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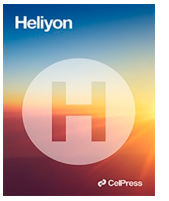

Review article

Conducting dyes as electro-active monomers and polymers for detecting analytes in biological and environmental samples

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A R T I C L E I N F O

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Redox-active dyes
Modified electrodes

A B S T R A C T

Currently, electrochemical sensors are regarded as an efficient tool for the biological and environmental sensing. Electrochemical sensors, such as voltammetric, amperometric, and impedimetric sensors, have gained great attention due to their simplicity, sensitivity, and selectivity. The performance of these electrochemical sensors could be enhanced by surface engineered nano/micro structured materials with conducting dyes/redox species. In this review, a great focus has been put on the redox-active dyes because of their electronic, optical, electrochromic, and conductivity properties. The mechanisms of oxidation and subsequent polymerization of different redox-active dyes at the surface of electrodes have been studied. Additionally, their role in catalyzing the oxidation or reduction of the target analytes at the surfaces of electrodes has also been highlighted. The redox-active dyes were used as electrochemical probes for detecting various analytes in biological and environmental samples. Overall, redox-active dyes are considered promising conducting polymers for the assessment of many analytes such as drugs, pesticides, surfactants, and heavy metal ions.

1. Introduction

Nowadays, scientific research on sensors has a great interest. The sensing principal is based on the signal transduction of unmeasurable physical or chemical responses to measurable ones [1]. It can be generally classified into optical and electrochemical sensor. Optical sensor allows the determination of the target analyte based on the remarkable change in absorption and fluorescence emission that can be quantified via ultraviolet–visible (UV–VIS) and fluorescence spectrophotometry, respectively [2]. Electrochemical sensor is a powerful analytical instrument that has gained a great attention as it is simple, inexpensive, and adaptable [3]. It has been widely used for the detection of different analytes in pharmacological, clinical, environmental, food, and industrial samples [4]. Compared with the traditional methods such as UV–VIS spectrophotometry, high-performance liquid chromatography, and mass spectrometry, which need long working times, organic solvents, tedious pretreatments, and sophisticated instrumentation, electrochemical sensor is an ideal alternative method because of its superior advantages [5]. Interestingly, it is presumed that the global electrochemical sensor market will record a compound annual growth rate (CAGR) of 11.2% during the period of 2023–2028 [6].

Electrochemical sensors are generally classified into potentiometric, amperometric, voltammetric, and impedimetric sensors. Potentiometric sensors, such as glucose sensors and ion-selective electrodes, are directly based on the Nernst equation, while amperometric sensors relies on equations like Cottrell that correlate between the measured current and the concentration of analyte during the application of unvaried potential [7]. Voltammetric sensors depend on applying potential to the electrochemical cell, and

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the produced current is associated with the concentration of the targeted analyte. The signal of voltammetric sensor can be monitored using cyclic voltammetry (CV), linear sweep voltammetry (LSV), differential pulse voltammetry (DPV), square wave voltammetry (SWV), anodic stripping voltammetry (ASV), or cathodic stripping voltammetry (CSV) technique. The difference between these techniques is attributed to the time waveforms. Impedimetric or electrochemical impedance spectroscopy (EIS) sensors are based on the immobilized receptors on the surfaces of electrodes that may result in increased or decreased resistance leading to respectively amplification or inhibition signal based on the chemical structure of the targeted analyte [8]. Additionally, the distance between the electrode and electrolyte affects the diffusion of redox-active species. On the other hand, this diffusion is influenced by the concentration of the redox-active species [9]. So that, electrode-electrolyte barrier distance and the concentration of redox-active species should be considered during EIS analysis.

Electrochemical sensors were broadly utilized in the detection of numerous drugs, natural products, and biological molecules [8, 10–12]. Additionally, they were used for the detection of inorganic pollutants, such as heavy metal ions, and organic pollutants, such as pesticides, in different samples [13]. The sensitivity and selectivity of the electrochemical sensors could be improved through using redox-active probes such as nanomaterials, ferrocene, hydroquinone, hexacyanoferrates, and conductive dyes [8]. Dyes were used as efficient conducting monomers or polymers for enhancing the electron transport at the electrode surface. Conducting polymers are broadly utilized in many fields such as medicine, energy storage, electronics, and optics owing to their high conductivity, low cost, and easy polymerization [14,15]. Conducting polymers have potential electronic, optical, electrochromic, and conductivity features making them appropriate to be used in electrochemical sensors and biosensors with enhanced analytical performance [16]. In this review, a great focus has been put on the conductive dyes as promising electrode modifier for detecting analytes in biological and environmental samples. The oxidation and subsequent polymerization mechanisms of these dyes at the electrode surfaces have been discussed. To the best of our knowledge, it is the first study that deals with all the redox-active (conductive) dyes used before in the construction of the electrochemical sensors.

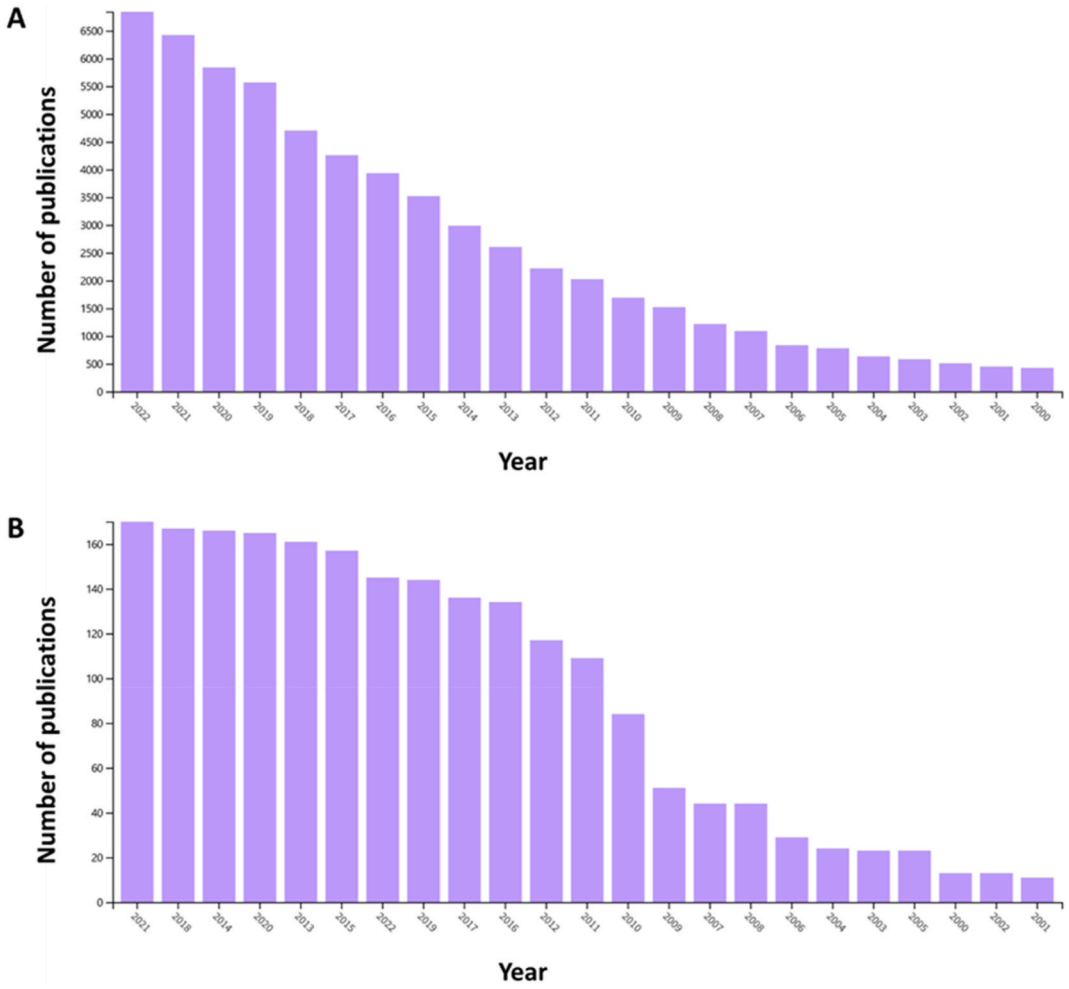


Fig. 1. Visualization of research impact in the field of (A) "electrochemical sensors" and (B) "conductive dyes" in the period 2000 to 2022 according to the results of web of science core collection.

