Sensing Technologies: Advanced Sensor Types and their applications

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Abstract

The term "molecularly imprinted polymers" (MIPs) refers to a class of synthetic receptors that can be created through various polymerization processes involving a target template and functional monomer (s), with functional groups specifically interacting with the template. The nature of the functional groups interacting with the target compounds can be used to tailor the interactions between the functional groups and the template. The basic process underlying the use of MIPs in electrochemical sensors and as solid phase extraction sorbents is the knocking out of the template using appropriate knocking out agents, which creates a recognition cavity that can selectively rebind to the target template.MIPs are great materials to employ for water analysis because they offer special characteristics in terms of stability, selectivity, and resistance to acids and bases in addition to being inexpensive and easy to produce. The current analysis summarizes the various ways that MIPs have been used to identify various types of contaminants in water and wastewater. With regard to food applications (sensors for the detection of food contaminants and the quantification of food nutrients and nutraceuticals, active food packaging applications, and sample preparation: removal, preconcentration, and detection of target analytes), a variety of MIP structures (hydrogels, membranes (nanofibers and films), and particles (core-shell and hollow-shell nanoparticles, microbeads, nanopillars, and nanotubes) were thoroughly reviewed and analyzed.

1. Introduction

1.1 The need for new devices

Analytical chemistry is a science of measurements, which allows us to understand and control our life and the world around. The methods, we use, for these measurements are in a process of constant evolution and replacement with those, which provide better, faster and more accurate results. The objective behind this process is to acquire and apply this information in real time to get a better picture of our environment and hence to control it.

In last few decades, the public consciousness about pollution has increased a great deal because of the endangerment of human and its environment by pollutants of different kinds. The task of modern analytical chemistry is to devise some methods in order to selectively detect potentially dangerous substances in shortest of the time possible, so that the damage to the humans and the environment can be minimized by controlling those substances.

For this purpose, various analytical instruments have been used with which the modern industry has achieved high levels both in detection limit and sample throughput. The spectrum reaches from the detection of a single molecule to a sample throughput of several thousand samples per day. However, these high-tech methods have three serious disadvantages:

- These are relatively expensive both in their purchasing and working cost.
- Well trained personnel are required for their operation.
- These can not be employed with mobility owing to their large size.

There are two different ways to solve these problems. One is to miniaturize already available devices i.e. UV, IR spectrometers or micro-columns for chromatography. The other possibility is to develop sensors.

1.2 Sensor

Sensors have many advantages over classical instruments because they are very cheap, small in size, simple to operate, and can be fabricated easily. Due to their miniaturization, several different sensors can be fabricating on one device to make a multi sensor array and can be used for remote measurements of different analytes unlike the classical instruments. Due to these characteristics, sensors can be used for online monitoring of some specific analytes and appropriate for all types of applications [1-2].

A sensor is a small sized device which transforms a physical change into an analytically useful electrical signals. Sensor actually considered as a part of interface between the world of analytical electrical devices like computers and physical world [3-4]. In this modern era, sensors have gotten substantial importance in research world because it is significant in electronic devices and enormous capability for information processing have been synthesized within the electronics. First decade of 21st century was celebrated as “Sensor decade”.

1.3 Chemical Sensor

A chemical sensor changes chemical information of the concentration of a particular sample part and total composition analysis, into analytically useful electrical signals as shown in figure 1. The chemical information generated due to the chemical reaction of an analyte (template) and also from physical property of any system investigated. It is the practice of continuously monitoring the target analyte's concentration or composition [5]. To better identify and quantify the target analyte in a mixture, a chemical sensor is typically employed; ideally, such a sensor would be both selective and sensitive [6].

A chemical sensor is a tiny device that converts chemical data into an analytically usable signal. This information might originate from a chemical interaction of the analyte or from a physical attribute of the system under study. The literature provides a variety of alternative chemical sensor definitions. The term "chemical sensing" refers to the practice of acquiring real-time data on the amount or content of a specific analyte. To isolate and quantify a single analyte from a mixture, chemists employ chemical sensors, which must be highly selective and sensitive to the target analyte.

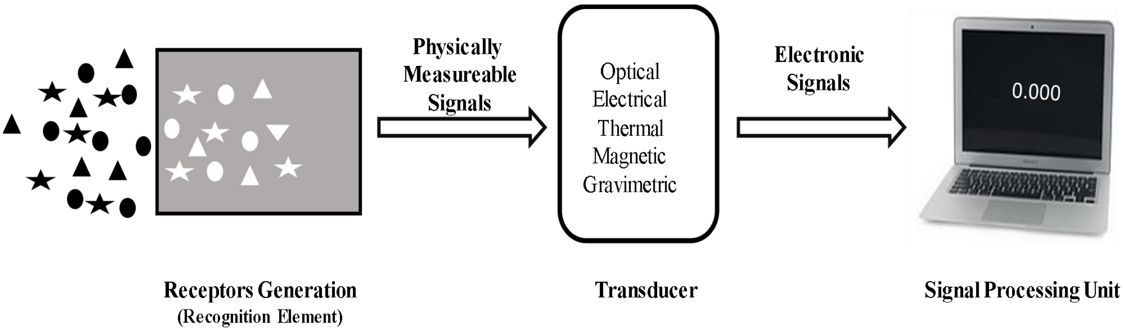

Figure 1: General layout of a chemical sensor

Sensors are normally designed to respond to a specific analyte in a sample; it should show low or little interference to the other analytes, which is called its selectivity. Sensitivity of chemical sensor is its ability to depict the changes in concentration of the target analyte. These Chemical Sensors are small, portable and low cost devices which can selectively recognize certain analytes without separation, enrichment and cleaning steps of sample preparation. Chemical sensors have great advantages over other methods which include simplicity in operation, low cost of working and above all, the possibility of continuous monitoring.

