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# Noble metal/metal–organic framework nanoparticle-based electrochemical sensors for evaluating fish quality: a comprehensive review

 Moustafa Zahran,<sup>†ab</sup> Shuang Tian,<sup>†c</sup> Ji Li,<sup>\*d</sup> Amal H. Marei,<sup>†e</sup> Ying Xie,<sup>f</sup> Qiyu Liu,<sup>f</sup> Jialin Huang,<sup>f</sup> Dezhi Wang,<sup>f</sup> Xingyao Ning,<sup>f</sup> Jiahao Wang,<sup>f</sup> Huizhong Chi<sup>\*f</sup> and Xiaolin Li<sup>\*b</sup>

Fish quality is a big-picture issue because of the possible presence of many chemical and biological pollutants, that may affect fish leading to environmental and health hazards. In this regard, researchers focus on developing efficient strategies for evaluating fish quality in terms of safety and freshness. Fish safety is determined based on assessing various pollutants, such as heavy metals, pesticides, dyes, and drugs, in fish tissue. Additionally, fish freshness evaluation is based on assessing some indicators including xanthine, hypoxanthine, uric acid, and histamine. Many chromatographic methods can assess all of these chemical indicators to evaluate the fish quality. However, these methods are expensive and often require sophisticated steps. Thus, electrochemical methods based on noble metal nanoparticles (NMNPs), metal–organic frameworks (MOFs) NPs, and their composites as electrode modifiers were investigated as potential replacements for the chromatographic ones. These materials showed high catalytic activity and electrical conductivity compared to the other electrode modifiers. In this review, we spotlight the role of NMNPs and MOF NPs in evaluating the quality of fish samples as a food source. Overall, NMNPs and MOF NPs are considered promising electrode materials for the electrochemical monitoring of fish quality.

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## Introduction

Fish, as a food source, is known for its complete protein, which contains many essential amino acids. However, fish, which absorb pollutants from the environment, may cause severe gastroenteritis and malnutrition, as well as many cancer types.<sup>1–3</sup> Fish pollutants may be microbes, such as bacteria, fungi, and viruses, or chemicals, such as heavy metals, pesticides, dyes, and drugs.<sup>4</sup> Thus, it is necessary to determine these pollutants to evaluate fish safety and quality. Fish freshness, which can be determined by the concentration of xanthine and

hypoxanthine in fish tissue, is also beneficial for quality evaluation.

The determination of pollutants and freshness indicators is accomplished using chromatographic methods, such as liquid chromatography,<sup>5</sup> high-performance liquid chromatography (HPLC),<sup>6</sup> ultra HPLC/tandem mass spectrometry (MS),<sup>7</sup> gas chromatography (GC)/MS,<sup>8</sup> and GC/tandem MS.<sup>7</sup> These chromatographic methods can efficiently assess the target analytes in fish samples. However, they require many highly sophisticated tools and utilization of organic solvents. Therefore, a great focus has been put on electrochemical methods as promising tools to evaluate fish quality.

Electrochemical methods have recently been studied as simple, fast, robust, and selective tools for assessing target analytes in fish samples.<sup>9</sup> The assessment strategy is based on the direct oxidation or reduction of the analyte at the working electrode giving characteristic redox signals. The efficiency of these electrochemical sensors can be enhanced by introducing dyes, polymers, and nanomaterials as highly conductive materials, to facilitate the electron transport at the electrode surfaces.<sup>10–12</sup> Herein, we are focused on nanomaterials as effective electrode materials, which can improve the performance of the electrochemical sensors in evaluating fish quality.

Nanomaterials have gained much attention owing to their outstanding role in various fields such as electronics and

<sup>a</sup>Guangdong Provincial Key Laboratory of Robotics and Intelligent System, Shenzhen Institutes of Advanced Technology, Chinese Academy of Sciences, Shenzhen 518055, P. R. China

<sup>b</sup>Institute of Intelligent Manufacturing Technology, Shenzhen Polytechnic University, Shenzhen, 518055, P. R. China. E-mail: lixiaolin0427@szpu.edu.cn

<sup>c</sup>Nutrition Department, Guangdong Women and Children Hospital, Guangzhou, 511442, P. R. China

<sup>d</sup>Office of Academic Research, Shenzhen Polytechnic University, Shenzhen, 518055, P. R. China. E-mail: lijij@szpu.edu.cn

<sup>e</sup>Department of Chemistry, Faculty of Science, El-Menoufia University, Shibin El-Kom 32512, Egypt

<sup>f</sup>School of Materials and Environmental Engineering, Shenzhen Polytechnic University, Shenzhen, 518055, P. R. China. E-mail: chihuizhong@szpu.edu.cn

<sup>†</sup> The two authors contributed equally to this work.



catysis.<sup>13,14</sup> Recently, nanomaterials such as multiwalled carbon nanotubes (MWCNTs),<sup>15</sup> nanofibers,<sup>16</sup> semiconducting nanoparticles (NPs),<sup>17</sup> noble metal nanoparticles (NMNPs),<sup>18</sup> metal-organic frameworks (MOFs)<sup>19</sup> have been incorporated into electrochemical sensors to determine targeted analytes. Among nanomaterials, NMNPs have desirable features in enhancing the performance of electrochemical sensors because of their high surface area and electrical conductivity. The high surface area of NMNPs allows more analyte molecules to be immobilized onto the electrode surface leading to an enhancement in the electrochemical signal.<sup>20</sup> Additionally, they can improve the electrochemical signal owing to the improved electron transport at the electrode surface. This electron transport improvement resulted from the partial oxidation of NMNP under applied potential, forming active sites that facilitate the electrochemical reactions at the electrode surface.<sup>21</sup> For example, silver (Ag) NPs acted as redox mediators for facilitating electron transport to determine different analytes such as thiols and dyes.<sup>22,23</sup> In addition to NMNPs, MOF NPs are considered brilliant electrode modifiers to improve the performance of electrochemical sensors.

MOFs, which are assembled by inorganic metal ions/clusters and organic ligands, are known for their large surface area with exposed active sites as well as various structures and compositions.<sup>24</sup> Their porosity allows adsorption and pre-concentration of target analytes leading to an improvement in the sensing process.<sup>25</sup> Thus, they were explored as promising materials for the electrochemical assessment of many analytes.<sup>26</sup> Many published review articles discuss the efficacy of NMNPs and MOF NPs as electrocatalysts in improving the electrochemical detection of many biological and environmental analytes.<sup>12,19,27,28</sup> However, there are no published review articles that discuss the role of NMNPs and MOF NPs in the electrochemical assessment of fish quality, to the best of our knowledge. Therefore, our review presents an outlook on these materials' performance, their challenges, and future directions.

## Chemical indicators of fish quality

### Fish freshness indicators

Fish freshness can be evaluated based on the determination of indicators, such as xanthine, hypoxanthine, 5'-guanosine monophosphate, uric acid, and histamine. After the die of fish, its body decomposes rapidly at room temperature, producing high purines and uric acid levels.<sup>29</sup> Firstly, ATP is degraded by guanine deaminase and xanthine oxidase to form xanthine, a derivative of guanine and hypoxanthine, which is finally degraded into uric acid.<sup>30</sup> The ATP can be converted into inosine monophosphate which is further converted into inosine and hypoxanthine.<sup>31</sup> Monitoring these indicators in fish samples can provide valuable information about the time of fish death and declare the early stages of its spoilage. Additionally, biogenic amines, such as histamine, produced in spoiled fish owing to the action of microorganisms and endogenous enzymes can be used as indicators to evaluate fish freshness.<sup>32</sup> The high histamine content may result in fish poisoning.<sup>33</sup> Food and Drug Administration (FDA) indicated that the

concentration of histamine in fish should not exceed 50 ppm.<sup>34</sup> Moreover, the 5'-guanosine monophosphate used in fish products as a flavour can be used as an indicator to evaluate fish freshness.<sup>35</sup>

