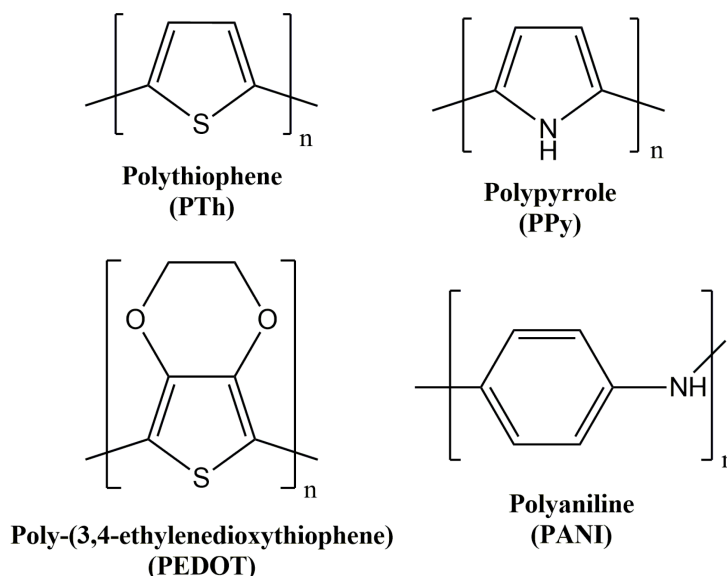
In this regard, nanostructures can also be conveniently tailored, leading to analytical sensing improvements <sup>[11]</sup>. In addition to their morphology, their electrochemical properties can be customized by electronic doping (*p* or *n*) extensively reported in the bibliography<sup>[14][15]</sup>. As an illustrative example, the *p*-doping of PPy is shown in **Figure 1**. In the first stage, a radical cation is formed by the oxidation of the polymer, inducing a local deformation within the polymer chains, leading to the formation of intermediate electronic states between the conducting and valence band. Subsequently, the polymer can be re-oxidized, increasing the number of charge carriers<sup>[16]</sup>.



**Figure 1.** Schematic representation of p-doping process of PPy and structure of electronic bands in different electronic states: (a) de-doped, (b) polaron, (c) bipolaron, (d) bipolaron coupling.

This is the reason why the electrochemical characteristics of the conducting polymers, such as electrical conductivity, can be modulated by electronic doping. This characteristic feature allows the employment of conducting polymers in several applications previously mentioned, such as electrochemical transducers in sensor devices, electrochromism, solar cells, batteries and supercapacitors, among others, proving their versatility in comparison with other electrode materials. Furthermore, the entrapment of enzymes within the polymeric layer may increase the selectivity of the overall system. In this sense, several examples, including horseradish peroxidase<sup>[17]</sup>, tyrosinase<sup>[18]</sup>, and glucose oxidase<sup>[19]</sup> can be found in the literature.

Despite the high number of intrinsic conducting polymers present in the bibliography, great attention was focused on polythiophene and the corresponding derivatives, as well as on polyaniline and polypyrrole (**Figure 2**).



**Figure 2.** Chemical structure of the most relevant intrinsic conducting polymers.

### 1.1. Polythiophene and Derivatives

Polythiophene (PTh) have several electroactive properties, such as the remarkable ability for doping/de-doping and high electrical conductivity, which makes them excellent choices for electrochromic supercapacitors and electrochemical applications<sup>[20]</sup>. Nonetheless, high potentials are required to oxidize the unsubstituted thiophene ring, which can induce the overoxidation of the resulting polymeric film. The inclusion of functional groups in the thiophene monomer leads to a significant reduction of the potentials required for the oxidation due to the presence of electron donor inductive effects<sup>[21]</sup><sup>[22]</sup>. Among all the polythiophene derivatives, poly(3,4-ethylenedioxythiophene) (PEDOT) raised based on its low oxidation potential, which provides a wide oxidation potential window. Hence, the direct electrochemical sensing of different electroactive species can be performed using PEDOT-based electrochemical devices<sup>[23]</sup><sup>[24]</sup>. Moreover, the entrapment of biological species onto PEDOT films by several procedures, such as sinusoidal current<sup>[25]</sup>, sinusoidal voltages<sup>[26]</sup>, and drop casting<sup>[27]</sup> has been achieved.

