Review:

Recent Advances and Future Prospects of Molecular Imprinting Polymers as a Recognition Sensing System for Food Analysis: A Review

Almajed Asaad Abdullah Sfoog[1], Norlaili Abu Bakar[1*], Nurulsaidah Abdul Rahim[1], Wan Rusmawati Wan Mahamod[1], Norhayati Hashim[1,2], and Siti Kamilah Che Soh[3]

[1]*Department of Chemistry, Faculty of Science and Mathematics, Universiti Pendidikan Sultan Idris, 35900 Tanjung Malim, Perak, Malaysia*

[2]*Nanotechnology Research Center, Faculty of Science and Mathematics, Universiti Pendidikan Sultan Idris, 35900 Tanjong Malim, Perak, Malaysia*

[3]*Faculty of Science and Marine Environmental, Universiti Malaysia Terengganu, 21030 Kuala Nerus, Terengganu, Malaysia*

** **Corresponding author:***

tel: +6015-48797323
email: norlaili@fsmt.upsi.edu.my

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Abstract: *Molecular imprinting polymers (MIPs) have been widely used to produce stable polymeric materials due to their highly selective binding sites to determine the analyte (target molecule) in food products. MIPs begin with a complex compound between the template molecule and the functional monomers that can be polymerized when there is a closely crossed link. MIPs left specific cavities after the removal of templates during washing, which complements the size and shape of the templates. The use of MIPs has contributed to novel advances in materials science, polymer science, natural science, and other multi-disciplinary systems. Optical chemical sensor is an exciting field in MIPs today due to comprehend the unique affirmation limit of associated polymers giving stable polymers with high molecular recognition capabilities. MIPs display a wide extent of relevance, incredible flexibility, security, and high selectivity; their internal affirmation districts can be explicitly gotten together with design molecules to achieve specific affirmation. This review covers the various achievements of sensors used in laboratory analyses. The advancement in the development of MIPs is evaluated with an accentuation on the preparation principle, the discovery process, the molecular recognition mechanism and future perspectives and challenges for MIPs in building an optical chemical sensor.*

Keywords: *molecularly imprinted polymer; optical chemical sensor; fluorescence sensor; food analysis; molecular recognition*

■ INTRODUCTION

Molecularly imprinted polymers (MIPs) are polymers created from molecular imprinting technology, which is a proficient technique for delivering useful materials with explicit recognition locales that are reciprocally mass-fitted and size-to-format atoms. It is noted that these polymers should be inflexible to hold a lasting memory in the imprinting process and that the imprinted areas must be widely coordinated to enable the layout atoms to effectively diffuse in and out [1-2]. The assortment of approaches for creating manufactured receptors can be categorized into reversible covalent or non-covalent (hydrogen, ionic, and Van der Waals) techniques, depending on the cooperation between the monomer and the layout. Generally, most MIPs are arranged by mass polymerization, which has a poor site of objective particles and a low rebinding limit because of a thick polymeric matrix [3].

To overcome the downsides of mass polymerization, increasingly modern and complex polymerization strategies have been proposed to acquire

MIPs with various structures, such as dots, solid structures, and coatings. At first, globules are commonly used for pressing segments for chromatography or solid-phase extraction (SPE) [4] and are normally integrated by suspension polymerization [5], seed polymerization [6], emulsion polymerization [7], and precipitation polymerization [8]. Solid packing polymers are used for *in situ* segments that are prepared to be used directly after the evacuation of the format [9-10]. Next, some MIPs rely on optical sensors with various materials to provide an overview and are discussed between optical and electrochemical sensors [11]. Besides, multi-step or one-advance extending and polymerization systems [12], electrochemical polymerization [13], joining on the strong segment, photo joining, and sol-gel methods have furthermore been used [14]. Additionally, surface etching over a nano-evaluated circle reinforces materials with a colossal express surface locale is reasonable for targeted analytes to make sharp MIP nano-circles [15-19]. Smart materials reflect ordinary receptor characteristics; for example, they help to set up responsive MIPs with redesigns of responsive polymers as the cross-section for sub-nuclear etching. Meanwhile, enhancements of responsive MIPs, including appealing responsive MIPs, pH-responsive MIPs, photo-responsive MIPs, temperature-responsive MIPs, and double- or multi-redesigns responsive.

The principle of MIPs is integrated sensing techniques including the monomer-template mixture is synthesized in the presence of a cross-linking agent and a suitable porogen solvent and then polymerized to generate a microporous or macroporous structure and mechanical stability that is solidified using an excess of a cross-linking agent to form the MIP [20]. During the crosslinking process, monomers were trapped into the MIP network along with the template and the solvent occupying the pores using the photopolymerization method for synthesized MIPs [21]. After removing the templates, the MIPs contain complementary functionalized cavities whose size and form mimic those of the template molecule [22]. These functional cavities have been designed precisely to recognize the template molecules in a specific and selective manner [23-25].

Over time, scientists in various fields have modified and developed the techniques for producing MIP, and several methods and applications that used the molecular recognition capacity were implemented [26]. This technique offers a convenient method of molecule recognition to be applied in sensors [27-29], optical sensors [30-32], chemical sensors [33-34], electrochemical sensors [35-37] and so forth.