### Fish safety indicators

Many possible chemical pollutants such as drugs, pesticides, dyes, heavy metals, preservatives, and toxins, may be found in fish samples.<sup>36,37</sup> Drugs and pharmaceuticals are accumulated in the fish tissue leading to an alteration of some physiological functions such as development and reproduction.<sup>38</sup> For example, antibiotics, such as ciprofloxacin, destroy cells and tissues. Additionally, they hurt the antioxidant defense, which is necessary for maintaining homeostasis in fish.<sup>39</sup> Moreover, olaquinox, a growth promoter, is known for its genotoxicity and carcinogenicity as well as its serious side effects on live-stock and humans.<sup>40</sup>

Pesticides may accumulate in fish's fatty tissues affecting plasma membrane permeability and leading to the loss of essential ions and nutrients responsible for fish's growth and survival.<sup>41</sup> They also have histopathological effects on the liver, blood vessels, and kidneys as well as an impact on the immune and endocrine systems.<sup>42</sup> Additionally, they influence the gill epithelium of fish resulting in morphological abnormalities.<sup>43</sup> Moreover, they may lead to mitochondrial depolarization and subsequent cell apoptosis.<sup>44</sup> The genotoxicity of the pesticides against fish was also reported.<sup>45</sup>

Dyes, extremely hazardous substances, have high cytotoxicity, genotoxicity, and carcinogenicity against fish.<sup>46</sup> Additionally, they can induce reproductive abnormalities in fish.<sup>47</sup> For example, malachite green, a toxic dye with carcinogenic and teratogenic effects, causes mutation in fish. Therefore, eating fish contaminated with malachite green dye may lead to serious human health problems.<sup>48</sup>

Heavy metal ions can accumulate in vital organs resulting in toxic effects, such as alterations in organosomatic indices. They also induce genotoxicity that can trigger cell apoptosis.<sup>49</sup> For example, eating fish poisoned by mercury ( $\text{Hg}^{2+}$ ) leads to the accumulation of Hg in the brain and liver, affecting the nervous system and kidney.<sup>16</sup>

Preservatives like formalin can be sprayed on fresh fish to prevent spoilage and extend the shelf life. However, consuming fish treated with excessive formalin concentration results in adverse effects such as nasopharynx and leukemia.<sup>17</sup>

Toxins, such as saxitoxin, are responsible for paralytic shellfish poisoning. Saxitoxin, a neurotoxin, blocks sodium channels in fish tissues.<sup>50</sup> Other pollutants, such as bisphenol A and 3,3',4,4'-tetrachlorobiphenyl, which act as endocrine disruptors, hurt fish safety.<sup>51,52</sup> Therefore, the detection of all these pollutants in fish samples became one of the food quality concerns. This review has extensively discussed the electrochemical detection of all these pollutants in fish samples using nanomaterials, especially NMNPs and MOF NPs, as electrode modifiers. Fig. 1 exhibits fish pollutants, drugs, heavy metal ions, preservatives, industrial chemicals, pesticides, toxins, and dyes previously detected by the electrochemical sensors. The



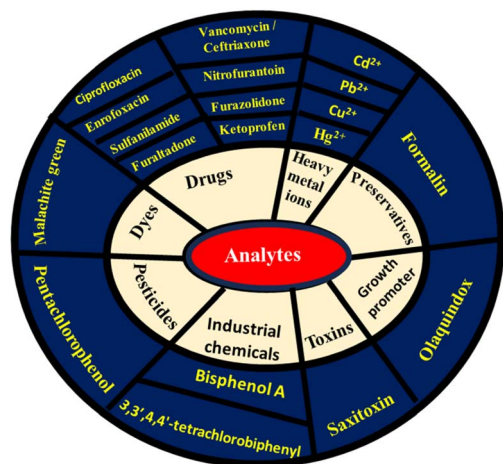


Fig. 1 Fish pollutants detected by NMNP- and MOF NP-based electrochemical sensors.

drugs included furaltadone, sulfanilamide, enrofloxacin, nitrofurantoin, furazolidone, ketoprofen, ciprofloxacin, vancomycin, and ceftriaxone, while heavy metals ions included copper ( $\text{Cu}^{2+}$ ) and  $\text{Hg}^{2+}$ . Malachite green dye, pentachlorophenol pesticide, and the endocrine disruptors, bisphenol A and 3,3',4,4'-tetrachlorobiphenyl, were also detected.

## Electrochemical sensors

Electrochemical sensors are attracting significant attention owing to their favorable features like high sensitivity, quick analysis, and good controllability.<sup>53,54</sup> The electrochemical sensor is an electronic instrument that transduces electrochemical reactions into measurable signals.<sup>55</sup> It consists of three electrodes, working, reference, and auxiliary. Ag/silver chloride (AgCl) and platinum (Pt) wire are frequently employed as reference and counter electrodes, respectively.<sup>56</sup> The working electrode is usually a glassy carbon (GCE), gold (Au), Pt, carbon paste electrode (CPE), or screen-printed (SPE) electrode.<sup>57</sup> The working electrode is a transducer where electrochemical oxidation or reduction occurs at its surface.<sup>11</sup> Usually, the working electrode is functionalized with conductive materials to improve its electrochemical performance. These materials include graphene, conducting polymers, MWCNTs, semiconducting materials, NMNPs, and MOF NPs.<sup>57</sup> Electrochemical sensing can be achieved using a variety of methods including potentiometric, voltammetric, amperometric, chemiresistive, electrochemical impedance spectroscopy (EIS), field-effect transistor (FET), electrogenerated chemiluminescence (ECL), and photochemical (PC).

Potentiometric sensors were extensively studied as valuable tools for detecting numerous target molecules, such as drugs,<sup>58</sup> biomarkers,<sup>59</sup> minerals,<sup>60</sup> heavy metals,<sup>61</sup> and organic species, with high performance.<sup>62</sup> Potentiometric sensing is based on determining the produced potential in the presence of different concentrations of the analyte of interest, as shown in Fig. 2A.

Voltammetric sensors are broadly applied to sensitively identify various organic, inorganic, and biological

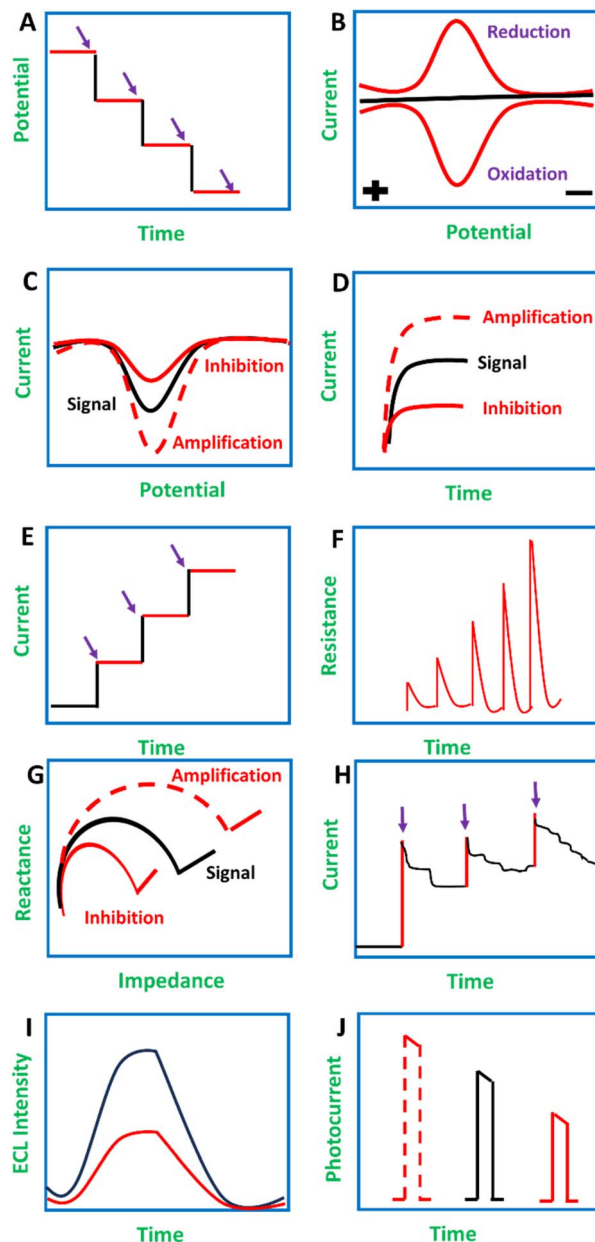


Fig. 2 The electrochemical responses of (A) potentiometric, (B and C) voltammetric, (D) chronoamperometric, (E) amperometric, (F) chemiresistive, (G) EIS, (H) FET, (I) ECL, and (J) PE sensors in the presence of an analyte.