2. Electrochemical sensors

Historically, there is a considerable concern to use electrochemical sensors for the assessment of different analytes. Fig. 1A exhibits the exponential increase of publications in the area of electrochemical sensors in the period of 2000–2022. Basically, the electrochemical sensor is composed of three main compartments: electrochemical cell, potentiostat, and signal monitoring apparatus (Fig. 2). The electrochemical cell is composed of reference, counter, and working electrodes. Reference electrode may be silver/silver chloride or saturated calomel electrode, while counter or auxiliary electrode may be formed of platinum, carbon, or polymer. The working electrode may be a platinum, a gold, a glassy carbon, or an indium-tin oxide electrode [8,17]. The working electrode may be modified with other conducting materials, which are known as redox probes, for improving the electro-catalytic activity via the enhancement of electron transfer. The modification of working electrode could be carried out using many techniques such as electro-deposition, drop casting, ink-mixing and printing, dipping, or modification by carbon paste.

Redox probes may be inorganic materials, such as metal nanoparticles (NPs), or organic materials, such as dyes, and other conducting materials. NPs exhibited the capability of constructing sensitive electrochemical sensors because of their superb electrical and mechanical features as well as their increased surface area [6,7,18–20]. NP-modified electrodes were previously used for detecting many inorganic and organic pollutants with high sensitivity and selectivity [7,21–23]. Additionally, redox-active dyes were used as electrode modifiers owing to their electrical conductivity and good chemical stability [24]. Fig. 3 represents the different types of redox-active species as electrode modifiers.

3. Redox-active dyes as electrochemical probes

Dyes are colorants which are used in many applications such as food, medicine, and textile industries [25]. Additionally, they are used as sensitizers for solar cell applications [26,27]. Reportedly, the annual production of dyes is about 700 million kg worldwide [28]. The chemical structure of dye has mainly chromophores and auxochromes which lead to the appearance of the color of dye. The available dyes could be classified as natural or synthetic dyes. The synthetic ones include direct, azo, reactive, acidic, basic, and anthraquinone dyes. Additionally, they can be categorized as cationic, anionic, and non-ionic according to the chemical structure [29]. On the other side, dyes can be classified into conductive or non-conductive dyes according to their electrochemical behavior. Recently, there has been a great concern about using conductive dyes in many applications. Fig. 1B shows the annual increase in the number of publications in the field of conductive dyes. Conductive or redox-active dyes include many dyes which contain hydroxyl or amine groups. These groups can be easily oxidized and reduced through proton and electron transfer. Table 1 lists the redox-active dyes, their classifications, and their chemical structures. The redox-active dyes are used for forming conductive layer on the surface of the electrode. Fig. 4 exhibits the adsorption of the redox dye monomer molecules at the electrode surface, their polymerization, and the subsequent removal of the excess monomer molecules. On the other side, disposable electrodes can be incorporated into electrochemical sensors for low-cost and rapid analysis of target analytes [30]. For example, disposable screen-printed electrodes have recently been incorporated into voltammetric [31], EIS [32], and amperometric [33] sensors for detecting analytes. Previously, conductive dyes were immobilized on screen-printed electrodes for the purpose of the enhancement of the electrochemical detection [34]. Accordingly, the disposable screen-printed electrodes and other ship electrodes can be efficiently used for the efficient determination of the analytes.

4. The mechanisms of the oxidation and polymerization of redox-active dyes

Redox-active dyes are easily oxidized at the electrode surfaces producing electrons as well as free radicals or carbocations. The released electrons catalyze the redox reactions of the target analyte leading to an enhancement in the analyte signal. Additionally, the free radicals or carbocations undergoes dimerization and polymerization processes. The proposed mechanisms of oxidation and polymerization of the redox-active dyes at the surfaces of electrodes are shown in Fig. 5. The alizarin is an anthraquinone derivative dye which contain quinone group. The quinone group is responsible for the reversible oxidation and subsequent polymerization of the dye. The poly(alizarin) could act as a proton receptor in the electrochemical reactions, which leads to an enhancement in the electron

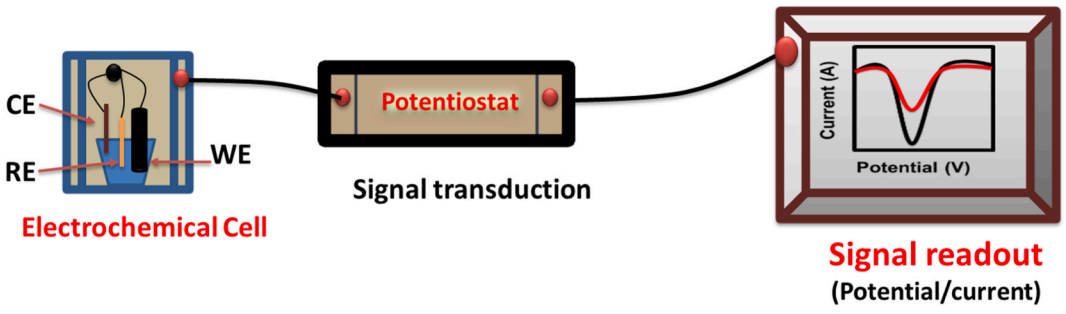


Fig. 2. Schematic representation of the compartments of an electrochemical sensor. Glassy carbon electrodes, Ag/AgCl, and platinum are examples of working electrodes (WE), reference electrodes (RE), and counter electrodes (CE), respectively.

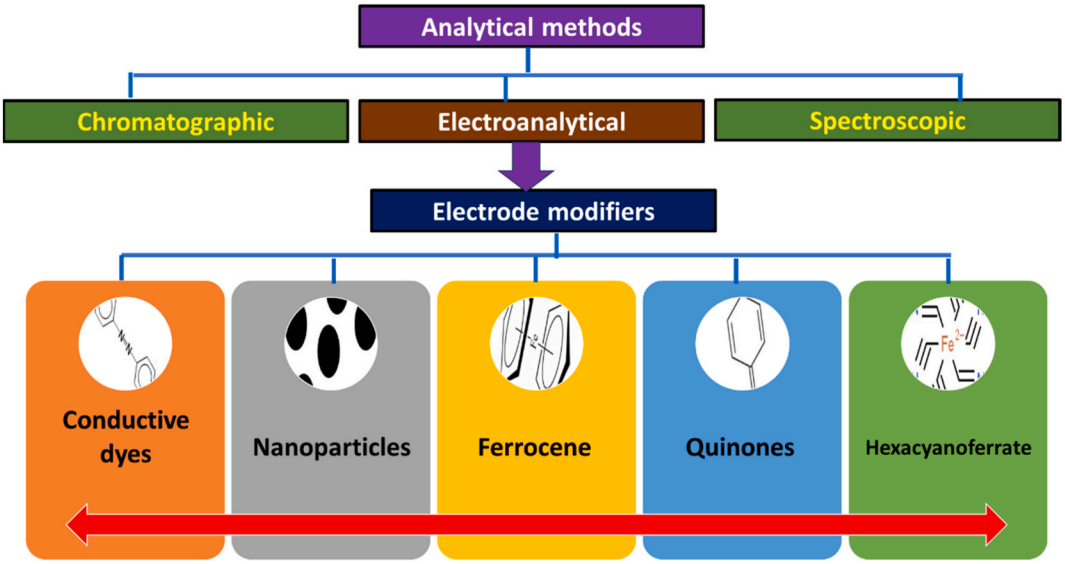


Fig. 3. Types of redox-active species as electrode modifiers.

transfer between the electrode and the electrolyte containing the analyte [35].