■ SYNTHESIS OF MOLECULARLY IMPRINTED POLYMER

MIPs are prepared in the presence of an analyte that directs the organization of functional groups on functional monomers into molecular stamping processes and the template works for the formation of complementary binding sites of a polymer. Certainly, molecularly imprinted materials have increased their forthcoming usage due to their high affinity and selectivity, chemical and physical stability, in addition to reproducibility and simplicity of their preparation method [38].

MIPs also has good properties of durability, resistance to high temperatures and pressure, as well as ease in production of these industrial receptors to solve problems in the field of chemical separation [39-41], SPE [42-44], chemical sensing [45-46], optical sensor [47-49] and drug development [50-51]. The selectivity of MIPs is directly related to the recognition of the target molecule, which is used as a template in the imprinted polymer process. The design and synthesis of MIPs are complex when dealing with the chemical bond, including the nature and level of the template, the functional monomer, the cross-linker, the solvent, the initiator, and the time as well as the polymerization methods.

In the synthesis of MIPs, Bhawani et al. [52] described those three different imprinting approaches are used in the process of preparing the MIPs containing the covalent imprinting approach, the non-covalent imprinting approach, and the semi-covalent imprinting approach. The covalent imprinting approach demonstrates that reversible covalent bonds are formulated between the template and the monomers

before the polymerization process. In the covalent bonding process, the template could be successfully removed by using the cleavage of these bonds to reform the rebinding of the target molecule. The covalent approach to the synthesis of MIPs could lead to the binding of homogeneous population sites by ensuring a high level of template-monomer stability. The approach is regarded to be highly restrictive in the presence of covalent bonds that are always required rather than experienced in harsh conditions [53]. Bhawani et al. [52] described that the semi-covalent approach is associated with intermediate options by which the template covalently bounds to the functions of the monomer and, the template rebounds based on the non-covalent interactions. The non-covalent approach is another important approach that could be described as more selective in their internal recognition sites adopted in the synthesis of MIPs, which described non-covalent interactions as relatively weaker interactions between the molecular template and functional monomers before the initiation of polymerization process [54]. The non-covalent methodology is also highly effective and versatile in molecularly imprinting which is quite similar to the recognition of patterns observed in nature, based on the interaction process between the monomer and the template that forms the complex through non-covalent or covalent interactions [55]. The association, as well as the disassociation of the imprint, could happen in the plain diffusion through in and out of the sites. The approach is very useful in the process of preparing MIPs by simplifying and determining the availability of different monomers to interact with the type of template for any type of imprinting process [56]. This approach is also associated with some drawbacks, as the interactions of template monomers are usually managed through the equilibrium process. A higher amount of monomer can be successfully used to show the equilibrium in the formation of the template-monomer complex. Excessive free monomers have been randomly incorporated into the polymeric matrix, which could lead to heterogeneous formation or binding of non-selective sites [57]. This non-covalent strategy has been preferred in the preparation of MIPs despite the drawbacks of the MIP preparation approach, including leakage of the template molecule and heterogeneity at the binding sites [58-59]. The methodology can be easily carried out and the removal of templates could easily practice by simplifying the solvent extraction [60].

MIP is necessary to correspond to the functions of the monomer with the function of the targeted analyte in an integrated way, whereas the chemical properties are crucial in the imprinting process. Functional monomers, such as methacrylic acid (MAA), are the choices for polymer synthesis in biological reactions [61].

A novel preparation method was conducted for using fluorescein methacrylate monomer as the fluorescence functional monomer in the synthesis of MIPs to estimate the sensing template for selective recognition [62]. Generally, the monomers are appropriately selected for the chosen template to include the distribution of the heterogeneous binding site in the polymerization with excellent binding capacity.

Monomer

The functional monomers shall be carefully selected because they play an important role in enhancing the formation of monomer-template complexes that carry acid-base functional groups, with solid associations with the format. However, this may lead to an increase in non-specific binding sites and assist in the formation of strong reactions between the functional monomers and the template during an imprinting process. Some of the functional monomers (acidic or basic) are the most widely used and widespread for forming imprinted polymers such as MAA in MIP formation, MAA is a popular and excellent functional monomer in creating accurate MIPs where they can be obtained commercially [63]. In another study, a basal monomer was chosen to calculate the dynamic properties for preparing a molecularly imprinted polymer complex [64].

On the other hand, in a recent study by Janczura et al. [65], five different functional monomers including methacrylic acid, 1-vinylimidazole, 4-vinylpyridine, allylamine and 1-allyl-2-thiourea were used separately

with cross-linker in the presence of different templates to synthesize selective molecularly imprinted polymer. The 1-vinylimidazole as a functional monomer with the template (4-hydroxyphenylacetic acid) in MIP leads to higher specificity as well as good selectivity with high affinity compared to other functional monomers. Affinity interactions are developed to form hydrogen bonds between these functional monomers with templates. Therefore, this leads to the development of recognition elements in MIPs and differences from NIPs were evaluated.