analytes.<sup>54,63,64</sup> The determination strategy relies on a changing potential application to a working electrode and calculating the produced current corresponding to the analyte concentration.<sup>55,65</sup> Voltammetric sensors include cyclic voltammetry (CV), square wave voltammetry (SWV), differential pulse voltammetry (DPV), square wave stripping voltammetry (SWSV), and square wave adsorptive stripping voltammetry (SWAdSV) techniques. CV is considered the simplest voltammetric technique that provides general information about the oxidizable and reducible components of the sample of interest.<sup>66</sup> It is also used to monitor the sensor signal. DPV and SWV are pulse techniques



with high sensitivity and minimized non-faradic current compared with the CV technique.<sup>11</sup> Moreover, stripping techniques such as SWSV and SWAdSV have been introduced for superior sensitivity and lower detection limits. The stripping techniques require a preconcentration step to electrodeposit the analyte at the surface of the electrode.<sup>66</sup> The sensor signal usually originates from the direct analyte oxidation or reduction. Fig. 2B shows a voltammogram displaying a plot of the measured current on the y-axis against the applied potential on the x-axis. On the other side, the obtained modifier peak can be used to indirectly determine the analyte, where peak current is amplified or inhibited based on the electroactivity of the analyte, as shown in the voltammogram in Fig. 2C.

Amperometric and chronoamperometric electrochemical sensors were investigated as potential tools for assessing target substances like drugs<sup>67</sup> and inorganic molecules.<sup>68</sup> Integrating nanomaterials into electrochemical instruments leads to fast and reliable detection of target molecules. The electrochemical responses of the chronoamperometric and amperometric sensors in the presence of target analytes are shown in Fig. 2D and E, respectively. Both diagrams plot the measured current on the y-axis against time on the x-axis.

Chemiresistive sensors have gained great importance in the food and health fields.<sup>69</sup> They were utilized in the assessment of hormones,<sup>70</sup> heavy metals,<sup>71,72</sup> and gases.<sup>73</sup> The sensing principle depends on the alterations caused by the analyte in the conductivity or resistance of the film placed between the metal electrodes. The signal is produced by plotting resistance on the y-axis against time on the x-axis (Fig. 2F). The conductivity of the sensing film is affected by the charges present on the analyte of interest.<sup>74</sup>

EIS is commonly applied to evaluate a substance's electrical features and to explain the electrocatalytic reaction mechanisms by studying the charge transfer processes.<sup>75</sup> It is susceptible to the solid-liquid interface and the processes occur in the solution further from the electrode.<sup>76</sup> Many EIS-based electrochemical sensors were used in many fields, including the pharmaceutical industry, disease diagnosis, health care system, and food industry.<sup>77</sup> The assessment principle relies on the increment or decrement of a redox probe impedance in the presence of the analyte of interest, which leads to signal amplification or inhibition, respectively. The impedimetric signal is obtained by plotting reactance on the y-axis against impedance on the x-axis, as shown in Fig. 2G.

FET-based sensors have gained great attention as an elegant tool for detecting chemicals and biomolecules.<sup>78</sup> They have many attractive features, such as instantaneous response, facilitated manipulation, and noiseless. FET sensor is composed of source, drain, and gate terminals, as well as the sensing channel material. The sensing channel may be molybdenum disulfide ( $\text{MoS}_2$ ), carbides, nitrides, or graphene.<sup>79,80</sup> For example, graphene FET was previously used for assessing SARS-CoV-2 spike protein.<sup>81</sup> In general, the sensing process of FET is based on the charge-modulated mechanism where the sensing channel materials interact with the target analyte, where changes in gate dielectric constant occur.<sup>79</sup> The real-time curve, where current on the y-axis is plotted against time on the x-axis, is shown in Fig. 2H.

ECL sensors are gaining considerable attention because of their attractive features like fast response, high sensitivity, and simple instrument.<sup>82</sup> ECL converts input electrical signal into optical readout, where electroactive species are released in the vicinity of the surface of the electrode undergoing high-energy electron-transfer reactions to generate excited states emitting light.<sup>83</sup> The ECL quenching in the presence of the targeted analyte is shown in Fig. 2I.

PC sensors have recently acquired popularity owing to their simple instrumentation, low background detection signal, and optimized sensitivity and selectivity.<sup>84</sup> They have been applied to determine biological and environmental analytes.<sup>85</sup> They rely on the photosensitive features of semiconducting materials.<sup>86,87</sup> Additionally, photoactive nanomaterials have been used to enhance the semiconductor's photon-to-current conversion efficiency.<sup>88,89</sup> Signal amplification and inhibition in the presence of the analyte are shown in Fig. 2J. The sensor signal is obtained by plotting photocurrent on the y-axis against time on the x-axis.

## Electrochemical sensors for determining analytes in fish samples

Electrochemical sensors are potential tools for detecting different analytes in fish samples. Bare working electrodes were tested for the assessment process. For example, a carbon paper-based electrochemical sensor was reported as an efficient tool for the assessment of ketoprofen drug in fish samples.<sup>90</sup> The detection mechanism was based on the reduction of ketoprofen at the surface of the carbon paper electrode. Fig. 3 shows the morphology of the carbon-fiber paper, the reduction mechanism of ketoprofen, and the resulting voltammogram.

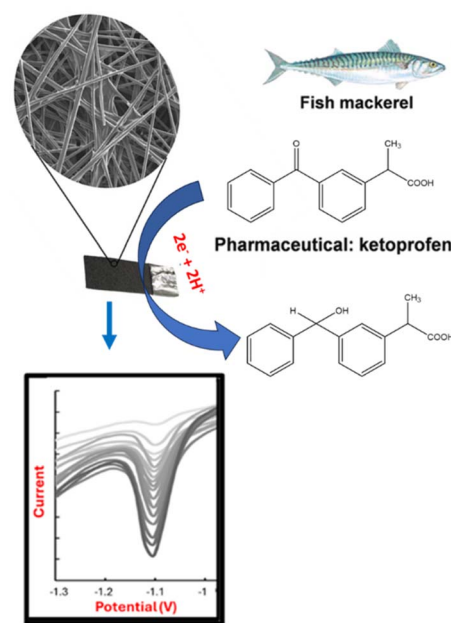


Fig. 3 Electrochemical determination of ketoprofen in mackerel using carbon fiber electrode. Reproduced from ref. 90 with permission from Elsevier, copyright 2021.



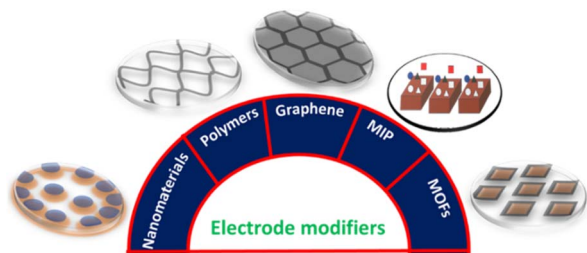


Fig. 4 An illustration of electrode modifier types used for detecting pollutants in fish samples.