The modification of electrodes with azo dyes is accomplished through the oxidation of phenolic hydroxyl to the benzoquinone in alizarin yellow R, Eriochrome black t, Evans blue, hydroxy naphthol blue, and sunset yellow dyes or the oxidation of amine group in methyl orange and methyl red dyes. The oxidation of hydroxyl or amine group is accompanied by the oxidation of the azo bond to the diimine structure.

The electrode modification with oxazine dyes, such as phenazine and thiazine dyes, could be achieved through the deprotonation of amine to form imine structures. The polymerization mechanism includes the production of radicals, dimers, and tetramers that can be combined via carbon-nitrogen couplings [36,37].

Triphenyl methane dyes could also be oxidized and polymerized at electrodes surfaces. For example, the oxidation of one of the hydroxyl groups in bromocresol green or bromocresol purple dye is achieved through one-proton and one-electron transfer forming the radicals that can be combined via carbon-oxygen couplings. Similarly, phenol red dye can be oxidized forming the phenolate radical cation and its resonant form which can be combined via carbon-oxygen couplings [38]. In short, the dimerization and polymerization of redox-active dyes generally occurred via carbon-oxygen and carbon-nitrogen couplings. The oxazine, phenazine, and thiazine dyes are dimerized and polymerized via carbon-nitrogen coupling, while triphenyl methane dyes are dimerized and polymerized via carbon-oxygen coupling.

5. Redox-active dyes for detecting analytes in biological and environmental samples

Redox-active dyes were used for detecting numerous pollutants in biological (Table 2) and environmental (Table 3) samples. These pollutants include drugs, fatty acids, vitamins, neurochemicals, antioxidants, amino acids, carbohydrates, biomarkers, ATP catabolites, phenolic compounds, pesticides, dyes, and heavy metal ions (Fig. 6).

5.1. Acridine dyes

Acridines are heterocyclic organic compounds which have a nitrogen atom in their chemical structures, are non-hazardous compounds with important biological activities including antimicrobial, antibacterial, and antiprotozoal activities [87]. They have been used as DNA intercalators, staining, and DNA quantification. Previously, acridine orange was studied as a fluorescent marker for both DNA and RNA via *bis*-intercalation of the chromophore into DNA [88]. Additionally, it was utilized in numerous industrial applications due to their attractive chemical and physical features as well as their biological activities. Recently, acridine orange has been reported as a corrosion inhibitor for minimizing the corrosion of metals [89]. It has also been used as a co-reactant for detecting mercury (II) and dopamine. The dimethyl amino of the acridine orange ring is considered as an active site for the production of electrochemiluminescence and the nitrogen in the heterocyclic ring assists in the complex formation with the metal ion [90].

The electrochemical activity of acridine orange made it a good electrode modifier for determining uric acid and cetyltrimethylammonium bromide (CTAB) surfactant [14,39]. For the detection of uric acid, the electro-polymerization of acridine orange on graphene/glassy carbon electrode (GCE) was accomplished via electrolysis micelle disruption method. The N atom with sp^2 hybridization in benzene ring of the acridine orange leads to empty orbitals providing filling sites for the electrons of graphene. The acridine orange was adhered to the surface of graphene/GCE via π–π conjugation [39]. The CTAB surfactant was detected on the basis

Table 1
Classification and chemical structures of redox-active dyes.

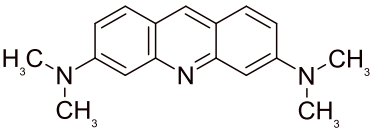
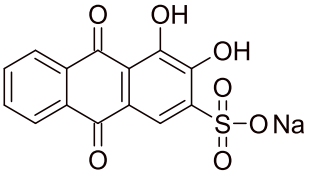
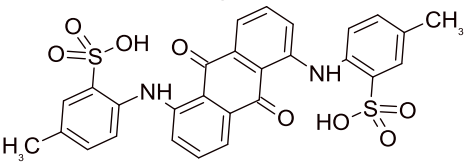
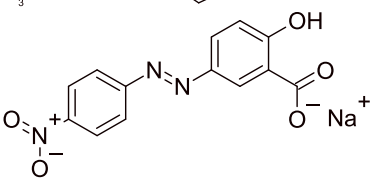
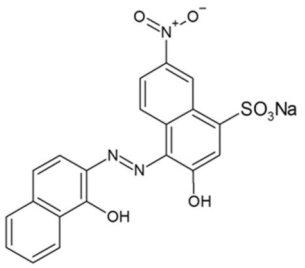
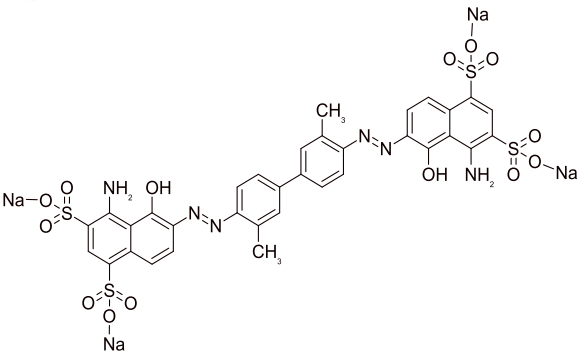
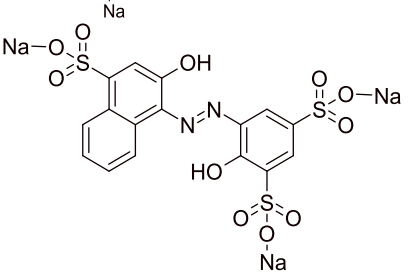
Class	dye	chemical structure
Acridines	Acridine orange	
Alizarins -Anthraquinone dyes	Alizarin red	
	Alizarin violet	
Alizarins-Azo dyes	Alizarin yellow R	
Azo dyes	Eriochrome black t	
	Evans blue	
	Hydroxy naphthol blue	

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Table 1 (*continued*)

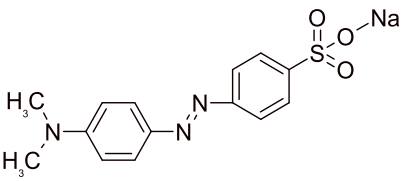
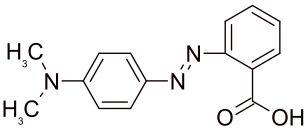
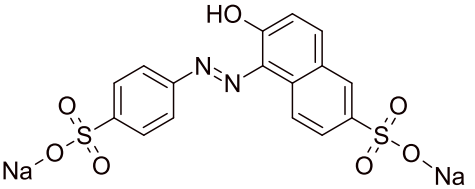
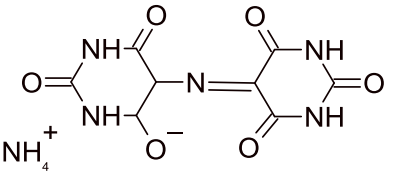
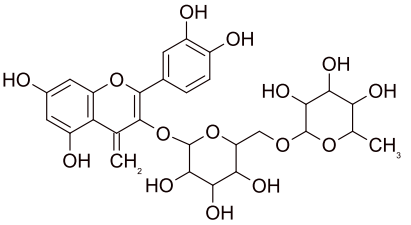
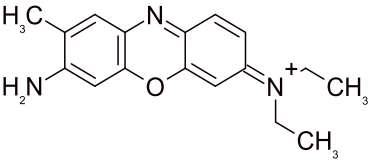
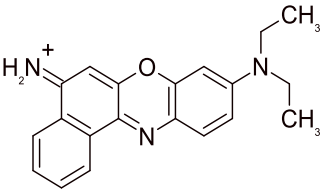
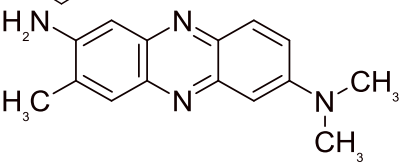
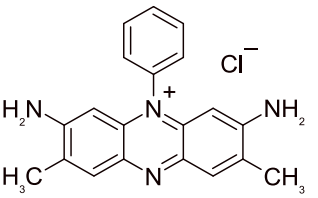
Class	dye	chemical structure
	Methyl orange	
	Methyl red	
	Sunset yellow	
Mordant dyes	Murexide	
Natural dyes	Rutin	
Oxazine dyes	Brilliant cresyl blue	
	Nile blue	
Phenazine dyes	Neutral red	
	Safranine O	