Normally, in a non-covalent imprinting approach, most researchers have focused on the selection of the functional monomer that is used in the recognition of molecularly imprinted materials for the pre-polymer compound [66]. However, the compositional rule of traditional MIPs has often been relatively difficult and complex dependent on the type of functional monomer [67]. In contrast, MIPs provides a simple and uncomplicated compositional rule according to the template-functional monomer ratio in the MIP synthesis [59]. MIPs were synthesized by the functional monomers [68], MIPs are also mechanically stable and highly selective, thus this method was developed for pre-treatment methods [69].

Template

The template is a key molecule for the preparation of an imprinted polymer that directs the organization of functional groups on functional monomers into molecular stamping processes. MIP has a specific binding capability with a specific affinity for the template based on imprinting approaches if the imprinting process is successful [70-71]. The template is selected for polymer imprinting based on its chemical and physical properties, including size, shape and functional group, as well as solubility in organic solvents, where templates linked to the MIPs have been successfully selected in the presence of multiple suitable organic porogen solvents, such as *p*-xylene [72] and formaldehyde templates [73]. More importantly, the templates possessed by MIP must be chemically inert and stable under these free radical polymerization conditions, produced either by a photochemical or thermochemical method and must prevent or inhibit free radical polymerization. In this case, the composite MIP prepared with the template must be reactive in the imprinted polymer and the advanced sensors must provide suitable selectivity when using a single MIP chemical sensor template for selectivity testing [74] and a MIP chemiluminescence sensor to detect pyrethroids as a molecular template [75]. Whereas Altintas et al. [76] synthesized nanoparticles using MIP, and diclofenac was chosen as a successful template for developing the optical sensor surface. The high-affinity nano MIP study indicates good potential for the detection of diclofenac in water with the nano MIP sensor.

In all atomic imprinting forms, the form assumes an essential job, and the molecular structure decides the sort of useful monomer to be used in the amalgamation since the compound bonds between them prove molecular recognition. The form should be artificially inactive in a perfect world under polymerization conditions and stable under union conditions e.g., temperature). MIP was prepared by thermal polymerization for functional monomers where dimethyl methyl-phosphonate was used as a template [77]. In other cases, when the template is being removed from the polymer after the washing process leaves empty cavities, MIP is considered to be a physical and chemical supplement to the molecular of the left template; cavities are formed in the polymer and complement the size and shape of the template, and it can affect the polymer selectivity. Hence, a dummy template can be used instead of the required template for the functional group in its structure when it does not have the required target [78]. Recently, MIPs have been reported for the detection of cobalt, cadmium, and lead ions as good templates in the aqueous medium. Prepared ion-imprinting polymers have powerful and good selectivity [79].

Cross-linker

The cross-linker has a stable and steady state; the cross-linker connects the building monomers to the template inside the polymer matrix [80]. An appropriate

cross-linker must have the capacity for polymerization under a free radical initiator, and should not be sensitive to any reactions to the analyte. A choice of appropriate components with functional monomers is available to vary degrees [81]. The major purpose of crosslinking monomers is to steady the binding sites within the imprinted polymer, and a large amount of cross-linker monomers can also be used in place by forming a high-strength solid polymer network to maintain MIP binding sites [82]. MIP synthesis also depends on cross-linking as polymer bond, ethylene glycol dimethyl acrylate (EGDMA) is a common cross-linker used in MIP synthesis for various applications because it has two active ethylene sites in its structure. During the polymerization process, the cross-linker (EGDMA) of the molecules is strong and compact with each other between the linear molecules that create the polymer network in an effective study to estimate the effect of cross-linker, monomer and polymer on the controlled delivery of dexibuprofen [83].

The effect of cross-linking density on polymer morphology and polymer-template recognition in bupivacaine molecularly imprinted MAA–EGDMA copolymers were examined by the replacement of EGDMA with methyl methacrylate (MMA). The use of MMA as a common monomer during the cross-linking process with EGDMA affects the morphology and recognition characteristics of molecularly imprinted MAA–EGDMA copolymers, which are the most commonly used polymer system in molecular imprinting science and technology [84-85]. In addition to the optimal combination, trimethylolpropane trimethacrylate and EGDMA produce rigid polymers with high levels of binding sites that are used in forming an imprinted polymer [86].

For sol-gel imprinting, (3-aminopropyl)triethoxy silane (APTES) was chosen as a functional monomer because of its amino group that interacts with templates via hydrogen bonds. Tetraethoxysilane acts as a cross-linker to form a polymeric siloxane network through the Si–O bond via hydrolysis [87]. The selection of EGDMA as a cross-linker for the preparation of a novel nanomaterial of the $Ni(OH)_2$ nanoarrays electrode was applied as a sensing platform for the detection of sulfapyridine in MIP-based electrochemical sensors [88]. A study of the chemical structure of monomers, crosslinkers and initiators used in molecular imprinting was carried out by Cormack and Elorza [89].

Initiator

Generally, MIP sensors depend on initiators, especially free radical polymerization, which is one of the best polymerization methods currently used to convert vinyl monomers into polymers under moderate reaction conditions. Azobisisobutyronitrile (AIBN) [90-91], 4,4′-azobis(4-cyanovaleric acid) [92], azobisdimethylvaleronitrile [93] and benzoylperoxide [94] are few suitable initiators for MIP synthesis.