In addition to bare electrodes, electrodes modified with electrocatalysts, such as graphene, molecularly imprinted polymers (MIPs), conductive polymers, nanomaterials, and MOF structures, were examined as effective sensing materials for determining analytes in fish samples. Fig. 4 shows a schematic of different electrode modifiers, graphene, MIPs, nanomaterials, and MOFs, used for fish analytes determination.

### Graphene-based electrochemical sensors

Graphene is a two-dimensional material formed from densely packed carbon atoms in a benzene-ring structure.<sup>91</sup> It has many attractive features like rapid carrier mobility and superior electrical conductivity. Therefore, it is regarded as a suitable electrode modifier to determine many targeted analytes such as drugs and heavy metal ions.<sup>92,93</sup> Recently, it has been investigated as a promising material to detect fish analytes. For example, a graphene FET sensor has been used to detect  $\text{Cu}^{2+}$  in crucian carp. The sensing mechanism was based on shifting the surface charge and changing the potential of the Au gate electrode after interaction with  $\text{Cu}^{2+}$ .<sup>94</sup> Additionally, a CPE containing reduced graphene oxide has been utilized to assess ciprofloxacin in fish samples using the DPV technique based on its direct oxidation.<sup>95</sup>

### MIP

MIP structures, synthetic polymers with binding sites, are designed to specifically bind to target molecules with great selectivity and high affinity.<sup>96</sup> They are widely utilized in fabricating many types of sensors such as colorimetric, fluorometric, and electrochemical.<sup>97</sup> They have been integrated into these sensors for optimizing their selectivity, stability, repeatability, and reproducibility.<sup>98</sup> Recently, MIP/cysteamine/Au SPE has been investigated as an electroanalytical tool for determining histamine in sardine and mackerel using the EIS technique. The positively charged histamine interacted with the negatively charged iron redox probe  $[\text{Fe}(\text{CN})_6]^{4-}/[\text{Fe}(\text{CN})_6]^{3-}$  resulting in an improvement in the charge transfer properties at the electrode surface. This improvement in charge transfer triggered an amplification in the sensor signal by increasing the concentration of histamine.<sup>99</sup> Histamine was also detected in *Rastrelliger kanagurta* (mackerel) using MIP-modified SPE using the DPV technique where histamine was oxidized.<sup>100</sup>

### Conductive polymers

Polymers, materials with large molecules formed by a repeating unit called monomers, have significant attention in sensing

applications because of their flexibility, mechanical resistance, and low cost.<sup>101</sup> Specifically, conducting polymers were extensively used as electrode modifiers due to their good electrical properties.<sup>102</sup> Recently, poly(5-amino-4H-1,2,4-triazole-3-thiol) film has been adsorbed on the anodized SPE to detect  $\text{Hg}^{2+}$  in fish samples. The sensing process was accomplished through the preconcentration of  $\text{Hg}^{2+}$  through its immobilization on the electrode surface, then electrodeposition of  $\text{Hg}^{2+}$  at the electrode surface through its reduction to  $\text{Hg}^0$ , and finally the anodic stripping of  $\text{Hg}^0$  to form  $\text{Hg}^{2+}$ .<sup>103</sup>

## Nanomaterials for electrochemical determination of analytes in fish samples

Nanomaterial-based electrochemical sensors are reliable tools for detecting analytes in fish samples. Fig. 5 exhibits a schematic illustration of different nanomaterials, electrospun nanofibers, MWCNTs, semiconducting nanomaterials, NMNPs, and MOF NPs, investigated for the electrochemical assessment of analytes in fish samples. The previously reported nanomaterial-based electrochemical sensors used for determining analytes in fish, the sensing mechanism, and the detection limits are listed in Tables 1–3. Table 1 lists the electrospun nanofibers, MWCNTs, and semiconducting nanomaterials, while Tables 2 and 3 list NMNPs and MOF NPs, respectively.

### Electrospun nanofibers

Electrospun nanofibers, nanofibers made from polymers *via* electrospinning, are brilliant sorbents to preconcentrate and isolate analytes because of their increased surface-to-volume ratio, nanoporosity, and increased adsorption capacity.<sup>128</sup> Recently, a hybrid of polyamide and chromotropic acid nanofibers electrospun on a GCE has been utilized to detect  $\text{Hg}^{2+}$  in canned fish samples using the DPV technique based on its direct reduction. The  $\text{Hg}^{2+}$  ions have specifically interacted with

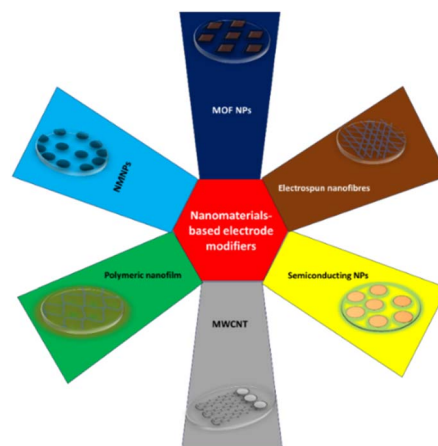


Fig. 5 Schematic representation of the nanomaterials used for the electrochemical detection of analytes in fish samples.



Table 1 Nanomaterials-based electrochemical sensors for analytes determination in fish samples<sup>a</sup>

Nanomaterial	Electrode material	Analyte	Fish	Techn.	Mechn.	LOD (nM)	Ref.
Electrospun nanofibres	PANFs/CANFs/GCE	Hg <sup>2+</sup>	Canned fish samples	DPV	Reduction	9.98	16
MWCNT	Nafion-MWCNT/GCE	Histamine	Fish sample	DPV	Oxidation	390	104
	MWCNTs/MIPs/GCE	Enrofloxacin	Fish and shrimp samples	DPV	Oxidation	0.0009	105
Semiconducting CdS NPs	CdS/chitosan/Pt electrode	Formalin	Mackerel fish	CV	Oxidation	166 666.6	17
Semiconducting CuO NPs	$\beta$ -Cyclodextrin/CuO/GCE	Chloramphenicol	Fish sample	SWV	Reduction	50	106
Semiconducting CuFe <sub>2</sub> O <sub>4</sub> NPs	CuFe <sub>2</sub> O <sub>4</sub> /Pt wire/Au paste	Formalin	Fish sample	Chem-i-resistive			107
Semiconducting ZnS NPs	MIP@CuInS <sub>2</sub> /ZnS/GCE	Sulfadiazine	Fish (local market)	DPV	Oxidation	2.1	108
Semiconducting ZnSe NPs	ZnSe/MWCNT/GCE	PCP	Carp (local market)	DPV	Oxidation	2	109
	CDTe-cDNA-aptamer/chitosan/ZnSe/Ag <sub>2</sub> Se/ITO	3,3',4,4'-Tetrachlorobiphenyl	Coral reef fish	PC	Oxidation	0.05 pg L <sup>-1</sup> and 0.22 ng L <sup>-1</sup>	52

<sup>a</sup> CANFs: chromotropic acid, CuInS<sub>2</sub>: copper indium sulfide, EIS: electrochemical impedance spectroscopy, GCE: glassy carbon electrode, GO: graphene oxide, g-SPE: gold screen-printed electrode, MIP: molecularly imprinted polymer, MWCNT: multiwalled carbon nanotube, PANFs: polyamide nanofibers, PCP: pentachlorophenol, PC: photochemical, SPCE: screen-printed carbon electrode, SWV: square wave voltammetry.

the functional groups of chromotropic acid leading to an improvement in the sensitivity and selectivity of the sensor.<sup>16</sup>