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Table 1 (*continued*)

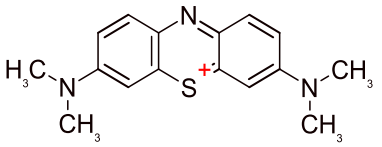
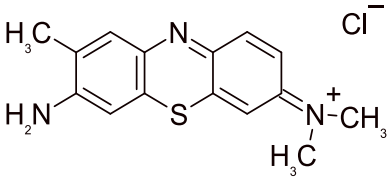
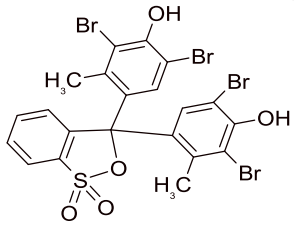
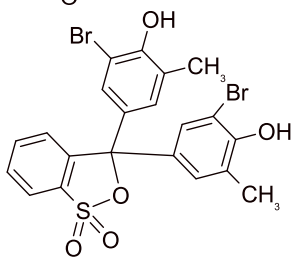
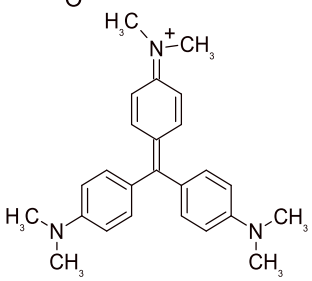
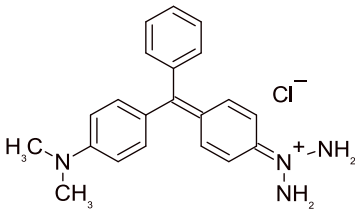
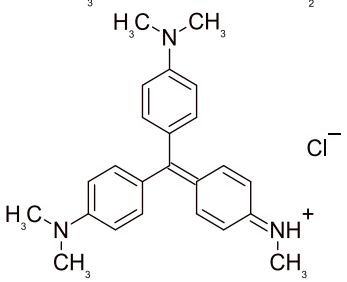
Class	dye	chemical structure
Thiazine dyes	Methylene blue	
	Toluidine blue	
Triphenyl methane dyes	Bromocresol green	
	Bromocresol purple	
	Crystal violet	
	Malachite green	
	Methyl violet	

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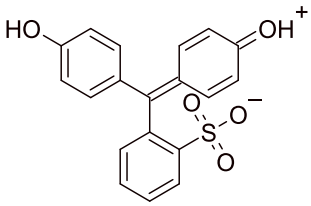
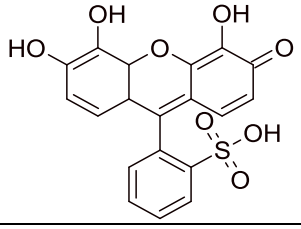
Table 1 (*continued*)

Class	dye	chemical structure
	Phenol red	
Xanthene dyes	Pyrogallol red	

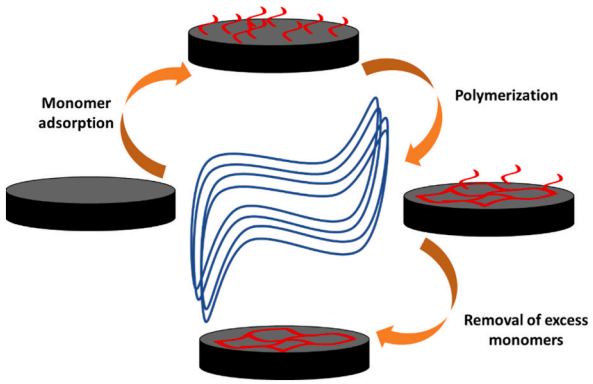


Fig. 4. Schematic representation of the formation of the conductive polymer layer at the electrode surface.

of difference in binding affinities of polystyrene sulfonate with electroactive acridine orange and electroinactive CTAB, where polystyrene sulfonate showed a stronger binding affinity toward CTAB than toward acridine orange. Based on amperometric assay through exchange mechanism, increasing CTAB concentration decreases the redox peak current of acridine orange [14].

5.2. Anthraquinone dyes

Alizarin red, which could be naturally obtained from plants, is used in many industrial applications such as dyeing and textile, as well medical applications such as staining the mineralized bones in biospecimens [91]. It can be used as electrode modifier for catalyzing redox reactions at electrodes surfaces. Previously, it was used for determining biological molecules such as nitrofuran, oxandrolone, and luteolin. Nitrofuran drug was detected using poly(alizarin red)-modified GCE. The sensing mechanism was based on the reduction of drug via two-proton and two-electron transfer [40]. Additionally, oxandrolone was detected in urine by poly(alizarin)/pyrolytic graphite electrode using DPV technique based on the inhibition of alizarin oxidation peaks [41]. Recently, luteolin drug has been determined by alizarin red S incorporated with carboxylic acid group functionalized multiwalled carbon nanotubes (f-MWCNTs) into GCE [35]. Alizarin red was used also for detecting environmental pollutants such as catechol, hydroquinone, and diuron. Catechol and hydroquinone were detected via their oxidation at poly(alizarin red S) incorporated with graphene doped with B and N on the surface of GCE, while diuron herbicide was detected through its oxidation at poly(alizarin red S) mixed with organosmectite at the surface of GCE [71,70]. On the other hand, alizarin violet, which was incorporated onto an acetylene black paste electrode, was used for determining molybdenum in plant foodstuffs. The sensing mechanism was based on the oxidation of molybdenum-alizarin violet complex via two-electron and one-proton transfer [42].

5.3. Azodyes

5.3.1. Alizarin yellow

Alizarin yellow is an acid-base indicator that can be used for preparing pH sensing materials [92]. It could be used for forming