### 1.2. Polyaniline

Polyaniline (PANI) is constituted by three ideal oxidation states: leucoemeraldine (reduced form), emeraldine (half-oxidized state), and pernigraniline (fully oxidized state)<sup>[28]</sup>. The reversible redox conversion between emeraldine base, non-conducting form, to emeraldine salt, conducting form, has been exploited in several electronic devices, e.g., gas sensors, supercapacitors, electrochromic devices, and electrochemical sensors<sup>[29]</sup><sup>[30]</sup><sup>[31]</sup><sup>[32]</sup>. Regarding electrochemical devices, the electroactivity of polyaniline film plays a relevant role. Low electroactivity of the polyaniline film at neutral and basic electrolytic media can be found, which can be ascribed to the deprotonation of emeraldine salt at higher pH. Polyaniline composites were developed and used for electrochemical sensing to avoid the deprotonation of the conducting polymer backbone. In this sense, self-doped polyaniline-based devices showed electroactivity at neutral media<sup>[33]</sup><sup>[34]</sup><sup>[35]</sup>.

### 1.3. Polypyrrole

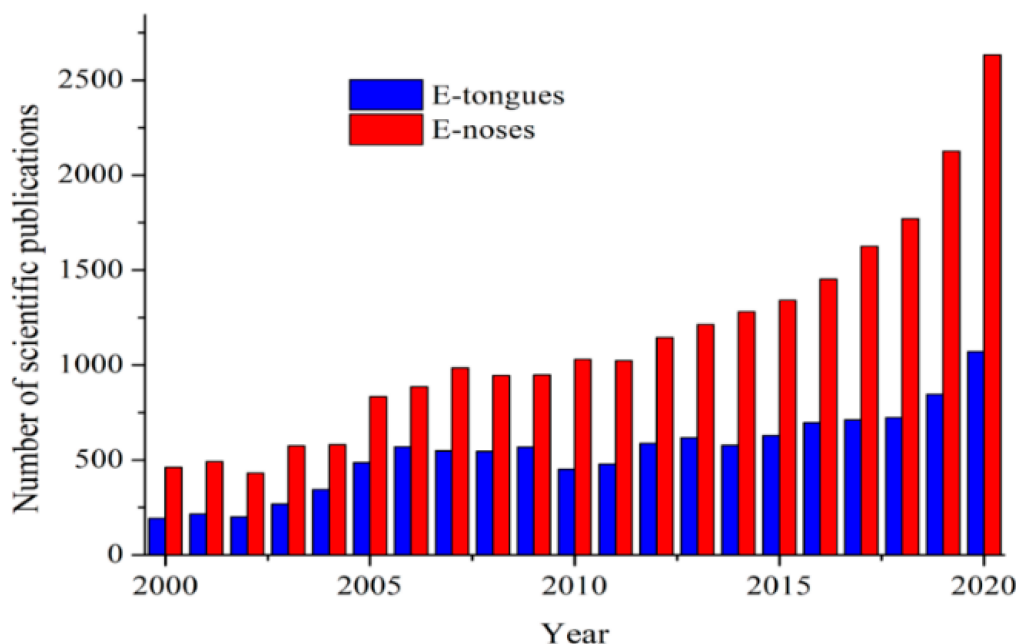
Polypyrrole (PPy) is a versatile conducting polymer, characterized by redox properties, high electrical conductivity, and reversible redox switching<sup>[36]</sup>. The redox conversion of neutral form (yellow) to oxidized form (black grey) is useful for electrochromic applications<sup>[37]</sup>. The electrochemical performance of polypyrrole composites towards several compounds

for electrochemical and gas sensors is also reported in several review papers<sup>[38][39][40]</sup>. Furthermore, the entrapment of several enzymes in conducting polypyrrole films to design biosensor devices is also reported in the bibliography<sup>[41][42][43]</sup>.

Based on the previous subsections, the feasible employment of these conducting polymers in complex and high demanding sensing devices, such as electronic tongues (E-tongues) and noses (E-noses), is evident.

#### 1.4. Electronic Systems: Electronic Tongues and Noses

Electronic systems (E-tongues/noses) emerged during the beginning of the 21st Century as useful low-time consuming tools to obtain qualitative and quantitative information about several biological, pharmaceutical, and food samples. Currently, the demand for these devices has been largely increasing in the last years. The terms electronic “tongue” or “nose” arose due to their mimicking properties of taste and smell senses, respectively. To illustrate their growing scientific interest, the number of reports related to their development published per year is shown in **Figure 3**.