### MWCNTs

MWCNTs are cylindrical structures formed from multiple concentric graphene layers, endowing them with exceptional mechanical strength, electrical conductivity, and thermal stability.<sup>129</sup> They are known for their large specific surface area attributed to their hollow structure, which provides many exposed reaction sites.<sup>130</sup> Thus, they are considered a good electrode modifier for sensitive detection of analytes. Previously, MWCNTs were used as an electrocatalyst to determine *N*-acyl-homoserine-lactones in freshwater fish using the EIS method. The sensing principle relied on the decrement in the impedance signal induced by the analyte. Additionally, MWCNT and Nafion were mixed and immobilized on the surface of the GCE for the electrochemical determination of histamine in fish samples through its direct oxidation using the DPV technique. The presence of MWCNT improved the current of the oxidation peak and decreased the potential.<sup>104</sup> Moreover, MWCNTs can be mixed with MIP for detecting enrofloxacin in fish and shrimp samples using DPV, where MWCNTs were used as a conductive carrier.<sup>105</sup>

### Semiconducting nanostructures

Semiconducting nanostructures are intermediate between metals and nonmetals. Thus, they carry features from metals and non-metals. They have many desirable characteristics such as a narrow energy bandgap that improves the degradation of water pollutants.<sup>131</sup> These nanostructures include cadmium sulphide (CdS), copper oxide (CuO) copper ferrite (CuFe<sub>2</sub>O<sub>4</sub>), zinc sulphide (ZnS), and zinc selenide (ZnSe). CdS nanostructures have recently gained much attention due to their big role in various applications like solar cells,<sup>131</sup> supercapacitors,<sup>132</sup> catalysis,<sup>133</sup> optical sensors,<sup>134</sup> and electrochemical sensors.<sup>135</sup> For example, CdS semiconductor is broadly utilized in electronics because of its potential features such as electronic and electrochemical properties.<sup>136,137</sup> It was previously used to electrochemically detect formalin in mackerel fish samples using the CV technique. The NPs create many active sites for improving the electron movement at the surface of the electrode.<sup>17</sup>

CuO NP was used as an efficient electrocatalyst in the electrochemical synthesis of ammonia.<sup>138</sup> Recently, it has been incorporated with  $\beta$ -cyclodextrin to form a nanocomposite to monitor chloramphenicol drug in fish samples employing the SWV technique. The efficiency of this electrochemical sensor originated from the excellent catalytic properties of CuO NPs.<sup>106</sup>

CuFe<sub>2</sub>O<sub>4</sub> was used as a catalyst in many energy and environmental applications.<sup>139–141</sup> The CuFe<sub>2</sub>O<sub>4</sub> inverse spinel nanopowder was used for constructing a chemiresistive sensor to determine formalin in fish samples. The sensor showed excellent p-type response and resolution toward formalin gas.<sup>107</sup>

ZnS NPs were applied in many applications such as oxygen evolution electrocatalysis,<sup>142</sup> solar cells,<sup>143</sup> and optical sensors.<sup>144</sup> ZnS has recently been incorporated with copper

Table 2 NMNP-based electrochemical sensors for detecting analytes in fish samples<sup>a</sup>

NPs	Electrode material	Analyte	Fish	Technique	Mechn.	LOD (nM)	Ref.
Au	Au-XOR/MWCNT-PEDOT/GCE	Xanthine	Fish (local market)	DPV	Analyte oxidation	54.5	110
	Fe <sub>3</sub> O <sub>4</sub> @Au core-shell/CPE	Xanthine	Tuna fish	SWV	Analyte oxidation	400	111
	MWCNTs/Au/SPE	Xanthine	Fish (local market)	CV	Signal inhibition	1.14	112
	Au/rGO/GCE	CH <sub>3</sub> Hg <sup>+</sup>	<i>Carassius auratus</i> , <i>Channa argus</i> , <i>Cyprinus carpio</i> , <i>Pelteobagrus fulvidraco</i> , <i>Silurus asotus</i>	DPSV	Analyte reduction/oxidation	0.12 µg L <sup>-1</sup>	113
	poly(ester-urethane) urea/Au NPs/CPE	Hg <sup>2+</sup>	(local fish market)	CV		1.17	114
	Au nanoshells/LIPG	Sulfanilamide	Fish and shrimps (local market)	DPV	Analyte oxidation	0.000035	115
	MnO <sub>2</sub> /Au/GCE	CH <sub>3</sub> Hg <sup>+</sup>	Fish and shrimp (local market)	SWASV	Analyte reduction/oxidation	0.051 µg L <sup>-1</sup>	116
	BSA/gDNA-CdS/aptamer/chitosan/CuO-Au NPs/ITO electrode	3,3',4,4'-Tetrachlorobiphenyl	Fish muscle (local market)	PC	Modifier signal amplification	17.3 pg L <sup>-1</sup>	51
	MXene/g-C <sub>3</sub> N <sub>4</sub> /Au-COOH/GCE	GMP	Fish broth	DPV	Analyte oxidation	29.2	35
	Ru(bpy) <sub>2</sub> <sup>2+</sup> /sulfonatedMWCNTs/Au NPs/GCE	GMP	Fish broth	DPV	Analyte oxidation	7.36 × 10 <sup>-6</sup>	117
Ag	Ag NPs/Au electrode	Xanthine	Fish sample		Analyte oxidation	150	118
	Ag/MoS <sub>2</sub> /ITO	Xanthine	Rohu fish	DPV	Signal inhibition	27	119
	Au-Ag/rGO/poly(L-histidine)/GCE	Vancomycin	Fish sample	SWV	Analyte oxidation	0.00011	120
		Ceftriaxone				0.000017	
	Ag-Ag <sub>2</sub> O/MWCNTs/GCE	Histamine	Fish sauce samples (local store)	DPV	Analyte oxidation	0.000018	121
	L-Cysteine/Ag-Fe <sub>3</sub> O <sub>4</sub> /SPCE	Cu <sup>2+</sup>	Fish sample (local market)	DPAdSV	Analyte reduction	0.34	18
	K <sub>3</sub> Fe(CN) <sub>6</sub> /Ag NP@aptamer/GCE	Saxitoxin	Mantis shrimps	DPV	Signal amplification	1	122
Pt	PTA/CeO <sub>2</sub> @Pt NPs	Xanthine	Fish sample	DPV	Analyte oxidation	1036	29

<sup>a</sup> A: amperometry, BSA: bovine serum albumin, CA: chronoamperometry, CPE: carbon paste electrode, CV: cyclic voltammetry, DPV: differential pulse voltammetry, DPAdSV: differential pulse adsorption solvation voltammetry, DPV: differential pulse stripping voltammetry, ECL: electrogenerated chemiluminescence, GEC: graphite-epoxy composite magneto-actuated electrodes, LIPG: laser-induced porous graphene, LOD: limit of detection, MIP: molecularly imprinted polymer, MWCNT: multiwalled carbon nanotube, NP: nanoparticle, ITO: indium-tin oxide, PE: photoelectrochemical, PGE: pencil graphite electrode, rGO: reduced graphene oxide, RU: hexaammineruthenium(III) chloride, SPCE: screen printed carbon electrode, SWASV: square wave anodic stripping voltammetry.

Table 3 MOF-based nanomaterials for determining analytes in fish samples<sup>a</sup>

Electrode material	Analyte	Fish	Techn.	Mechn.	LOD (nM)	Ref.
Cu-MOF NPs/GCE	Hg <sup>2+</sup>	Canned tuna fish	DPV	Analyte reduction	0.06	123
Cu-MOF NPs/CPE	Xanthine	Fish sample (local market)	DPV	Analyte oxidation	0.89	124
	Hypoxanthine				2.1	
Ni-MOF/GCE	Histamine	Fish sample	CV	Analyte oxidation	0.31	125
Au NPs/Co-MOF-NH <sub>2</sub> /carbon paper	Pb <sup>2+</sup>	Fish and shrimp	ASV	Metal ion reduction followed by oxidation	0.34	126
	Cd <sup>2+</sup>				0.10	
ZrO <sub>2</sub> @Ni/Co-MOFs@Au NPs/GCE	Cd <sup>2+</sup>	Fish	DPV	Signal inhibition	1.7 × 10 <sup>-6</sup>	127
Ag NPs/Cu-MOF/GCE	Malachite green	Fish sample (local market)	DPV	Oxidation	2.20	48
AgNPs/Ni-MOF	Olaquinox	Fish muscle	DPV	Signal amplification	2.2	40