The importance of the functionalized radical initiator (4,4′-azobis(4-cyanovaleric acid)) has been demonstrated by standard precipitation polymerization in the formation of highly selective polymeric molecules [55]. The radical initiator is used to prepare MIPs for a synthesis process that achieves good capabilities to improve the functioning of the molecularly imprinted synthetic receptors [55]. In contrast, 2,2-AIBN was used as a photoinitiator for photopolymerization and the prepared polymer was exposed to UV light for 3 h [95-96]. However, Lim and Holdsworth's study [97] observed that the selectivity and the binding efficiency of the template increased with an increase in the initiator concentration in MIPs.

Furthermore, the initiator must include significant importance in the reaction mixture under the polymerization conditions [97]. Finally, polymerization is usually done by the presence of an initiator and by the formation of free radicals in photochemical, thermal, or chemical ways. The photoinitiators are more common in imprinting technology and their concentrations are very low compared to monomers in MIP processes. 2,2-AIBN as an initiator has been applied in MIPs processes such as MIP sensor [98] and chemosensor [99]. An overview of the template, initiator, monomers, cross-linkers, type of polymerization and solvents used in the synthesis of molecularly imprinted polymer is listed in Table 1.

■ FUTURE PROSPECT OF MIP AS A RECOGNITION SENSING SYSTEM

MIP-Based Optical Sensors in Food Analysis

Optical sensors have MIPs as a recognition section in sensor development. Fluorescent technology is also the most common technique used in MIP optical sensors that converts changes in the optical properties of materials into analytical signals [112] and stationary phase chromatography [113]. The MIPs appear to be sufficiently effective to react selectively with their analyzing capacity; thus, increasing their usage in sensor systems as shown by the increase in the number of publications. Many analyses can be identified by optical sensors with a specific sensibility due to MIPs. The MIP optical sensor has drawn a great deal of attention to the potential for food and environmental safety, whereas the MIP also has a stable sensitivity and high accuracy for detecting τ-fluvalinate in real samples [114]. The simple colorimetric method introduces a MIP-based optical sensor to enhance detection capabilities that can identify 3-phenoxy benzaldehyde in real samples [115]. In this manner, several studies are using MIP to develop optical sensors that can be used to produce a sensor for detecting chlorpyrifos in drinking water [116] and apple juice [117].

Optical sensing depends on the kind of reaction taking place, which can either be chemical or physical in order to obtain an analytical signal. Using this technique, it is possible to show the binding of the analytes with the polymer if the reactions entering the polymer are very strong. Regardless, these strategies are complicated and time-consuming. The optical sensor does not need a second analytical stage and is therefore a promising method. The system depends on the adsorption of the target molecule in the MIP [118].

The polymer state can provide information about MIPs once the polymer has been synthesized. MIP optical sensor fabrication involves the use of a non-covalent self-assembly approach by adding layers of silica

Table 1. Some materials were suitable for MIP sensors in food analysis

Template molecule	Functional monomer	Cross-linker	Initiator	Porogen solvent	Preparation method	Application in real samples	Recovery (%)	Ref.
Di (2-Ethylhexyl) phthalate (DEHP)	Methacrylic acid (MAA)	Ethylene glycol dimethacrylate (EGDMA)	2,2′-azobisisobutyronitrile (AIBN)	Toluene	Precipitation polymerization	Not mentioned (its aqueous solution)	-	[100]
Andrographolide	MAA	EGDMA	Benzoyl peroxide (BPO)	Acetonitrile: toluene (3:1)	Precipitation polymerization	Not mentioned (its aqueous solution)	-	[101]
Metsulfuron-methyl Chlorsulfuron	4-Vinylpyridine (4-VP)	Divinylbenzene (DVB)	AIBN	Acetonitrile	Precipitation polymerization	Crops, vegetables, and oils samples	74.8–110.5	[102]
Diclofenac (DFC)	MAA	*N,N*-Methylenebis(acrylamide) (MBAA)	Potassium persulfate (KPS)	Acetonitrile/ toluene (1:1)	Precipitation polymerization	Water samples	-	[103]
Diazepam (DZM)	MAA	EGDMA	BPO	Chloroform	Bulk polymerization	Serum samples	95.31	[104]
Atenolol	MAA	EGDMA	BPO	Propanol	Bulk and precipitation polymerization	Serum samples	95.46	[105]
Erythromycin (ERY)	Acrylic acid (AA)	*N,N′*-Methylene bisacrylamide (MBA)	AIBN	Acetonitrile	Bulk polymerization	Water samples	-	[106]
Malachite green	MAA	EGDMA	AIBN	Acetonitrile	Bulk polymerization	Fish muscles	100	[107]
Climbazole (CBZ) Clotrimazole (CMZ) Miconazole (MNZ)	MAA	EGDMA	AIBN	Toluene	Emulsion polymerization	Fish samples	89.2–101.5	[108]
Aflatoxins	MA	DVB	AIBN	Dimethyl sulfoxide (DMSO)	Emulsion polymerization	Fish feed samples	93	[109]
Ziram	MAA	EGDMA	AIBN	Chloroform	Suspension polymerization	Not mentioned (its aqueous solution)	96.76	[110]
p-hydroxybenzoic acid (*p*-HB)	4-VP	EGDMA	AIBN	Acetonitrile	Suspension polymerization	Water samples	70	[111]