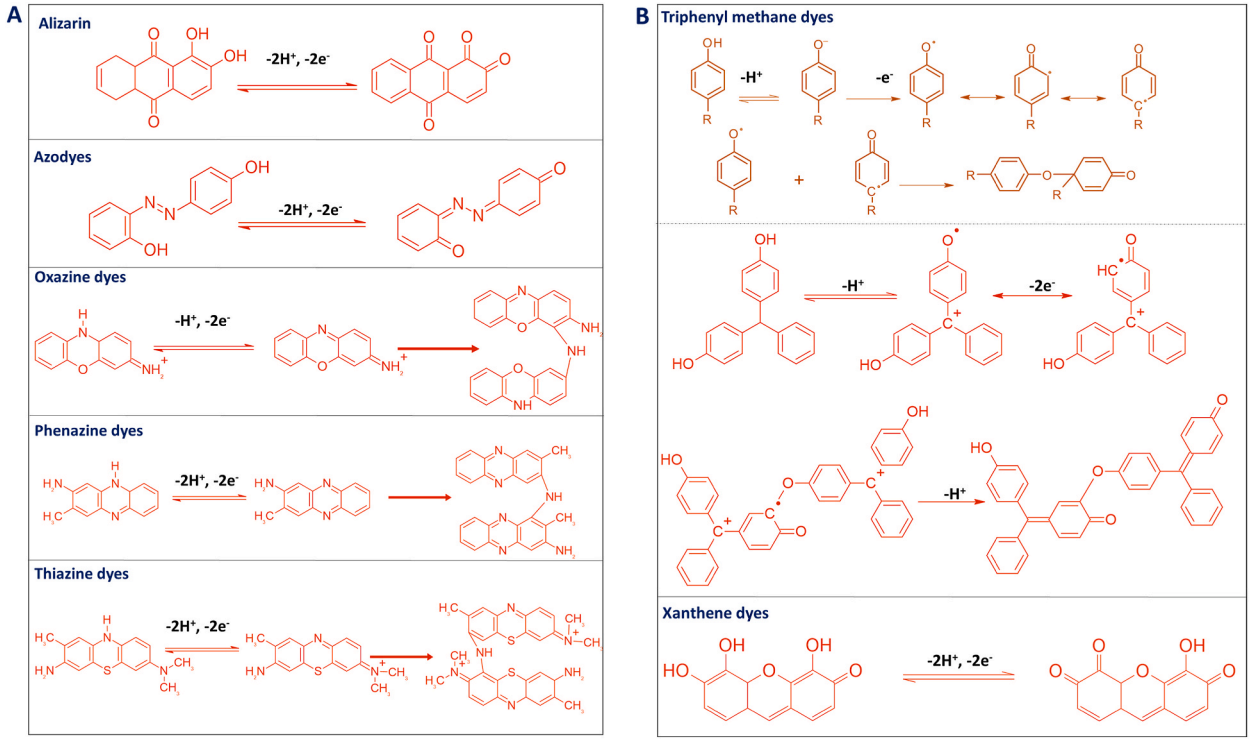


Fig. 5. A–B The proposed oxidation mechanisms of the of redox-active dyes.

composites with preferred physical, chemical, photophysical, and photochemical properties. These composites can be used in many applications such as organic photovoltaic cell [93,94]. Additionally, alizarin yellow can be act as a redox probe for detecting analytes. Previously, poly(alizarin yellow R)/carbon quantum dots immobilized on GCE was utilized for detecting L-cysteine based on its amperometric oxidation [43].

5.3.2. *Eriochrome black t*

Eriochrome black t was used for the construction of many colorimetric and fluorescent sensors as it coordinates with metal ions generating color changes [95]. It can be oxidized and polymerized at electrode surfaces acting as electrocatalysts for detecting drugs and environmental pollutants. For example, poly(Eriochrome black t) was used for sensing many drugs such as methdilazine, lansoprazole, omeprazole, and 3-aminopyridine [48–50]. Methdilazine, lansoprazole, and omeprazole were detected using poly(Eriochrome black t)-modified carbon paste electrodes using voltammetric techniques based on the direct oxidation of each drug [49,48]. 3-Aminopyridine was detected using poly(Eriochrome black t)-modified GCE based on its oxidation via two-electron and four-proton transfer [50]. On the other side, GCE modified with poly(Eriochrome black t), graphene oxide, and Au NPs was used for detecting hydroquinone, catechol, and resorcinol as environmental pollutants based on their direct oxidation [74].

5.3.3. *Evans blue*

Evans blue is a highly water-soluble dye which was utilized for disease diagnosis. It was also applied in the determination of blood volume and the permeability of blood vessels. Recently, it has been used for coloring fabrics such as cotton and silk [96]. Previously, it was used as redox-active dye for detecting hydroquinone and catechol based on their oxidation using DPV technique [75].

5.3.4. *Hydroxy naphthol blue*

Hydroxy naphthol blue is an azo dye which has many applications in the field of diagnosis and supercapacitors [97,98]. It is a redox-active dye which can be oxidized and polymerized at electrodes surfaces for detecting target analytes. Previously, poly(hydroxy naphthol blue) was used as electrode modifier for detecting hydroquinone, catechol, and resorcinol based on their oxidation using DPV technique [76].

5.3.5. *Methyl orange*

Methyl orange is broadly utilized as a pH indicator with a pH transition range of 4.4–3.2 [99]. It is a redox-active dye that can be oxidized and polymerized at the electrodes surfaces for enhancing the sensitivity of the electrochemical sensors during the determination of biological and environmental analytes. For example, the biological molecules, amodiaquine, cholesterol, and vanillin, were detected using poly(methyl orange)/MWCNT/GCE, poly(methyl orange)/NiO/MoS_2/screen printed carbon electrode, and poly

Table 2
Redox-active dyes for the electrochemical detection of analytes in biological samples.

Redox dye	electrode material	analyte	technique	mechanism	detection limit (μM)	Ref.
Acridine orange	poly(acridine orange)/graphene modified GCE	uric acid	DPV	analyte oxidation	0.02	[39]
Alizarin red	poly(alizarin red)/GCE	nitrofurazone	DPV	analyte reduction	0.33	[40]
	poly(alizarin red)/PGE	oxandrolone	DPV	modifier signal inhibition	0.0005	[41]
	poly(alizarin red)/MWCNT-COOH/GCE	luteolin	Amperometry	analyte oxidation	0.17	[35]
Alizarin violet	molybdenum-alizarin violet complex/ABPE	molybdenum	AdASV	complex oxidation	0.002	[42]
Alizarin yellow	poly(alizarin yellow R)/CQD/GCE	L-cysteine	amperometry	analyte oxidation	0.09	[43]
Brilliant cresyl blue	poly(brilliant cresyl blue)/MWCNT/GCE	glucose	amperometry		2.9	[44]
Bromocresol green	poly(bromocresol green)/pencil graphite electrode	ertapenem meropenem	SWV	analyte oxidation	0.08 0.32	[13]
	poly(bromocresol green)-PdNPs/amide functionalized SWCNT/PGE	inosine hypoxanthine xanthine uric acid	SWV	analyte oxidation	0.0009 0.001 0.0011 0.0004	[45]
Bromocresol purple	poly(bromocresol purple)-MWCNT/CPE	L-tyrosine	amperometry	analyte oxidation	0.19	[46]
Crystal violet	poly(crystal violet)/MWCNT/GCE	luteolin	DPV	analyte oxidation	0.005	[47]
Eriochrome black t	poly(Eriochrome black t)/CPE	methdilazine	SWV	analyte oxidation	0.0257	[48]
	poly(Eriochrome black t)/DES-CPE	lansoprazole omeprazole	DPASV	analyte oxidation	0.009 0.006	[49]
	poly(Eriochrome black t)/GCE	3-aminopyridine	SWV	analyte oxidation	0.54	[50]
Malachite green	poly(malachite green)/GCE	tetracycline	AdASV	analyte oxidation	1.6	[51]
Methyl orange	poly(methyl orange)/MWCNT/GCE	amodiaquine	DPV	analyte oxidation	0.089	[52]
	poly(methyl orange)/NiO/MoS_2/SPCE	cholesterol	DPV	analyte oxidation	0.00001	[34]
	poly(methyl orange)/graphene paste electrode	vanillin	DPV	analyte oxidation	0.0735	[53]
Methyl red	TiO_2–graphene/poly(methyl red)/GCE	acetaminophen	DPV	analyte oxidation	0.025	[54]
	methyl red/graphene oxide nanocomposite/carbon paste electrode	nitrite	amperometry	analyte oxidation	0.011	[55]
Methylene blue	Au NPs/poly(methylene blue)/MWCNTs/GE	nevirapine	DPASV	analyte oxidation	0.053	[56]
	poly(methylene blue)/CPE	catechin	DPV	analyte oxidation	0.0049	[57]
	poly(methylene blue)/aptamer/MIP/COOH–ZnO NPs/GCE	Cardiac Troponin I	DPV		1.04×10^{-6}	[58]
Murexide	CuO NPs/polyaniline/murexide/GCE	cholesterol	EIS	increased charge transfer resistance	0.001	[59]
	poly(murexide)/pencil GE	tryptamine serotonin dopamine	DPV	analyte oxidation	0.329 0.395 0.227	[60]
Neutral red	poly (AA) dendrimer/Ag NPs/MWCNT/poly(neutral red)-modified electrode	paracetamol ascorbic acid	SWV DPV	analyte oxidation analyte oxidation	0.053 0.053	[61] [62]
	AChE/poly(neutral red)/Fe_2O_3 NPs/GCE	acetylcholine	amperometry		1.04	[36]
Nile blue	aptamer-reduced graphene oxide/nile blue/Au NPs/GCE	dopamine	SWV	analyte oxidation	0.001	[63]
	poly(amido amine)/poly(Nile blue)-modified electrode	paracetamol dopamine ascorbic acid riboflavin	CV	analyte oxidation analyte reduction	2.2 1.63 1.63 2.63	[64]
Phenol red	reduced graphene oxide/poly(phenol red)/GCE	ciprofloxacin	DPV	analyte oxidation	0.002	[65]
Pyrogallol red	poly(pyrogallol red)/CPE	paracetamol	DPV	analyte oxidation	0.11	[66]
Safranine O	poly(safranine O)/GCE	quercetin	SWV	analyte oxidation	0.005	[67]
Sunset yellow	poly(sunset yellow)	dopamine	DPV	analyte oxidation	0.9	[20]
Toluidine blue	MWCNT-COOH/poly(toluidine blue)/GCE	dopamine	DPV	analyte oxidation	0.0104	[37]