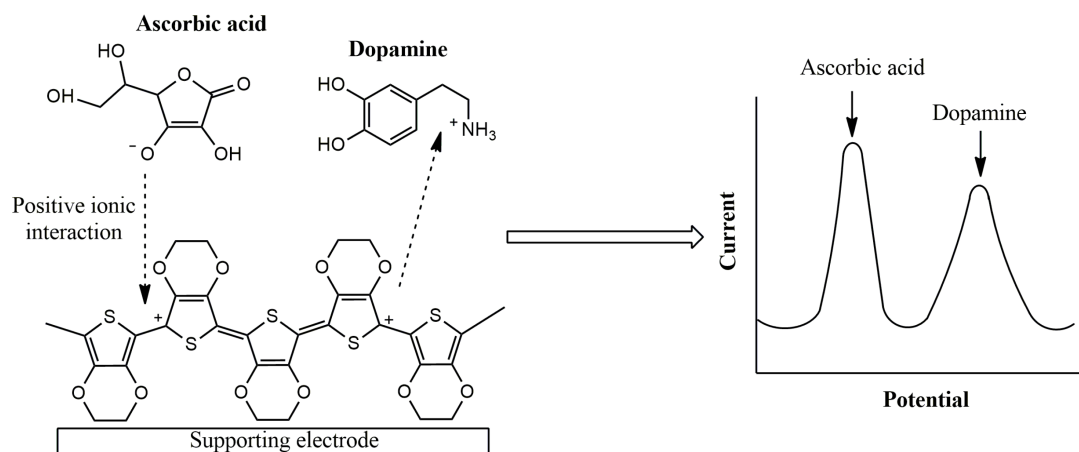
**Figure 3.** Number of scientific publications containing “electronic tongues” (E-tongues) and “electronic noses” (E-noses) terms published per year. Information obtained from Science Direct database (2021).

E-tongues/noses are mainly constituted by two blocks: a sensing unit, able to produce signals from target chemical species, and their processing part usually based on multivariate calibration methods. The latter permits the discrimination of samples, control of chemical adulteration (qualitative analysis), and correlation between experimental results and chemical parameters (quantitative analysis) by monitoring several features of the target samples<sup>[44][45][46][47][48]</sup>.

## 2. Electronic Tongues (E-Tongues) Based on CPs

### 2.1. Sensing Unit: Electrochemical Sensors

The sensing mechanism of conducting polymer-based electrochemical devices has been deeply studied in different pieces of research<sup>[49][50]</sup>. As an illustrative example, the electrochemical oxidation of ascorbic acid and dopamine using PEDOT-modified sensors in a neutral medium was evaluated. Attractive ionic forces between ascorbate, usually found at neutral medium, and the conducting layer was proposed, leading to an electrocatalytic effect for ascorbic acid oxidation. On the other hand, a repulsive interaction between dopamine and the p-doped conducting film was established<sup>[51]</sup>. Therefore, the PEDOT sensor allows the simultaneous voltammetric determination of both analytes in neutral medium (**Figure 4**).



The interaction between the polymeric backbone and the target analyte was also reported for other heterocyclic conducting polymers, such as 3-amino-5-mercapto-1,2,4-triazole<sup>[52][53]</sup>, polyaniline<sup>[54]</sup>, and poly-(N-dimethylaniline)<sup>[55]</sup>.

**Table 1.** Electrochemical sensors based on polythiophene (PTh), poly-(3,4-ethylenedioxythiophene) (PEDOT), polyaniline (PANI), and polypyrrole (PPy) currently reported in literature.

Electrochemical Device	Analyte	Working Media	Sample	Analytical Parameters		Ref.
				LD	LR	
				( $\mu\text{M}$ )	( $\mu\text{M}$ )	
PTh						
MWCNT/PTh/Pt	BPA	PBS pH 7.5	Water	0.009	0.05–0.4	[56]
MnO <sub>2</sub> /PTh/rGO/GCE	MP	PBS pH 7	Human urine and blood	0.0057	0.5–10	[57]
GO-4-ATP-Au-PTh/Au GCE	Nicotine	PBS pH 7	Serum, urine, cigarette	0.17	1.0–30	[58]
PTh-AgBr	Glucose	NaOH	Human blood plasma	0.31	4–5000	[59]
PTh-Ag/GCE	L-Tryp	PBS pH 7	Soybeans extract	0.020	0.2–400	[60]
PEDOT						