nanoparticles to MIPs as thin film and has been developed and tested for the discovery of testosterone in aqueous solutions by HPLC. This technique was first used to create a testosterone photosensitive sensor in aqueous samples [119]. Recently, in any case, the strategy for molecularly imprinted films has been appropriately distinguished; approaches based on molecularly imprinted films may provide an easy, fast, and cost-effective way to perform sensitive electrochemical measurements [120]. Thus, a few MIP-based audits to improve optical sensors can be found depending on the nature of the analyte molecule. Therefore, we need to know the concentration of the analyte and the monomers in the MIP of various materials as it provides from an optical discovery perspective.

Complex food frameworks are difficult to recognize via traditional methodological strategies; hence fluorescent treatment is required [121]. Recently, MIP-based mimetic sensors with great precision and simple pre-treatment techniques have been a functioning exploration territory [122]. MIP-based detecting frameworks offer extraordinarily favorable circumstances over regular diagnostic systems; they remember a high particularity for constant investigation of complex blends, ease, and straightforward activity without the requirement for broad-based pre-treatment. MIPs have just been applied to a wide variety of optical sensors such as fluorescence and conventional semiconductor [123]. Molecularly imprinted silica film takes into consideration the arrangement of sensors for practically a wide range of analytes for the detection of cyfluthrin (CYF) in fish and sediment samples; this method developed MIP-based rapid sensor for analyzing CYF in fish and sediment samples. The limit of detection (LOD) of CYF in fish and sediment samples was found to be 1.0 and 1.3 μg/kg, respectively [124]. A new strategy has been proposed for estimating cypermethrin (CYP) in fish samples using MIP-based silica layers as an Opto-sensing material method, which is an accurate, stable, and sensitive method for determining CYP in fish samples under optimal conditions, with a detection limit of 1.2 μg/kg [125]. Furthermore, another type of MIP silica layer has been made through selective surface grafting technology for saxitoxin in shellfish samples at a LOD of 0.3 μg/kg [126]. On the other hand, there is a way to modify MIP-based chemical sensors using multi-walled carbon nanotubes to detect melamine in food products, with a detection limit of 5.6×10^{-13} mol/L [127]. The quality of food products is currently determined in terms of safety and security for human consumption.

MIP-Based Electrochemical Sensors in Food Analysis

These detection procedures are principally used in applications to fluid sample; the MIP is applied to the working terminal that is gathered with the counter and reference cathodes in the cell. Because the estimation is currently based on a consistent potential, the strategy is called amperometric, and current estimations during changing possibilities are known as potentiometric techniques. The pinnacle current and voltage are checked relative to the grouping of the objective particle. The number of varieties of these two general techniques can be overpowering, but the essential ideas continue as before [128].

There are many methods of MIP technology that have been invented and reconstructed electrochemical sensors. Xu et al. [129] introduced the electroreduction method that includes MIP-based AuNPs-RGO composite as an electrochemical sensor fabricated for methylmercury (CH_3Hg^+) sensing in fish samples at a LOD of 0.12 μg/L. A new molecularly imprinted electrochemical sensor has been reported for the detection of melamine in feed and milk samples, the glassy carbon electrode has been adjusted through gold and polyaniline compounds kept on the glassy carbon electrode surface and have been utilized to build the electrode sensitivity and to amplify the sensor signal for the target of the melamine molecule through the formation of hydrogen bonds at a LOD of 1.39×10^{-6} μmol/L [130]. Another interesting approach is that the MIP-based affinity sensor on an Au electrode surface dependent on capacitive transduction has been developed by electro-polymerization, which can transfer information of benzo(a)pyrene detection in river water; this method introduces great significance to transfer information that has a wide linear range and is more sensitive in MIP sensors [131].

Wang et al. [132] established a novel method to extract olaquindox in real-life samples using a developed MIP electrochemical sensor to detect trace olaquindox in food and feedstuffs using Au nanoparticles by electrodeposition in the imprinting process. However, this method shows that it had good selectivity and high sensitivity toward the target molecule (olaquindox) at a LOD of 2.7 nM.

In electro-polymerization, in order to avoid a portion of the previously described limitations, MIP can be manufactured around a mineral-coated fiber optic sensor on its oxidation surface, wherein the polymer thickness is obtained well around the sensor surface giving an increased refractive index, and the dispersion spectrum light increases with an increase in the target molecule content from MIP sensor, as a result of which the light is dispersed when the template molecules get attached to the cavities in the MIP [133].