<sup>a</sup> ASV: anodic stripping voltammetry, CNFs: carbon nanofibers, CPE: carbon paste electrode, CV: cyclic voltammetry, DPV: differential pulse voltammetry, GCE: glassy carbon electrode.

indium sulfide (CuInS<sub>2</sub>) to form a nanocomposite with superior catalytic activity towards sulfadiazine drug oxidation. The nanocomposite was used as a brilliant electrode material for constructing an MIP electrochemical sensor to assess sulfadiazine in fish samples using the DPV technique.<sup>108</sup>

ZnSe is considered a crucial electronic and optoelectronic substance that is used in many fields like solar cells,<sup>145</sup> batteries,<sup>146</sup> and sensors.<sup>147</sup> As an electrode modifier, it was utilized to determine analytes in fish samples electrochemically. For example, ZnSe quantum dots decorated MWCNTs exhibited electrocatalytic activity towards pentachlorophenol oxidation and subsequent determination in carps. The DPV technique was used to monitor the sensor signal.<sup>109</sup> Additionally, the nanoflower-like ZnSe was incorporated with silver selenide (Ag<sub>2</sub>Se), cadmium telluride (CdTe) NPs, and chitosan onto indium-tin oxide to determine 3,3',4,4'-tetrachlorobiphenyl in coral reef fish using PC sensor. The ZnSe, Ag<sub>2</sub>Se, and CdTe nanomaterials showed high photocatalytic activity and improved the efficiency of photo-to-current conversion, leading to an enhancement in the PC sensor performance.<sup>52</sup> Fig. 6 illustrates the preparation of the nanocomposite and its immobilization onto the working electrode for the electrochemical determination of 3,3',4,4'-tetrachlorobiphenyl in coral reef fish *via* PC sensing.

The semiconducting nanostructures as sensing materials can detect fish analytes based on the optical, chemical, or

electrical changes occurring at the electrode surface in the presence of a fish analyte. At first, the fish analyte is adsorbed at the semiconductor-modified electrode leading to alterations in the semiconductor properties. Fish analytes can be directly detected based on the optical changes or electrical resistance changes of the semiconductor material.<sup>52,107</sup> On the other side, the semiconducting NPs can create many active sites to improve the electron movement at the surface of the electrode for direct detection of the fish analyte through its oxidation or reduction. For example, formalin,<sup>17</sup> sulfadiazine,<sup>108</sup> and pentachlorophenol<sup>109</sup> were detected based on their oxidation, while chloramphenicol was detected based on its reduction.<sup>106</sup>

### NMNPs

Noble metals refer to the chemically resistant and precious metals such as Au, Ag, Pt, palladium (Pd), ruthenium (Ru), rhodium (Rh), iridium (Ir), and osmium (Os). In the nanoform, they have a larger surface area, biocompatibility, and good optical, catalytic, and electrical features.<sup>148</sup> NMNPs have gained significant attention due to their wide range of applications across diverse fields. For example, in medicine, they can improve medicines' therapeutic index, reduce multidrug resistance, and enhance drug bioavailability in drug delivery systems.<sup>149</sup> Additionally, their role in diagnosing many diseases and determining biological and environmental molecules was reported.<sup>11,12,150</sup> Due to these attractive properties, many methods were followed to fabricate NMNPs.

NMNPs can be fabricated using top-down and bottom-up approaches. The top-down approach is physically achieved by breaking bulk metal into nano-dimension form *via* thermolysis, nanolithography, or  $\gamma$ -radiation.<sup>151,152</sup> Contrary to the top-down approach, the bottom-up one depends on the assembly of atoms to form NMNPs. This assembly can be accomplished *via* chemical, sonochemical, or electrochemical methods. The chemical method is based on the reduction of metal ions using reducing agents, followed by nucleation and aggregation to produce NMNPs.<sup>23</sup> The sonochemical method creates acoustic cavitation during the irradiation, which promotes the formation of free radicals and enables the metal ions to be reduced, forming NMNPs.<sup>153</sup> The electrochemical method is based on the reduction and subsequent deposition of NMNPs at the

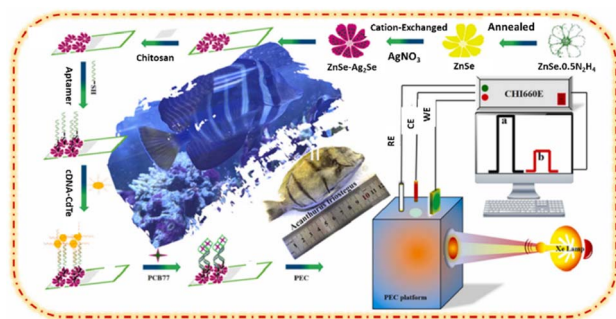


Fig. 6 Schematic illustration of PC sensor for determining 3,3',4,4'-tetrachlorobiphenyl in coral reef fish. Reproduced from ref. 52 with permission from Elsevier, copyright, 2022.





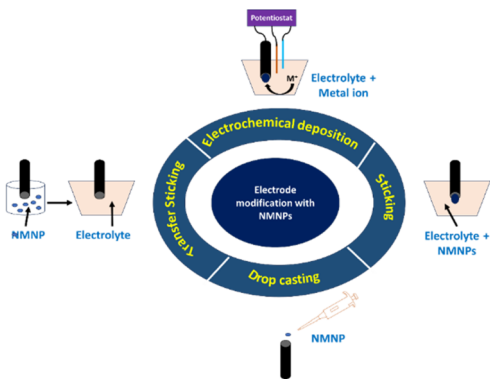


Fig. 7 A schematic illustration of electrode modification *via* electrochemical deposition, drop casting, sticking, and transfer-sticking techniques.

electrode or the formation of NMNPs in the electrolyte solution.<sup>154</sup> The electrode modified with the deposited NMNPs (*in situ* technique) can be directly used to detect target molecules. *Ex situ* techniques, such as drop casting, sticking, and transfer-sticking, can be followed for the electrode modification, as shown in Fig. 7.

In the drop-casting technique, the NMNPs are directly deposited on the electrode surface, while the sticking technique is based on the immersion of the electrode in the NMNP suspension mixed with the electrolyte. Additionally, the modification can be performed by transfer-sticking, where the electrode is immersed in the NMNP suspension for a particular time and then transferred into the cell containing the electrolyte.<sup>155</sup>

After electrode modification, the target molecules can be determined directly *via* their oxidation/reduction at the NMNP-modified electrode or indirectly based on the electrochemical response of the NMNPs or other redox mediators. In direct determination, the oxidation/reduction signal of the target molecule is improved in the presence of NMNPs owing to their increased surface-to-volume ratio which assists in immobilizing more molecules of the target analyte leading to an amplified signal.<sup>155</sup> Additionally, the high conductivity of NMNPs makes the electron transport at the electrode surface an easy process.<sup>156</sup> This electron transport improvement is attributable to the partial oxidation of NMNP under the applied potential, leading to the formation of active sites that facilitate the electrochemical oxidation/reduction of analyte at the electrode surface.<sup>21</sup> On the other hand, the target molecule can be determined based on the NMNP as a signal indicator. The amplification or inhibition in the NMNP signal depends on the chemistry of the target analyte.<sup>157</sup>

**Gold (Au).** Au NPs were extensively used for detecting target analytes using electrochemical sensors.<sup>158</sup> Recently, Au NPs immobilized on laser-induced porous graphene electrode by drop-coating have been used to detect sulfanilamide in fish and shrimp samples based on its oxidation. The current response of sulfanilamide on the surface of the modified electrode is higher than the response of the bare electrode because of the high

electrocatalytic activity of Au NPs.<sup>115</sup> Additionally, Au NPs can be mixed with other materials, such as reduced graphene oxide, metal oxide NPs, polymers, MWCNTs, and MXene for superior catalytic activity and sensitive determination of targeted analytes.