Electrochemical Device	Analyte	Working Media	Sample	Analytical Parameters		Ref.
				LD	LR	
				( $\mu\text{M}$ )	( $\mu\text{M}$ )	
PEDOT/IL/GCE	DA	PBS pH 7.4	Human urine	0.033	0.2–328	[61]
UiO-66-NH <sub>2</sub> @PEDOT/GA/GCE	PCMC	ABS pH 6	Tap water	0.2	0.6–18	[62]
PEDOT/AG/GCE	AC	PBS pH 7	Local tablets	0.041	0.15–5881	[63]
Cu <sub>2</sub> O/PEDOT/MWCNT	Glucose	NaOH	Human blood serum	0.04	0.495–374	[64]
GC/PEDOT-AuNPs-SV	CA	PBS pH 7	Juice	4.24	10–1000	[65]
PEDOT-Tyr/SNG-C	CA	PBS pH 7	Wine, beer	4.33	10–300	[66]
PEDOT/PEDOT-SH/Au	Nitrite	PBS pH 6.9	Tap water, milk	0.051	0.15–1000	[67]
PEDOT/Au	UA	PBS pH 6.6	Milk	7.0	6–200	[68]
GCE/PEDOT-MC/AgNPs	Rutin	PBS pH 3	Tablets	0.0035	0.005–0.5	[69]
Pt/PEDOT-PBNPS	H <sub>2</sub> O <sub>2</sub>	ABS pH 5.5	Human blood	1.4	5–1000	[70]
PANI						
Co <sub>3</sub> O <sub>4</sub> @PANINFs/GCE	Glucose	PBS pH 7.4	Human serum	60	100–8000	[71]
TiO <sub>2</sub> @PANI@Au/GCE	Hydrazine	NH <sub>3</sub> /NH <sub>4</sub> <sup>+</sup> pH 9	Power plant sewage	0.15	0.9–1200	[72]
PANI/SnO <sub>2</sub> /GCE	Nitrite	PBS pH 6	-	0.04	0.12–7777	[73]
GCE/PANI-Fe <sub>3</sub> O <sub>4</sub>	DA	PBS pH 7	Water	0.176	0.2–2.4	[74]
GCE/PANI-NiO	DA	PBS pH 7	Water	0.166	0.2–2.4	[74]

Electrochemical Device	Analyte	Working Media	Sample	Analytical Parameters		Ref.
				LD	LR	
				( $\mu\text{M}$ )	( $\mu\text{M}$ )	
$\alpha\text{-Fe}_2\text{O}_3/\text{PANI}/\text{GCE}$	UA	PBS pH 7	Human urine	0.038	0.01–5	[75]
NiO-NPs@PANINS/SPE	Glucose	NaOH	Human blood serum	0.06	1–3000	[76]
MeGO/PANI	AA	PBS pH 7.4	-	2.0	8–5000	[77]
PPy						
$\text{Fe}_3\text{O}_4@\text{PPy}/\text{MWCNTs}/\text{GE}$	AT	BR pH 4	Serum, tablets	0.0230	0.0314–201	[78]
AuNP/PPy/GCE	L-dopa	PBS pH 7	Urine	0.075	0.1–6.0	[79]
PDA/PPy/GCE	UA	PBS pH 8	Human serum, urine	0.11	0.5–40	[80]
PGE/CuO-NPs/PPy	TR	PBS pH 8.5	Tablets	0.001	0.005–380	[81]
PPy:LAC	Lactate	$\text{KNO}_3$	Human tear, rat blood	81.0	100–10,000	[82]
AuCu/PPy/Cu-TCCP	$\text{H}_2\text{O}_2$	PBS pH 8	Medical $\text{H}_2\text{O}_2$ solution	0.0067	0.71–24,100	[83]

AA: ascorbic acid; ABS: acetic buffer solution; AC: acetaminophen; AT: atorvastatin; ATP: adenosine triphosphate; BPA: bisphenol A; BR: Britton-Robinson; CA: caffeic acid; CuO-NPs: copper oxide nanoparticles; DA: dopamine; PTh: polythiophene; GA: graphene aerogel; GCE: glassy carbon electrode; IL: ionic liquid; LAC: lactate; LD: limit of detection; LR: linear range; L-Tryp: L-tryptophan; MC: mesoporous carbon; MP: methyl parathion; MWCNT: multi-walled carbon nanotubes; PANI: polyaniline; PANINS: polyaniline nanofibers; PBNPS: Prussian blue nanoparticles; PBS: phosphate buffer solution; PCMC: p-chloromethylcresol; PEDOT: poly-(3,4-ethylenedioxythiophene); PGE: pencil graphite electrode; PPy: polypyrrole; rGO: reduced-graphene oxide; SPE: screen-printed electrode; SV: sinusoidal voltage; TCCP: meso-tetra-(4-carboxyphenyl)-substituted porphyrins; TR: tramadol; and UA: uric acid.

## 2.2. Analytical Application of E-Tongues

Electronic tongues have been successfully applied in the analysis of a wide range of samples. Their multiple applications in different ambits employing potentiometric and/or voltammetric sensor arrays were overviewed by many researchers<sup>[84][85][86][87]</sup>.