Zhang et al. [134] proposed a new method of molecularly imprinted electrochemiluminescence sensor for sensitive monitoring for the direct determination of bisphenol A in seawater and fish samples using magnetic glassy carbon electrode that appears to have high selectivity and sensitivity; the use of MIP has been reported in actual samples for bisphenol determination at a LOD of 2×10^{-4} μg/L. MIP technology was used as one of the most promising advanced technologies for detecting histamine in fish with good selectivity, and MIP was also applied as a signal of food safety [135]. Recently, a surface plasmon resonance was performed to prepare histamine sensing using electropolymerization. The MIP findings showed a strong binding affinity of histamine sensing compared to non-imprinted polymer indicating an excellent specificity in molecularly imprinted at a LOD of 2.0 μg/mL. Manufactured MIPs sensing has a high-quality assurance capability and a high selectivity sensor for the detection of the harvesting freshness of fish and shrimp [136].

MIP-Based Sensors of Solid-Phase Extraction in Food Analysis

MIPs can generally be based on several strategies, such as SPE and solid-phase microextraction (SPME) [137]. Most applications of MIP-based SPE are packed in cartridges and columns and may deal with test lattices simply because the analytes are frequently present at low fixations in food frameworks, thus; pre-treatment testing is typically required to extricate, confine, and concentrate the analytes of enthusiasm from complex grids. Jia et al. [138] proposed a simple strategy for estimating chloramphenicol in fish, pork and chicken using the MIP-based microtiter chemiluminescence method, which is an accurate and sensitive method for determining chloramphenicol in food products at a LOD of 5.0 pg/g, which is outstanding than other techniques.

The analysis is eclectically retained in the extraction column and the interfering particles pass through the column easily. The analytes are absorbed into aqueous samples by SPE-dependent MIP through hydrophobic reactions. Rinse the analyte several times when the column is washed with a suitable solvent to eliminate the interfering compounds with no adverse effect on eclectic reactions between target molecules for MIP in food samples [139-140] and canned food samples [141].

MIP cartridges are used for selective SPE testing of analyte analogs in food samples [142]. Consequently, an unlimited number of analytes are used in the manufacture of imprinted non-covalent polymer to determine analytes, such as fenarimol in food samples at a LOD of 0.03–0.06 μg/mL [143]. Whereas in the study of Zhu et al. [144], ciprofloxacin was used as a template in the MIP-based SPE to extract ciprofloxacin in real samples for the selective extraction of a target molecule at a LOD of 0.11 μg/L.

Since SPE has risen quickly as a well-known method for performing specific extraction techniques, a snappy review of distributed techniques in the course of the most recent decades in food study uncovered that most of the intrigue has been centered on analytes in the real samples. The MIP-based SPE methods have the following specifications: cheap, easy, quick, effective, powerful, and safe [145]. Generally, pre-treatment techniques in a food study incorporate SPE and SPME processes. Among them, molecularly imprinted solid-phase extraction (MISPE) and molecularly imprinted

solid-phase microextraction (MISPME) have been perceived to be the most proficient pre-treatment methods to improve systematic affectability. The SPE depends on allowing the example segments to scatter into the strong sorbents. At that point, the blend is legitimately stuffed into a void SPE cartridge. In the long run, the analytes are eluted after an appropriate washing venture to evacuate the meddling substance. Phenolic compounds used as SPE adsorbents have been extracted from water samples using multi-template MIP; this results in high selectivity, high thermal stability, and uniform spherical morphology [146].

Currently, MIPs used for MISPME are of great importance as they offer the upsides of comfort, time, and particular clean-up of the analytes [60]. Moreover, MISPE and MISPME can be effectively joined into mechanized scientific systems on account of the specific assimilation of MIPs for a specific analyte or collection of analytes [147-148]. A sol-gel precursor has also been performed to prepare MIP-derived fibers for the detection of analytes in food samples [149-150].

Most MISPE-based diagnostic strategies are performed in disconnected mode. The three stages for MISPE are (i) example stacking, (ii) washing, and (iii) elution. Soluble stacking is chosen to allow the rebinding of the analyte to express goals. After the stacking stage, the washing stage is intended to eliminate unwanted debasements from the cartridge, but the dissolvable elution must be updated by its ability to undermine contact between analysts and polymers. SPME has shown itself to be a normal device for its more points of interest, for example, effortlessness of activity, dissolvable less nature, and the accessibility of business strands [151-153]. In other methods, the matrix solid-phase dispersion is set up with olaquindox as the format, methacrylic corrosive as a utilitarian monomer, and EGDMA as the cross-linking specialist. The readied materials have been used as solid-phase materials for matrix solid-phase dispersion to enhance olaquindox and subsequently tested by HPLC. Results showed that it has great recognition, specific capacity, and quick adsorption-desorption elements for olaquindox under streamlined conditions [60]. Moreover, the accomplishment of ideal explicit recognition conditions for target analytes, particularly for different objective analytes, is a noteworthy test. To understand this objective, MIPs incorporated for explicit analytes or gatherings of fundamentally related species, as opposed to ordinary SPE sorbents, have been effectively utilized as SPE sorbents. MIP was evaluated as an adsorbent for SPE of trace cadmium (Cd) and lead (Pb) from seafood digestion; this method involves better sensitivity and was successfully applied for the synthesis of MIP materials for their analysis to detect the analytes in food matrices used in a well-studied matrix due to its good quality at a LOD of 0.15 and 0.5 μg/L, respectively, for cadmium and lead [154].