Au NPs were immobilized with reduced graphene oxide on the electrode surface for the detection of methyl mercury ( $\text{CH}_3\text{Hg}^+$ ) in *Carassius auratus*, *Channa argus*, *Cyprinus carpio*, *Pelteobagrus fulvidraco*, and *Silurus asotus* using DPSV, where  $\text{CH}_3\text{Hg}^+$  is firstly reduced and deposited on the surface of electrode as a preconcentration step, then, oxidation took place giving oxidation peak as a signal. Both Au NPs and reduced graphene oxide enhanced the sensing process due to their excellent catalytic features.<sup>113</sup>

Au NPs mixed with metal oxides, such as manganese dioxide ( $\text{MnO}_2$ ), CuO, and magnetic iron oxide ( $\text{Fe}_3\text{O}_4$ ) were used to determine analytes in fish samples. For example, the synergistic effect of Au NPs and  $\text{MnO}_2$  resulted in increased surface area, improved conductivity, and superior catalytic activity towards  $\text{CH}_3\text{Hg}^+$ . The detection process was based on the reduction of  $\text{CH}_3\text{Hg}^+$  into  $\text{Hg}^0$  and the subsequent stripping using the SWASV technique.<sup>116</sup> Additionally, a PC aptameric sensor based on CuO/AuNPs was used for the electrochemical detection of 3,3',4,4'-tetrachlorobiphenyl in fish muscles. The local surface plasmon resonance of Au NPs under the electromagnetic field induced charge separation and increased the photoelectric conversion efficiency.<sup>51</sup> Moreover,  $\text{Fe}_3\text{O}_4$ @Au core-shell was used to determine xanthine in tuna fish using SWV electrochemically.<sup>111</sup>

Au NPs incorporated with poly(ester-urethane) urea were utilized as a CPE modifier to determine  $\text{Hg}^{2+}$  in fish and shrimps obtained from a local market. Urethane and urea groups in poly(ester-urethane) were responsible for binding  $\text{Hg}^{2+}$  before the electrochemical detection. Owing to the poor electrical conductivity of poly(ester-urethane), Au NPs were added to increase the conductivity of the modification layer.<sup>114</sup> Additionally, Au NPs were mixed with poly(3,4-ethylenedioxythiophene) and MWCNTs to provide a favorable environment for fast electron transfer during xanthine oxidation.<sup>110</sup>

Au NPs/MWCNTs composite was used as an SPE modifier, which was treated with a mixture of two cross-linkers, *N*-hydroxysuccinimide and 1-ethyl-3-(3-dimethylaminopropyl carbodiimide), and then coupled with xanthine oxidase to determine xanthine in fish samples. The sensing mechanism was based on the inhibition of the modifier signal in the presence of xanthine.<sup>112</sup> Additionally, Au NPs/MWCNTs composite incorporated with 2,2'-bipyridine hydrated ruthenium ( $\text{Ru}(\text{bpy})_2\text{Cl}_2$ ) was used to determine 5'-guanosine monophosphate based on its oxidation at the GCE.<sup>117</sup>

Au NP/MXene/g- $\text{C}_3\text{N}_4$  composite has recently been reported as a GCE modifier to detect 5'-guanosine monophosphate. The carboxylated AuNPs exhibited high electron transfer efficiency, while MXene and g- $\text{C}_3\text{N}_4$  showed strong adsorption capacity and specific catalysis, respectively. The 5'-guanosine monophosphate was detected based on its oxidation at the modified GCE *via* two-proton and two-electron transfer.<sup>35</sup>



**Silver (Ag).** Ag NPs and their composites were used as electrode modifiers to determine many analytes electrochemically.<sup>155,159–161</sup> Their role in constructing electrochemical sensors to detect analytes in fish samples has recently been highlighted. For example, Ag NPs have been examined to determine xanthine in fish through amperometric biosensing.<sup>118</sup> Additionally, they were incorporated with other NMNPs, metal oxides, and metal sulphides to improve the measurement process. For example, Ag/Au nanocomposite has been reported as a GCE modifier to determine vancomycin and ceftriaxone in fish with a high performance. The assessment mechanism was based on the oxidation of vancomycin and ceftriaxone at the modified GCE using a SWV technique.<sup>120</sup> Also, a composite of Ag–Ag<sub>2</sub>O NPs and MWCNTs was used as an electrocatalyst to determine histamine in fish sauce based on its oxidation *via* the DPV technique. Ag–Ag<sub>2</sub>O layer exhibited improved surface area, catalytic activity, and reduced overpotential towards histamine.<sup>121</sup> Additionally, Ag nanowires have been incorporated with magnetic NPs onto an SPE electrode to assess Cu<sup>2+</sup> in fish samples based on its reduction using differential pulse adsorption solvation voltammetry technique. The composite was functionalized with L-cysteine to capture Cu<sup>2+</sup> *via* Ag–S bond.<sup>18</sup> Ag NPs were also used to construct an aptamer-based electrochemical sensor to determine saxitoxin in mantis shrimps. The sensing process was based on the effects of saxitoxin and aptamer/Ag NPs on the Prussian blue signal.<sup>122</sup> Moreover, Ag/MoS<sub>2</sub> nanosheets were used to determine xanthine in Rohu fish based on the modifier signal inhibition with increasing xanthine concentration.<sup>119</sup>

**Platinum (Pt).** Pt NPs were incorporated into electrochemical sensors to determine a variety of biological analytes owing to their high electrical conductivity and electrocatalytic activity.<sup>162,163</sup> Recently, Pt NPs, which were incorporated with cerium dioxide and polymer 3-amino-5-mercapto-1,2,4-triazole, have been reported as an electrocatalyst to determine the concentration of xanthine, for evaluating fish freshness.<sup>29</sup> Xanthine was determined based on its oxidation at the modified electrode using a DPV technique.

### MOFs for electrochemical determination of analytes in fish samples

MOFs are crystalline frameworks obtained by strong interaction between metal nodes and organic linkers.<sup>164</sup> They are hybrid materials that combine the flexibility of organic materials and the rigidity of inorganic systems.<sup>65</sup> They have many advantages such as increased surface area, porosity, plentiful active centers, and controllable morphology. Thus, it was used in many applications such as batteries,<sup>165</sup> supercapacitors,<sup>166</sup> and electrocatalysts.<sup>167</sup> Due to these significant applications, MOFs were prepared using mechanochemical, hydro/solvothermal, sonochemical, microwave, and electrochemical techniques, as shown in Fig. 8. The mechanochemical is a solvent-free technique, where the metal salt and ligand are ground together in a mortar and pestle or a ball mill to obtain the MOF material.<sup>168</sup> The hydro/solvothermal technique, the commonly used technique, is based on dissolving metal salts and ligands in aqueous

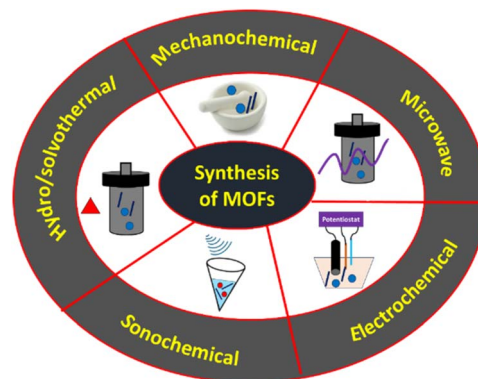


Fig. 8 Schematic illustration of MOF synthesis techniques.

or organic solvents before heating the solution at high temperatures and under elevated pressure for a particular time.<sup>169</sup> MOFs can also be obtained at room temperature by mixing the metal salt and ligand in the solvent for a particular time.<sup>170</sup> In the sonochemical technique, an ultrasound is applied to improve the chemical/physical changes in a liquid medium through a cavitation process, where growth and subsequent destruction of bubbles occur. These bubbles collapse leading to an energy release which is responsible for nucleation site formation and finally MOF production.<sup>171</sup> They can also be produced electrochemically at electrode surfaces by reducing metal ions and subsequent deposition with ligands at the electrode surfaces as MOFs.<sup>19</sup> The electrode with deposited MOFs can be directly used to determine target molecules. Additionally, MOFs prepared by other techniques can be adsorbed at electrode surfaces with binders. These MOF-modified electrodes have high electrocatalytic activities due to the large surface area of MOFs and the presence of pores that can adsorb more analyte molecules leading to an enhancement in the electrochemical signals.<sup>25</sup> However, the poor electrical conductivity of many prepared MOFs with crystal structures limits their utilization in electrochemical sensing. So, MOFs were prepared in the nanoform, prone to an amorphization, or mixed with conductive materials such as NMNPs to improve their electrical conductivity.<sup>19,172</sup> Accordingly, many various MOF materials were studied for detecting multiple analytes.<sup>55,173</sup> Herein, we are focused on MOF NPs and their composites with NMNPs as promising materials to determine analytes in fish samples.