The preliminary studies regarding electronic tongues containing conducting polymers developed by C. Mattoso and coworkers involves the use of ultrathin layers of PPy electrodeposited and their application in the distinction of some beverages<sup>[88][89][90]</sup>. One year later, De Saja developed an E-tongue by using PPy, PTh, and PANI coatings as voltammetric sensor arrays. Each one provided characteristic voltammetric signals, increasing the cross-selectivity of the

resulting device and discriminating solutions with different tasting properties<sup>[91]</sup>. Moreover, conducting polymers were tested as well for qualitative analysis of tea and coffee samples and the quantitative determination of specific analytes contained in green Korean tea<sup>[92]</sup>. Notably, subsequent sensor arrays composed by conducting electrodeposited polymer coatings are relevant for food analysis at industrial scale. **Table 2** shows several electronic tongues employed in electroanalysis of some foodstuffs.

**Table 2.** Electronic tongues containing sensors based on conducting polymer coatings applied in the analysis of food samples.

Sensor Array		Sample	Use	Multivariate Calibration	Ref.
No CP Sensor	CP Sensor				
SNG-C	PEDOT/Pt	Musts	Discrimination of samples collected at different ripening times	PCA iPLS PLS	[93]
-	PEDOT/Pt	Red wines	Classification of different samples and origin	PCA PLS	[94]
Pt Au	PEDOT/Pt	Fruit juice	Discrimination between samples from different fruits	PCA PLS-LDA	[95]
IDE PA6/IDE	PA6/PANI/IDE (0.25–5.0% PANI)	Bovine milk	Discrimination of samples according to tetracycline residue content	PCA	[96]
CE AuCE rGO-CE rGO-AuCE	PANI-CE PANI-AuCE	Vinegar, sugar	Multiflavor detection	PCA	[97]
C/SPE NiO/C/SPE MWCNT/C/SPE SWCNT/C/SPE Pt	PANI/C/SPE	Red wine	Phenolic content	PCA	[98]
SWCNT/SPCE MWCNT/SPCE	PPy-DSA/SPCE	White wine	Discrimination according to varietal origin	PCA LDA	[99]
CPE-CoPc CPE-LuPc <sub>2</sub> CPE-LuPc <sub>2</sub>	PPy-dopant/Au Dopant: SO <sub>4</sub> , DSA, FCN, AQDS, PWA, TSA	Red wine	Evaluation of chemical adulteration	PCA PLS	[100]

Sensor Array		Sample	Use	Multivariate Calibration	Ref.
No CP Sensor	CP Sensor				
GdPc <sub>2</sub> /CSPE	PPy-dopant/CSPE	Beef	Determination of ammonia and putrescine	PCA	[101]
DyPc <sub>2</sub> /CSPE	Dopant: FeCN, NP, Mo			PLS-LDA	
CSPE					
-	PPy-dopant/Pt	Beer	Evaluation of bitterness and alcoholic strength	PCA	[102]
	Dopant: DSA, H <sub>2</sub> SO <sub>4</sub> , FCN, AQDS, PWA, TSA			PLS	
-	PPy-dopant/Pt	Olive oil	Evaluation of bitterness	PCA	[103]
	Dopant: FCN, NP, PWA, H <sub>2</sub> SO <sub>4</sub> , MO, AQS			PLS	
-	PPy-dopant/SPCE	Wine	Classification of wines according to vintage year	PCA	[104]
	Dopant: DSA, SO <sub>4</sub> , FCN			LDA	
Graphite-epoxy	PANI	Wine	Classification of wines and recognition of the oxygenation effect	PCA	[105]
PtNPs	PPy				
CuNPs					

AQDS: anthraquinone-2,6-disulfonic acid, disodium salt; AQS: anthraquinone-2,6-disulfonic acid; CNT: carbon nanotubes; CoPc: cobalt phthalocyanine; CPE: carbon paste electrode; CuNPs: copper nanoparticles; DSA: sodium 1-decanesulfonate; FCN: potassium hexacyanoferrate (II); IDE: interdigitated electrodes; LDA: linear discriminant analysis; LuPc<sub>2</sub>: lutetium bis-phthalocyanine; MO: sodium molybdate; MWCNT: multi-walled carbon nanotubes; PA6: polyacrilamide; PANI: polyaniline; PCA: principal component analysis; PEDOT: poly-(3,4-ethylenedioxythiophene); PLS: partial least squares regression; PPy: polypyrrole; PWA: phosphotungstic acid; PtNPs: platinum nanoparticles; rGO: reduced-graphene oxide; SNG-C: sonogel-carbon; SPCE: screen-printed-carbon electrode; SPE: screen-printed electrode; SWCNT: single-walled carbon nanotubes; and TSA: p-toluenesulfonic acid.