(*continued on next page*)

Table 2 (*continued*)

Redox dye	electrode material	analyte	technique	mechanism	detection limit (μM)	Ref.
	poly(toluidine blue) MIP	breast cancer biomarker Carbohydrate Antigen 15–3 (CA 15–3)	DPV	modifier signal inhibition	0.10 U/mL	[68]
	HRP-PSA/poly(toluidine blue)/GCE	hydrogen peroxide	DPV & amperometry	analyte reduction	1	[69]
	toluidine blue-metal organic framework/GCE	indole-3-acetic acid	SWV	modifier signal inhibition	1.4 pg/mL	[24]

AA: amido amine, ABPE: acetylene black paste electrode, AChE: acetylcholinesterase, AdASV: adsorptive anodic stripping voltammetry, CPE: carbon paste electrode, CV: cyclic voltammetry, CQD: carbon quantum dots, DES: deep eutectic solvent, DPV: differential pulse voltammetry, DPASV: differential pulse anodic stripping voltammetry, EIS: electrochemical impedance spectroscopy, GCE: glassy carbon electrode, GE: graphite electrode, HRP-PSA: horseradish peroxidase-conjugated prostate-specific antigen, MIP: molecular imprinted polymer, MOF: metal organic framework, MWCNT: multiwall carbon nanotube, MWCNT-COOH: carboxylic acid group functionalized multiwall carbon nanotube, NP: nanoparticle, PGE: pyrolytic graphite electrode, SPCE: screen-printed carbon electrode, SWCNT: single-walled carbon nanotubes, SWV: square wave voltammetry.

Table 3
Redox dyes as electrocatalysts for the detection of analytes in environmental samples.

Redox dye	electrode material	pollutant	technique	mechanism	detection limit (μM)	Ref.
Acridine orange	acridine orange/PSS/PEI/GCE	CTAB	DPV	modifier signal inhibition	0.3 μg/mL	[14]
Alizarin red	poly(alizarin red s)/chitosan/BCN-graphene/GCE	hydroquinone catechol	CV	analyte oxidation	0.19 0.11	[70]
	poly(alizarin red s)/DODAB/organosmectite/GCE	diuron	DPV	analyte oxidation	0.48	[71]
Brilliant cresyl blue	poly(brilliant cresyl blue)/GCE	hydroquinone catechol	DPV	analyte oxidation	0.06 0.056	[72]
	poly(brilliant cresyl blue)/MWCNT/GCE	catechol	amperometry		3.9	[44]
Crystal violet	poly(crystal violet)/Pencil graphite electrode	hydroquinone catechol	DPV	analyte oxidation	0.0304 0.0278	[73]
Eriochrome black t	reduced GO/poly(Eriochrome black t)/Au NPs/GCE	hydroquinone catechol resorcinol	DPV	analyte oxidation	0.015 0.008 0.039	[74]
Evans blue	poly(Evans blue)/Anodized GCE	hydroquinone catechol	DPV	analyte oxidation	0.13 0.12	[75]
Hydroxy naphthol blue	poly(hydroxynaphthol blue)/MWCNT/GCE	hydroquinone catechol resorcinol	DPV	analyte oxidation	0.24 0.24 0.26	[76]
Malachite green	Poly(malachite green)/graphene nanosheets/GCE	methyl parathion	amperometry		0.002	[77]
Methyl orange	poly(methyl orange)/GCE	4-nitrophenol	DPV	analyte oxidation	0.17	[78]
Methyl red	poly(methyl orange)/MWCNT/GCE	carbendazim	LSV	analyte oxidation	0.009	[79]
Methylene blue	poly(methylene blue)/GCE	sulphide	amperometry amperometry	analyte oxidation	0.27 0.15	[80]
Murexide	murexide/poly(4-nitroaniline)/GCE	Hg^{2+}, Cu^{2+}, Ni^{2+}, and Co^{2+}	EIS	increased charge transfer resistance		[81]
	poly(murexide)/GCE	hydroquinone catechol	DPV	analyte oxidation	0.23 0.24	[82]
Neutral red	MWCNTs-neutral red-Au NPs/SPCE	Cr^{6+} V^{5+}	LSV	analyte reduction	0.025 0.42	[83]
Nile blue	poly(Nile blue)/prussian blue NPs/GCE	catechol	amperometry		26.4	[84]
Phenol red	Poly(phenol red)/GCE	hydroquinone	AdASV	analyte oxidation	0.025	[38]
Pyrogallol red	Poly(pyrogallol red)/CPE	hydroquinone catechol	DPV	analyte oxidation	0.018 0.021	[85]
Rutin	poly(rutin)/GCE	hydroquinone catechol	DPV	analyte oxidation	0.0052 0.0088	[86]

AdASV: Adsorptive anodic stripping voltammetry, CV: cyclic voltammetry, DPV: differential pulse voltammetry, DODAB: dioctadecylmethylammonium bromide, EIS: electrochemical impedance spectroscopy, GCE: glassy carbon electrode, GO: grapheren oxide, LSV: linear sweep voltammetry, MWCNT: multiwall carbon nanotube, NP: nanoparticle, PEI: Polyethyleneimine, PSS: polystyrene sulfonate, SPCE: screen-printed carbon electrode.

(methyl orange)/graphene paste electrode, respectively [34,52,53]. Additionally, 4-nitrophenol, a phenolic pollutant, was detected using poly(methyl orange)/GCE [78]. The sensing mechanism was based on the direct oxidation of the analyte at the surface of the modified electrode.