Fig. 9 An illustration of the electrochemical determination of Hg<sup>2+</sup> in tuna fish at Cu-MOF NP-modified electrode. Reproduced from ref. 123 with permission from Elsevier, copyright 2020.



MOF NPs, such as Cu-MOF NPs, were previously used as electrocatalysts to determine  $\text{Hg}^{2+}$ , xanthine, and hypoxanthine in fish samples.<sup>123,124</sup> The determination of  $\text{Hg}^{2+}$  was achieved based on its reduction at the modified GCE. The reduction peak was monitored by CV and DPV techniques.<sup>123</sup> Fig. 9 exhibits an illustration of the Cu-MOF NP-modified GCE used for the determination of  $\text{Hg}^{2+}$  in canned tuna fish. Firstly, the Cu-MOF was adsorbed on the GCE in the presence of a linker containing hydroxyl and amino groups to adsorb  $\text{Hg}^{2+}$  before its reduction. The canned tuna fish sample containing  $\text{Hg}^{2+}$  was analyzed by the electrochemical sensor showing a distinctive peak corresponding to  $\text{Hg}^{2+}$  reduction.

On the other side, xanthine and hypoxanthine were determined based on their oxidation *via* two-proton and two-electron transfer at Cu-MOF NP-modified electrode using the DPV technique.<sup>124</sup> Moreover, Ni-MOF was transformed into Ni@C NPs with large surface area to act as a GCE modifier for the assessment of histamine in fish samples based on its oxidation using the CV technique.<sup>125</sup> In addition to MOF NPs, the MOF composites with NMNPs were used to determine fish sample analytes.

MOFs incorporated with NMNPs, such as Au NPs and Ag NPs, were evaluated as promising materials for determining pollutants in fish samples. For example, Au NPs/Co-MOF-NH<sub>2</sub> as a carbon paper modifier was utilized to determine  $\text{Pb}^{2+}$  and  $\text{Cd}^{2+}$  in fish samples. The metal ions,  $\text{Pb}^{2+}$  and  $\text{Cd}^{2+}$ , were reduced and then oxidized using ASV technique. The presence of AuNPs increased the anodic peak of  $\text{Cd}^{2+}$ , while the addition of MOF-NH<sub>2</sub> to AuNPs/carbon paper led to the appearance of two distinguished peaks corresponding to  $\text{Pb}^{2+}$  and  $\text{Cd}^{2+}$  with high oxidation currents.<sup>126</sup> Additionally, Au NPs/Co-MOF incorporated with zirconium dioxide was used as a GCE modifier to construct an aptameric sensor to determine  $\text{Cd}^{2+}$ . The sensing process was based on the inhibition of the modifier signal by increasing the concentration of  $\text{Cd}^{2+}$ .<sup>127</sup> Ag NPs were also incorporated with Cu-MOF and Ni-MOF to respectively determine malachite green and olaquinox using the DPV technique. The malachite green was determined based on its oxidation at the modified electrode, while olaquinox was determined based on the signal amplification of the  $\text{K}_3[\text{Fe}(\text{CN})_6]$  probe with increasing olaquinox concentration.<sup>40,48</sup>

## Summary and perspective

Fish is a crucial source of nutrition for millions of people worldwide. Therefore, their quality in terms of safety and freshness is gaining significant attention. Fish safety can be achieved by minimizing the risk of contamination with various pollutants such as hazardous chemicals. The concentrations of these chemicals should be determined to evaluate fish safety. Fish freshness evaluation is also accomplished by determining chemical indicators, such as xanthine, hypoxanthine, 5'-guanosine monophosphate, uric acid, and histamine. Compared to the traditional chromatographic methods, the electrochemical methods, such as voltammetric, chemiresistive, and PC, represent simple and robust tools for fish quality evaluation.

Nanomaterials, such as NMNPs, MOF NPs, and their composites, were introduced as efficient electrode modifiers for superior improvement in the response of the electrochemical sensors toward fish analytes. The efficiency of these nanomaterials over the other electrode modifiers comes from their large surface area-to-volume ratio, high electrical conductivity, selective interaction with the targeted analytes, and boosted electrocatalytic activity. For example, xanthine, the common freshness indicator, was determined by using Au/MWCNTs, Au-xanthine oxidoreductase/MWCNT,  $\text{Fe}_3\text{O}_4$ @Au core-shell, Ag NPs, polymer/ $\text{CeO}_2$ @Pt NPs, and Cu-MOF NPs. Among these electrode modifiers, Cu-MOF NPs showed the highest electrocatalytic activity towards xanthine with the lowest detection limit. Additionally, Cu-MOF NPs as an electrode modifier were used to assess  $\text{Hg}^{2+}$  with a lower detection limit than Au nanocomposite. Moreover, Ag nanocomposite can assess histamine with a lower detection limit than Ni-MOF NPs. The differences between the sensitivity of different nanomaterials towards the same analyte are attributable to the electrochemical method used in monitoring the signal, the addition of other conductive materials such as polymers and MWCNTs, or the chemical structure of the analyte and its affinity to the modifier.

Although these modified electrochemical sensors have many advantages, they also have some challenges, such as the complex sensor design, the ineffective *in situ* real-time analysis, and the limited range of detectable targets. Future developments could focus on developing electrochemical sensors based on micro-electrodes which could be used for the *in situ* analysis to facilitate real-time determination of fish analytes. Additionally, developing electrode modifiers with different recognition probes could allow the determination of various analytes simultaneously.

## Conclusion

Nanomaterial-based electrochemical sensors are regarded as a favorable tool for detecting chemical pollutants, such as heavy metals, pesticides, dyes, drugs, preservatives, and toxins, as well as freshness indicators, such as xanthine, hypoxanthine, uric acid, and histamine in fish samples. Various nanomaterials, such as electrospun nanofibers, MWCNTs, semiconducting nanomaterials, NMNPs, and MOF NPs, were reported as effective electrode modifiers to determine analytes in fish samples. In this review, a great focus has been put on NMNPs, MOF NPs, and their composites owing to their increased electroactive surface area, boosted electrocatalytic activity, and high electrical conductivity. It also highlights the synthesis methods of these nanomaterials and their electrochemical performance to assess the fish analytes. Overall, NMNPs and MOF NPs are promising materials for developing electrochemical sensors to evaluate fish quality with a great ability to be introduced into real-world applications.

## Data availability

No primary research results, software or code have been included and no new data were generated or analysed as part of this review.



## Author contributions

Moustafa Zahran: conceptualization, writing original draft. Shuang Tian: writing original draft, Ji Li: writing review & editing. Amal H. Marei: writing original draft. Ying Xie: writing original draft. Qiyu Liu: writing original draft. Jialin Huang: writing original draft. Dezhi Wang: writing original draft. Xingyao Ning: writing original draft. Jiahao Wang: writing original draft. Huizhong Chi: writing review & editing. Xiaolin Li: supervision, writing review & editing.

## Conflicts of interest

There are no conflicts to declare.

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