A selective and sensitive MIP-based fluorescence sensor was developed for molecular recognition and has been successfully applied for sulfadiazine detection in seawater and shrimp samples at a LOD of 0.004 μg/L for seawater and 0.79 μg/kg for shrimp samples. The high-selectivity MIP study indicates good detection of sulfadiazine in real samples with the MIP sensor [155]. Recently, another sensitive MIP method has been established through new technologies that rely on SPE for the identification of patulin in apple-based food products [156].

MIP-Based Sensors of Chromatographic Stationary Phase in Food Analysis

MIP-based sensors have shown promising with their applications in food product analysis. Atomic imprinting as an intriguing procedure applied in the chromatographic division has the unmistakable preferred position of its anticipated elution order; the MIP-based SPE column was used with HPLC to estimate the extraction and enrichment capability of the MIP-SPE column and for the determination of analytes in the real samples [157]. The MIP chromatographic stationary stage needs a few conditions to be filled as MIP-HPLC stationary stages: high selectivity, great hydrodynamic conduct, and high limit. Another significant rule is the homogeneity of the particles used as pressing materials in HPLC. Chromatographic MIP-dependent detachment is frequently used for chiral partition, isomer division and enantio-separation in the food field.

Bai et al. [158] developed an advanced MIP electrochemical sensor to determine olaquindox in fish and feedstuffs with a strong agreement for results based on HPLC at a LOD of 7.5 nmol/L. HPLC and slim electrophoresis have been used for enantiomeric applications. Due to the simplicity of approval and segmentation, several studies have given uncommon attention to irregular targets of chromatographic division. For a particular enantio-separation, the principal issue in chromatography is whether the analytes and their enantiomers are isolated or not. The best-preferred position of the utilization of chiral molecularly imprinted stationary stages is the simple decision of the elution request of the analytes, and solid elution conditions can be dodged, and this method gives high efficiency and good selectivity [159]. Subsequent MIPs can be utilized as enantio-selectors in chromatography. Likewise, solid molecularly imprinted backings as stationary stages have attracted critical enthusiasm for HPLC. Consolidating the benefits of MIP and solid segments, and these solid materials are regularly arranged, the extraction of MIPs can be used as sorbents for the chromatographic determination of the specific analyte in the samples [160].

Zhai et al. [161] suggested simple, sensitive, and developed method of an imprinted polymer to determine flumequine in fish samples at a LOD of 0.32 ng/g. MIPs were prepared using silanized graphene oxide doped in a methanol-water system. MIPs are based on a feasible extraction method and have been successfully applied in an analytical technique for the selective, sensitive, and simultaneous detection of bisphenol A and nonylphenol in different fish samples. MIP-based sensors allow the manufacture of low-cost devices with a LOD of 2.4 and 4.7 ng/L for bisphenol A and nonylphenol, respectively, in food samples [162].

This research indicates the development of a molecularly imprinted stir bar with HPLC for the highly selective extraction and good sensitivity to detect semi-carbazide (target molecule) in fish samples; pre-treatment was convenient and effective with satisfactory results at a LOD of 0.59 ng/mL [163]. Table 2 shows the summary of molecularly imprinted polymers developed for sensors.

■ CHALLENGES OF MIPS IN FOOD ANALYSIS

Ansari described MIPs, despite their challenges, has a great potential for identifying a variety of food analyses due to some unique properties of MIPs [175]. Although the rapidly expanding literature on MIPs used as optical, and chemical sensors in food analysis, the achievements of MIPs have not reached the required level practically.

MIPs have been applied in several fields for various MIP-based sensors, including optical, chemical and electrochemical sensors. The application of MIPs has been faced with strong challenges in the determination of SPE and most advanced applications with pre-treatment methods to determine the analytes in food samples and to evaluate how successfully these analytes could be reduced to make food healthier for human life. Currently, the pre-treatment methods of MIP-based samples are mostly based on several techniques, including SPE, which is known to be physically and chemically stable [152,176]. The study also presents the use of MIPs as sensing receptors for the development of the most widely used electrochemical sensors in polymerization, which are used in the analysis of food and environmental samples soon [177-178].

Many problems hamper the use of conventionally prepared MIP sensors (such as films, membranes, pellets, and beads), including difficulties with the heterogeneous binding, poor selectivity, undesired adsorption, non-specific binding, and removal of the template incompletely to binding sites, so that the templates are chosen based on the chemical substance for MIP-based optical sensors [11].

Although there are some drawbacks: SPE-based sorbents are not reusable [179], the lack of a specific template between template-monomer interactions in the equilibrium process [59], and the destruction of binding sites during the MIP synthesis protocol [180]. Novel methods need to improve the performance of the MIP sensor in food analysis, which is a huge challenge. Consequently, there will be simple and effective methods for preparing MIPs to create convergent and highly selective binding sites that can be applied to MIP sensors.