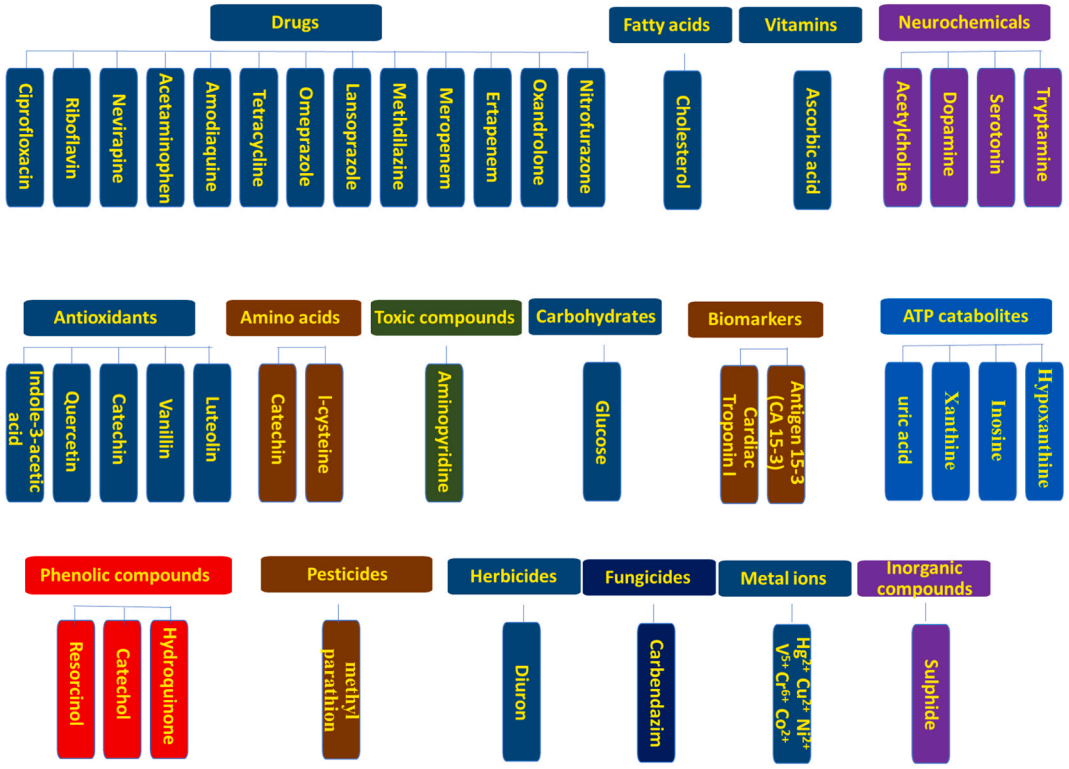


Fig. 6. The classification of different pollutants detected by conductive dyes.

5.3.6. *Methyl red*

Methyl red is commonly applied in textile dyeing process [100]. It can be used as a redox mediator for the detection of target analytes. For example, poly(methyl red) incorporated with TiO_2–graphene at the surface of GCE was used for detecting acetaminophen drug through its oxidation via two-electron and two-proton transfer [54]. Recently, methyl red and graphene oxide nanocomposite has been utilized for the construction of carbon paste electrode to detect nitrite in food samples through the oxidation mechanism using amperometric method [55]. Additionally, poly(methyl red) immobilized with multiwalled carbon nanotube (MWCNT) on the surface of the GCE was used for detecting carbendazim [79].

5.3.7. *Sunset yellow*

Sunset yellow, a water-soluble synthetic azo dye, is broadly utilized in food products [101]. It can be used as electrode modifier for preparing electrochemical sensors. Previously, poly(sunset yellow)-modified carbon paste electrode was used for determining dopamine through oxidation via two-electron and two-proton transfer using DPV technique. Additionally, the simultaneous separation of dopamine, ascorbic acid, and uric acid was accomplished by CV and DPV techniques [20].

5.4. *Murexide (mordant dye)*

Murexide, a metallochromic indicator, is used as complexometric indicator for determining Cu, Ni, Co, and rare earth metals. It is also used as a scavenger for superoxide and hydroxyl radicals. Additionally, it is utilized as a chromogenic agent for spectral detection of metals [81]. Murexide was used for the assessment of many biological molecules such as cholesterol, tryptamine, serotonin, and dopamine. It was incorporated with copper oxide (CuO) NPs and polyaniline nanofibers onto the surface of the GCE for the assessment of cholesterol. Murexide provides the biocompatibility of the sensor, good dispersion, and hydrophilic features through introduction of more hydroxyl groups which compensate positive charges on polymer chains and suppress the deprotonation processes [59]. On the other hand, poly(murexide)-modified pencil electrode was used for determining tryptamine, serotonin, and dopamine through their oxidation using DPV technique [60]. Murexide could also be used for assessing environmental pollutants such as heavy metal ions, hydroquinone, and catechol. For example, Hg^{2+}, Cu^{2+}, Ni^{2+}, and Co^{2+} were detected using murexide/poly(4-nitroaniline)/GCE via EIS method. The incorporation of murexide led to an increase in the appeared signal [81]. On the other hand, hydroquinone and catechol as model pollutants were detected using poly(murexide)/GCE based on their oxidation using DPV technique [82].

5.5. Rutin (natural dye)

Rutin, a flavonoid, is an important ingredient in food due to its antioxidant, anticancer, and anti-inflammatory activities. It can act as redox mediator for sensitive determination of target analytes. Previously, it was polymerized at GCE for detecting hydroquinone and catechol based on their oxidation using DPV technique [86].

5.6. Oxazine dyes

5.6.1. Brilliant cresyl blue

Brilliant cresyl blue, a cationic phenazine dye, is considered a redox conducting dye with better electrocatalytic features and enhanced electron transfer compared with many electrode modifiers. The electro-polymerization of brilliant cresyl blue in binary and ternary deep eutectic solvents has recently been studied [16]. Previously, poly(brilliant cresyl blue)/MWCNT/GCE was used for detecting glucose via amperometric oxidation. MWCNT were utilized as electrode modifier for the enzyme glucose oxidase to detect glucose [44]. Additionally, poly(brilliant cresyl blue)-modified GCE was used for determining hydroquinone and catechol based on their oxidation via two-proton and two-electron transfer [72]. Moreover, poly(brilliant cresyl blue)/MWCNT/GCE was used as an amperometric biosensor for detecting catechol using tyrosinase enzyme [44].

5.6.2. Nile blue

Nile blue is an important redox mediator which makes the electron transfer at electrode surface an easy process. Previously, aptasensor based on nile blue, which was mixed with reduced graphene oxide and Au NPs, was used for detecting dopamine. Nile blue act as redox probe for assessing dopamine where its oxidation peak current decreases with the increase of dopamine concentration [63]. Additionally, poly(amido amine)/poly(nile blue)-modified electrode was employed for the detection of paracetamol, dopamine, ascorbic acid, and riboflavin drugs [64]. Moreover, poly(Nile blue)/prussian blue NPs/GCE was used for the assessment of catechol using amperometric technique [84].

5.7. Phenazine dyes

5.7.1. Neutral red

Neutral red dye is utilized in many electrochemical biosensors as a redox probe for facilitating the microbe-electrode electron transfer. The polymer of neutral red was used for constructing modified carbon felt cathodes which can be used for improving the reduction of carbon dioxide to formate at the surface of the electrode [102]. On the other hand, the neutral red polymer is considered as a brilliant electron transfer mediator for detecting various biological and environmental analytes. For example, poly(amido amine) dendrimer/Ag NPs/MWCNT/poly (neutral red)-modified electrode was used for assessing paracetamol and ascorbic acid based on their oxidation via two-electron and two-proton transfer using SWV and DPV techniques, respectively [61,62]. Additionally, an acetylcholinesterase electrochemical biosensor based on poly(neutral red) and magnetic NPs for the assessment of acetylcholine [36]. On the other hand, neutral red mixed with MWCNT and Au NPs was utilized as an electrochemical sensor for the determination of Cr^{6+} and V^{5+} through their reduction using LSV technique [83].