### 3. Electronic Noses (E-Noses) Based on CPs

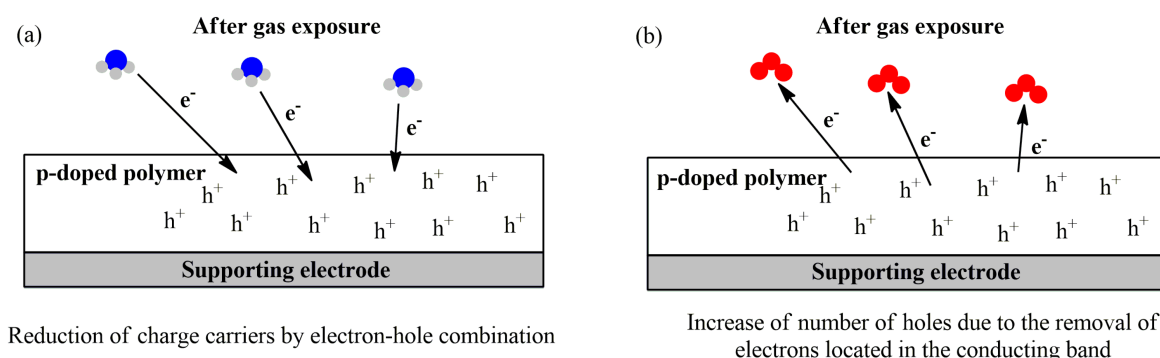
#### 3.1. Sensing Unit: Chemiresistors

In principle, three types of sensing units can be employed in gas sensing: chemiresistors, quartz crystal microbalance gravimetry, and optical sensors<sup>[106]</sup>. Among all of them, the first one is, by far, the most employed unit in electronic noses, and thus, this review will be focused on chemiresistors as the sensing unit.

The sensing performance of chemiresistor-based conducting polymers has been widely reported in bibliography. In brief, after exposing the conducting film to gases, the resistance changes depending on the initial concentration of the flowing gas. The overall resistance (S) measured with the p-doped polymeric device was calculated by the ratio between the resistance in air (R<sub>a</sub>) and the resistance in the presence of the flowing gas (R<sub>g</sub>) by using the following equation<sup>[107]</sup>.

$$S = \frac{|R_g - R_a|}{R_a} \times 100 \quad (1)$$

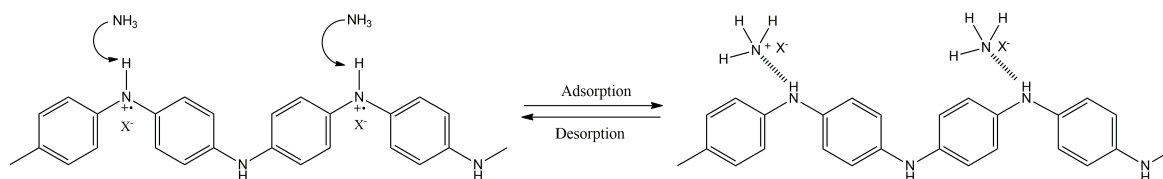
The sensing mechanism of p-doped conducting polymers towards different pollutants is overviewed in several review papers<sup>[108][109][110][111]</sup> and shown in **Figure 5**. The target gas can act as an electron donor of the conducting polymer layer, leading to a decrease in the number of holes by electron-hole combination, and thus, increase the charge resistance. On the other hand, electrons from the conducting band of the polymer can be removed by an electron-acceptor compound, leading to the increase in the number of holes, and, hence, decrease the electrical resistance.



**Figure 5.** Overall mechanism of gas sensing using a reducing gas ( $\text{NH}_3$ ) (a) and an oxidant gas ( $\text{O}_3$ ) (b) as examples.

The p-doped polymer resistance changes depending on the nature of the target analyte: oxidant gases, such as  $\text{NO}_2$  and  $\text{O}_3$ , induce an increase in the number of major charge carriers, decreasing the resistance (**Figure 5b**). Reducing gases, such as  $\text{NH}_3$ ,  $\text{CO}$ , and  $\text{H}_2\text{S}$ , induce the opposite effect by decreasing the charge carriers of the conducting film (**Figure 5a**)<sup>[112]</sup>.

The protonation/deprotonation of the conducting layer due to the vapor exposure is also reported in the literature for some conducting polymers, such as PANI<sup>[113][114][115]</sup>. **Figure 6** illustrates a possible interaction between ammonia, a reducing gas widely employed as model analyte, and PANI.