Table 2. Summary of the sensor applications based on the use of MIPs in food analysis

Template molecule	Monomer/Cross-linker/Initiator/Porogen solvent	Linearity range	Analytical method	Application in real samples	LOD	Recovery (%)	Ref.
Octopamine (OA)	3-Aminopropyl triethoxysilane(APTES)/Tetraethylorthosilicate(TEOS)/ammonia /methanol	0.1–10 mg/L	Optical nanosensor	Fermented samples	0.062 mg/L	80.38–87.17	[164]
β-Lactoglobulin (β-LG)	Polyacrylic acid (PAA), and *N*-Isopropylacrylamide(NIPAM)/*N*,*N*,*N*',*N*-tetramethylethylenediamine (TEMED)/Ammonium persulfate (APS)/Ethanol	0.1–0.8 mg/mL	Fluorescence sensor	Milk products	0.043 mg/mL	86.0–98.4	[165]
Cyromazine	α-methacrylic acid/trimethylolpropane trimethacrylate/phosphate-buffered saline (PBS)	0.5–3.0 µmol/L	Electrochemical sensor	Tomato, cowpea and water samples	0.5 µmol/L	90.14–101.67 and 90.64–101.1 and 91.1–108	[166]
Ciprofloxacin (CIP)	aniline (ANI)/o-phenylenediamine (PDA)/Ethanol aqueous solution	0.001–10.0 µM	Electrochemical sensor	Pharmaceutical samples and biological fluids	0.09 µM	–	[167]
2,4-dinitrophenol (DNP) and 2,4,6-trinitrotoluene (TNT)	Methacrylic acid (MAA)/Ethylene dimethacrylate (EGDMA)/2,2-azobis(2,4-dimethylvaleronitrile) (AIVN)/Ethanol	1.5–8.0 and 1.3–6.5 µM	Electrochemical sensor	Environmental field	0.59 and 0.29 µM	–	[168]
CIP	MAA/2-vinylpyridine (2-VP)/ EDGMA/2,2-azobis(isobutyrontrile) (AIBN)/chloroform/methanol (9:1, v/v)	–	Solid-phase extraction	Pure water	–	105	[169]
Norfloxacin (NOR)	MAA/ EGDMA/ AIBN/acetonitrile	0–200 mg/L	Solid-phase extraction	Seawater and fish samples	0.15 µg/L and 0.10 µg/kg	90.1–102.7	[170]
Profenofos (PFF)	MAA/EDMA/AIBN/acetonitrile	1.0–1000.0 µg/L	Solid-phase extraction	Seawater, food samples, rice and fish	0.33 µg/L	97	[171]
Phenylalanine	Acrylamide (AA)/EGDMA/AIBN/ acetonitrile	1 to 10 mg/mL	Chromatographic Stationary phase	Dietary supplement sample	1 mg/mL	–	[172]
Organophosphorus (7 OP)	MAA/EGDMA/AIBN/chloroform	1–20 ng/mL	Chemiluminescence sensors	Milk	1 to 3 pg/mL	83.6–94.2	[173]
Thiamethoxam and Thiacloprid	2-vinylpyridine EGDMA/4,4′-azobis (4-cyanovaleric acid)/*N*,*N*-dimethylformamide	1–100 µg/L	Chromatographic stationary phase	Light and dark honey	0.045–0.070 µg/kg	96.8–106.5 and 95.3–104.4	[174]

There is an urgent need to create new imprinting techniques in which the development of MIP-based material requires further progress in the imprinting process by applying dummy templates to improve MIP performance. Modern MIPs, such as imprinted sol-gel or hydrogel, must also be developed for the detection of analyte compounds in food products. MIPs can be associated with several other modern techniques, including nanotechnology, which contributes to the expansion of MIPs methodologies in the formation of imprinted sites in order to achieve significant achievements in the uses of MIPs and their applications for food analysis soon. Moreover, MIP-based optical sensors of organic molecules will be developed in the future due to the great practical value of MIPs in food sciences.

■ CONCLUSION

This review summarized the latest applications of MIP sensors that can be applied in food products. Molecular imprinting continues to receive increased attention to achieve its potential in other fields of optical sensors that have been used to determine the analytes in food products. Future trends in this technology will be promising not only because of its selectivity and sensitivity for the detection of analyses, but would also offer some perspectives on the future development of sensitive MIP optical chemical sensors. The current

review comprehensively presented the preparation methods for MIP as an optical, and chemical sensor and their important applications in food analysis. This review has discussed some of the novel MIP-based preparation methods associated with many modern technologies, such as MIP-based optical sensors, MIP-based electrochemical sensors, MIP-based SPE sensors, and MIP-based chromatographic stationary phase sensors in food analysis. MIPs are synthesized by polymerization involving porogen type, template, functional monomer, and cross-linker because the preparation of MIPs is observed to have selectivity, high sensitivity and homogeneous binding site.

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■ AUTHOR CONTRIBUTIONS

Almajed Asaad Abdullah Sfoog, Norlaili Abu Bakar, and Nurulsaidah Abdul Rahim wrote and revised the manuscript. Wan Rusmawati Wan Mahamod, Norhayati Hashim, and Siti Kamilah Che Soh revised the manuscript. All authors agreed to the final version of this manuscript.

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