5.7.2. Safranine O

Safranin dye, which is highly soluble in water, is applied in dyeing cotton, leather, and fibers [103]. Safranine O, a cationic phenazine dye, is broadly utilized in the preparation of pharmaceuticals. It could be polymerized at the surfaces of electrodes forming a stable redox-active polymer film. Poly(safranine O) has many active sites facilitating the electrode reaction of target analytes leading to an enhancement in the sensitivity of the electrochemical sensor. Previously, poly(safranine O)-modified GCE was utilized for detecting quercetin, food antioxidant, based on its oxidation using SWV technique [67].

5.8. Thiazine dyes

5.8.1. Methylene blue

Methylene blue, a cationic dye, is broadly utilized in textile industries [104]. It was extensively studied as conducting polymer for targeting analytes. For example, poly(methylene blue)-modified carbon paste electrode was previously investigated as an electrochemical sensor for determining catechin, antioxidant flavan-3-ol, based on its oxidation via two-electron and two-proton transfer using DPV technique [57]. Additionally, poly(methylene blue) incorporated with MWCNT and Au NPs onto graphite electrode was tested as voltammetric sensor for detecting nevirapine drug based on its oxidation via two-electron and two-proton transfer [56]. Moreover, an aptameric sensor, which is based on poly(methylene blue) and ZnO NPs, was utilized for targeting cardiac troponin I, a regulator of skeletal and cardiac muscle contractions, using DPV technique for the monitoring of the signal of poly(methylene blue). The sensing process relies on the inhibition of the oxidation peak current of poly(methylene blue) after incubation with cardiac troponin I [58]. On the other hand, sulphide as water pollutant was previously determined using poly(methylene blue) based on its oxidation using amperometric and photoamperometric techniques [80].

5.8.2. Toluidine blue

Toluidine blue dye has unique electrochemical features which led to its utilization as redox probe in many sensing applications

[68]. For example, poly(toluidine blue) and MWCNT were immobilized on GCE for detecting dopamine based on its oxidation via two-electron and two-proton transfer using DPV technique [37]. Also, poly(toluidine blue) as molecular imprinted polymer sensor was used for targeting breast cancer biomarker Carbohydrate Antigen 15–3 (CA 15–3). Ferro/ferricyanide redox probe produced a signal which was monitored by DPV technique upon binding of CA 15–3 to the imprinted surface. There was a decrease in the redox probe signal with increasing the concentration of the biomarker [68]. Additionally, the interaction of horseradish peroxidase-conjugated prostate specific antigen HRP(PSA) with the receptor, poly(toluidine blue), was used for assessing hydrogen peroxide in plasma. Poly(toluidine blue) act as supporting substrate for enhancing the surface area making the rebinding of HRP(PSA) an easy process [69]. Moreover, toluidine blue-functionalized metal-organic framework films were prepared for the electrochemical immunosensing of indole-3-acetic acid. The sensing process relied on the inhibition of the modifier signal [24].

5.9. Triphenyl methane dyes

5.9.1. Bromocresol green

Bromocresol green is commonly used as pH indicator. Also, it could be utilized as a tracking dye for DNA agarose gel electrophoresis. Additionally, it is used in the textile industries and in the determination of serum albumin for the diagnosis of liver and kidney failure. It is a non-biodegradable molecule due to its complex aromatic configuration and has a high degree of stability [105,106]. It can be electropolymerized at electrode surface to enhance the electron transfer for increasing the sensitivity of electrochemical sensors. For example, poly(bromocresol green)-modified pencil graphite electrode was used for determining ertapenem and meropenem based on their oxidation using SWV technique [13]. Additionally, poly(bromocresol green) was mixed with Pd NPs and amide functionalized single-walled carbon nanotubes to form a modified pyrolytic graphite electrode as an electrochemical sensor for detecting inosine, hypoxanthine, xanthine, and uric acid based on their oxidation using SWV technique [45].

5.9.2. Bromocresol purple

Bromocresol purple is a triphenyl methane dye with a negatively charged sulfonate. It was utilized as the sensing part of indicator films because of its high sensitivity to pH changes [107]. It was applied in many fields such as sensors, photodiode, and other devices [108]. It was used for detecting many biological and pharmaceutical molecules. Previously, poly(bromocresol purple)/MWCNT-modified carbon paste electrode was used for targeting L-tyrosine based on its oxidation using amperometric technique [46].

5.9.3. Crystal violet

Crystal violet, a triphenyl methane dye with high staining capability, is applied in many textile industries to dye cotton and silk, as well as in the production of paints and inks [109,110]. Polymerization of crystal violet at the surfaces of electrodes improve the electron transfer for enhanced detection of the targeted analytes. For example, poly(crystal violet) immobilized with MWCNT on GCE was used for detecting luteolin, flavonoid with antioxidant activity, based on its oxidation using DPV technique. There was electrostatic interaction between the electron-rich π system and MWCNT, which has carboxylate groups, and crystal violet dye of positive charge [47]. Besides biological analytes, poly(crystal violet) was used for detecting hydroquinone and catechol as model pollutants in water. Firstly, poly(crystal violet)-modified pencil graphite electrode was prepared. Next, hydroquinone and catechol were electrochemically oxidized and monitored using DPV technique [85].

5.9.4. Malachite green

Malachite green, a cationic triphenyl methane dye, is broadly used in fish, textile, leather, and printing industries [111,112]. The polymerization of malachite green can enhance the electron transfer at the surfaces of electrodes. Previously, poly(malachite green) adsorbed on the GCE surface improved the oxidation of tetracycline, an antiobiotic drug, using AdASV technique [51]. Additionally, poly(malachite green) immobilized with graphene nanosheets–nafion composite on GCE was used for detecting methyl parathion pesticide using chronoamperometric method [77].

5.9.5. Phenol red

Phenol red is commonly utilized as a pH indicator. It exhibits a rich redox feature that is essential for its application in electrode modification [38]. Poly(phenol red) has an active quinone group in the polymeric chain providing two electrons to redox reaction. Also, the presence of OH and SO_3H functional groups make it a good electron donor for the transfer of charge between the electrode and the targeted analyte. Previously, poly(phenol red) mixed with reduced graphene oxide was immobilized on the GCE for detecting ciprofloxacin drug based on its oxidation via two-electron and two-proton transfer using DPV technique [65]. Additionally, poly (phenol red)-modified GCE was utilized for determining hydroquinone as environmental pollutant based on its oxidation using AdASV technique [38].

5.10. Pyrogallol red (xanthene dye)

Pyrogallol, an important derivative of phenol, is broadly utilized in applications such as pharmaceuticals, cosmetics, and plastics. It can be polymerized at electrode surface for enhancing the electron transfer of the targeted analytes. For example, poly(pyrogallol red)-modified carbon paste electrode was tested as an electrochemical sensor for detecting paracetamol drug based on its oxidation via DPV technique [66]. Additionally, it was used for detecting hydroquinone and catechol pollutants based on their oxidation via two-electron

and two-proton transfer using DPV [85].

6. Conclusion

Redox-active dyes are promising probes for determining organic, inorganic, and biological molecules. They include many dye classes such as acridine dyes, anthraquinone dyes, azodyes, mordant dyes, natural dyes, oxazine dyes, phenazine dyes, thiazine dyes, triphenyl methane dyes, and xanthene dyes. They can enhance the electron transport at the surfaces of the electrodes making the oxidation or reduction of the targeted analytes an easy process. In this review, the previously reported redox-active or conducting dyes, their oxidation, and subsequent polymerization mechanisms have been discussed. Additionally, their effective role in catalyzing the oxidation or reduction of the targeted analytes has been indicated. However, there many redox-active dyes such as acridine orange, alizarin violet, alizarin yellow, brilliant cresyl blue, Evans blue, hydroxy naphthol blue, methyl violet, safranin O, and sunset yellow that require further studies in the field of electrochemical sensing.

Author contribution statement

All authors listed have significantly contributed to the development and the writing of this article.

Data availability statement

No data was used for the research described in the article.

Declaration of competing interest

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

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