**Figure 6.** Possible interaction mechanism between ammonia and PANI.

As it can be observed in the previous figure, PANI can be deprotonated under ammonia exposure, leading to the de-doped state of PANI. This process is reversible, and thus, PANI can be protonated again, leading to the desorption of ammonia. The performance of some chemiresistors based on conducting polymers, in terms of concentration detected, response and recovery times, are shown in **Table 3**.

**Table 3.** Chemiresistors based on polythiophene (PTh), poly-(3,4-ethylenedioxythiophene) (PEDOT), polyaniline (PANI), and polypyrrole (PPy).

Gas Sensor Device	Target Gas	Range (ppm)	Sensing Performance			Ref.
			Gas Conc. (ppm)	Recovery Time (s)	Response Time (s)	
$\text{SnO}_2/\text{PTh}$	$\text{NO}_2$	10–200	10	-	2.07	[116]
P3CT/CNT	NMPEA	0.004–0.032	0.004	40	20	[117]
PEDOT:PSS/ $\text{FeCl}_3$	$\text{NH}_3$	0.2–200	0.5	-	20	[118]

Gas Sensor Device	Target Gas	Range (ppm)	Sensing Performance			Ref.
			Gas Conc. (ppm)	Recovery Time (s)	Response Time (s)	
WO <sub>3</sub> -PEDOT:PSS	LPG	500–3000	500	54	29.4	[119]
PANI/PVDF	NH <sub>3</sub>	0.2–5	0.2	235	174	[120]
PANI/SnO <sub>2</sub>	NO <sub>2</sub>	5–55	37	25	17	[121]
SnO <sub>2</sub> /rGO/PANI	H <sub>2</sub> S	0.05–10	2	78	82	[122]
PANI-NF	LPG	100–1000	700	200	50	[123]
PPy/rGO	NH <sub>3</sub>	1.0–4.0	1.0	300	60	[124]
PPy thin film	NO <sub>2</sub>	10–100	10	374	218	[125]
PPy nanoribbons	CH <sub>3</sub> CH <sub>2</sub> OH	-	100	31	2	[126]
PPy-Ag	CH <sub>3</sub> COCH <sub>3</sub>	25–600	580	150	175	[127]
PPy-CNT	H <sub>2</sub>	1–100	10	-	>1.0	[128]

CNT: carbon nanotubes; LPG: liquified petroleum gas; NF: nickel ferrite; NMPEA: n-methylphenethylamine; P3CT: poly[3 - (6-carboxyhexyl)thiophene-2,5-diyl]; PANI: polyaniline; PEDOT: poly-(3,4-ethylenedioxythiophene); PPy: polypyrrole; PSS: poly(styrenesulfonate); PTh: polythiophene; PVDF: polyvinylidene; and r-GO: reduced-graphene oxide.

### 3.2. Analytical Application of E-Noses

Among the multiple possible applications of E-noses, the early diagnosis of diseases and the evaluation of food quality in a non-invasive manner are the most relevant for society. Importantly, electronic nose devices based on conducting nanocomposites have proved their suitability in both scenarios during the last decade [129][130][131][132].

Despite its high interest currently, investigations of the role of conducting polymers in electronic noses started in the previous century, with the development of PPy, PTh, and PANI derivatives to detect alcoholic volatile compounds[133]. However, new discoveries have been carried out at the beginning of this century, leading to great improvements in the development of these devices. In this regard, Stella and coworkers developed an E-nose system based on PEDOT, PANI, and PPy coatings for the distinction of three Italian olive oils by using their aromatic substances content as a differentiating parameter[134]. Contrarily, other authors have vastly explored the role of several dopants. For example, Barisci et al. developed gold tracks supported on silicon chip coated with PPy doped with 12 different chemical compounds to detect aromatic hydrocarbons, benzene, toluene, ethylbenzene, and xylene[135]. In spite of the lack of concise explanations, the authors must be praised for the wide spectrum of polymers assayed. In fact, the evaluation of different dopants in CPs seems to be the quintessence of a great number of pieces of research. Particularly, PANI coatings with different doping agents are commonly reported in bibliography as sensor arrays in E-nose devices to monitor several parameters in foodstuffs and human body fluids[136][137][138]. **Table 4** shows some illustrative examples recently reported in the literature.

**Table 4.** Electronic noses based on polyaniline (PANI) films applied for analytical purposes in the last decade.

PANI Sensor Array	Sample	Use	Multivariate Calibration	Ref.
PANI-dopant/IDGEs Dopant: CSA, DBSA, HCl	Strawberry Grape Apple	Discrimination of samples according to aromatic substances	PCA	[139]
PANI-HCl/PGIEs PANI-HCl/IDEs	Strawberry Grape Apple	Detection of different aromas	PCA	[140]
PANI-dopant/IDGEs Dopant: HCl, TSA, CSA, MSA	Cow's estrus	Determination of estrus times of cows	PCA	[141]
PANI-dopant/IDEs Dopant: HCl, TSA, CSA, MSA	Bananas	Monitoring of bananas ripeness	PCA	[142]
PANI-dopant/PGIEs Dopant: CSA, HCl, DBSA	Gummy candies	Monitoring of aromas during candy storage	PCA	[143]
PANI-CSA/Chitosan PANI-DBSA/TiO <sub>2</sub> PANI-DBSA/CNT	Simulated human breath	Preliminary diagnoses of kidney disease	PCA LDA	[144]
PANI/AuNPs	Human breath	Early diagnoses of renal diseases	PCA LDA	[145]
PANI-dopant/MWCNT PANI-dopant/GO Dopant: CSA, DBSA, HCl	Essential oils	Determination of quality of essential oils	PCA	[146]

CSA: camphorsulfonic acid; DBSA: dodecylbenzenesulfonic acid; GO: graphene oxide; IDE: interdigitated electrode; MSA: methanesulfonic acid; MWCNT: multi-walled carbon nanotubes; PANI: polyaniline; and TSA: p-toluene sulfonic acid.

## 4. Future perspectives: integration of E-tongues and E-noses in commercial systems

It is not ambitious to think that the analytical applications of E-Tongues/Noses possess a great impact, not only in the foodstuff ambit but also in the health and environmental sector. Besides, this impact is rising sharply, reflecting the great need in society for these devices. Therefore, their implementation in commercial devices is exceedingly pursued by many sensor companies. Currently, there are some examples of its commercialization.

### 4.1 Commercial prototypes of E-tongues

Concerning E-tongues, Alpha M.O.S and Insent Inc. offers two models ( $\alpha$ Astree and TS-5000Z, respectively) that have been used in the evaluation of food quality in the last decade<sup>[147][148][149][150][151]</sup>. Other laboratory prototypes were also employed for pharmaceutical analysis, providing very satisfactory results, as those obtained with commercial systems<sup>[152]</sup>.

## 4.2 Commercial prototypes of E-noses

Regarding E-noses, a commercial system containing several conducting polymers as sensor arrays (Cyranose 320®), offered by Sensigent, was employed in the screening of several diseases (breast and lung cancer<sup>[153][154][155]</sup>, asthma<sup>[156][157]</sup> and amyotrophic lateral sclerosis<sup>[158]</sup>, among others), identification of foodstuffs (rice, wines<sup>[159]</sup> and fruits<sup>[160]</sup>) and classification of road asphalt samples<sup>[161][162]</sup>. Additionally, fecal VOCs can be inspected as well, informing about the microbial enterotype of infants<sup>[163]</sup>. Other companies also supply E-noses. For example, AromaScan A32S® (Osmetech Inc.) provides useful information about the diagnose of urban trees, being able to discriminate VOCs from healthy and decay woody samples<sup>[164]</sup> and the assessment of the quality of catfish meat<sup>[165]</sup>. In this work, off-flavour in catfish filets can be identified from good-flavour ones by means of PCA. Notably, the new device tested displayed promising features for the analysis of commercial beverages<sup>[166]</sup>.

## 4.3 Final remarks: challenges of electrochemical/gas sensing devices

Despite the excellent analytical results provided at laboratory scale in food, pharmaceutical and medical sectors, only some timid examples can be found commercially available. In our modest opinion, the inclusion of CPs and their development may pave the way to keep growing and reach the desired applicability of E-tongues and E-noses systems. Nowadays, in order to climb up into higher technological readiness levels (TRLs), the developed devices must be able to perform reliable, robust, fast, accurate and in-situ measurements using diverse samples, by using a non-complex, low cost and portable instrumentation. The stability of the conducting coatings is another issue to take into account, since the repeatability of the responses provided with the devices can be affected. The conducting film may be passivated after performing successive electrochemical assays, as well as film overoxidation can take place at high potentials. Furthermore, stability can be affected by swelling/deswelling phenomena. With the aim to minimize these factors, several parameters, including analyte concentration, film characteristics (e.g thickness and morphology) and instrumental conditions should be carefully controlled. Further research in this sense is under study to accomplish all the commercial requirements mentioned.

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