Chemical and bio-sensors are of substantial importance and getting famous in the arena of modern analytical chemistry. Increased interest and demands are essentially due to particularly in diagnostics, environmental investigation, food analysis and for the detection of monitoring and chemical warfare agents [7-8].

A chemical sensor is composed of three major parts,

- Sensitive layer
- Transducer
- Electronics and data storage system

A polymer or antibody (either natural or synthetic) in the recognition layer allows the sensor to specifically monitor the chemical components of the surrounding environment. The analyte of interest interacts with this recognition layer when it is exposed to it. The transducer system picks up variations in one of the layer's physical properties [9] as a result of the interaction, such as mass, optical absorbance, reflectance, polarity, impedance, voltage, or fluorescence behavior. An electronic device called a transducer changes the form of energy being transferred. If we can measure the physical variables connected to the output form of energy, we can make educated guesses about the intake form. The electronic and data storage system picks up on, amplifies, and processes these transmissions [10].

2. Classification of Chemical Sensors

The most common way in which chemical sensors are further classified is by the type of transducer used. The many different types of transducers currently in use for chemical sensing can be broken down into distinct categories based on the underlying signal-generation mechanisms shown in figure 2. These devices can be distinguished as electrical, optical and acoustic wave (or mass-sensitive) transducers and so are the resulting chemical sensors.

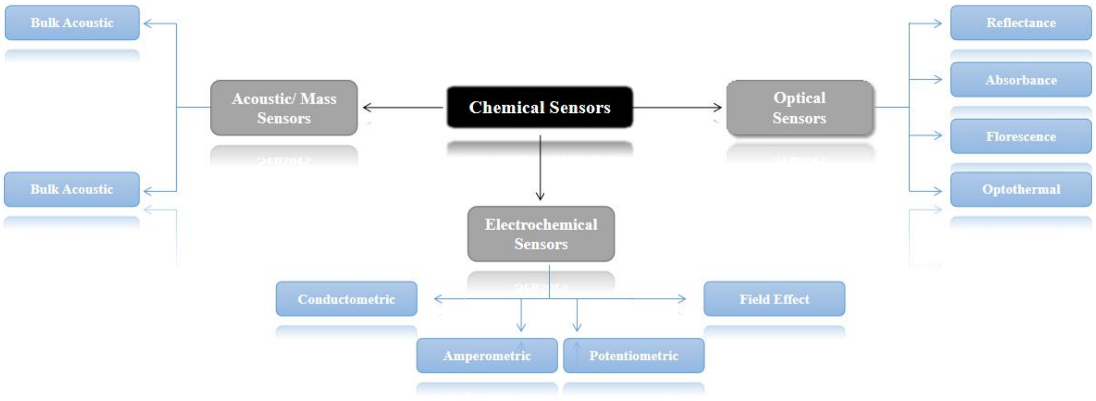

Figure 2: Classification of chemical sensors

2.1 Electrochemical sensors

Electrochemical sensors are better developed and most flexible among all the other chemical sensors. Depending on their operation, they are classified into amperometric sensors, potentiometric sensors, field effect sensors, and conductometric sensors [11]. In all these sensors, at least two electrodes are used where a chemical reaction or charge transfer occur results into the change in potential, conductance, and current values. These changes can be measured when these electrodes are connected to electric current (either ac or dc) except in potentiometric sensors. The electrodes in these sensors are often made of platinum or gold or carbon-coated metals or can be modified to improve their working life [12].

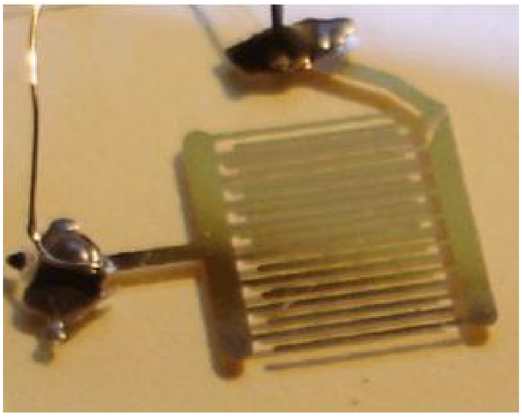

Figure 3: IDE used as transducer for the electrochemical measurements of sensitive layer. The desired structure of IDE (gold) was fabricated via screen printing on the glass substrate. Transducer has 9 fingers with equal gaps and width (300*300) and length of every finger is 7mm (Latif, Najafi et al. 2013).

2.2 Amperometric sensors

Amperometric sensor measure the change in current, the magnitude of which is proportional to amount of target analyte, caused by an electrochemical reaction. The Clark oxygen sensor is an early form of an amperometric chemical sensor, having been designed in 1956. In these sensors an elecroactive redox substance is connected to the transducer and when a target analyte interact with electroactive substance then a redox reaction occurs which can be measured in the form of current. The change in current is directly related to concentration of the specific analyte. So we can say that the information obtained in this group of chemical sensors is in the form of current-concentration relationship. Amperometric sensor has been developed for sensing a variety of analytes like NOx, CO, and lot of other vapros10 due to their low price, high sensitivity and small sizes [13].

2.3 Potentiometric sensors

Potentiometric measurements can only be carried out at zero current. The information obtained from these sensors are in the form of voltage-concentration relationship. In these sensors potential difference (voltage) between working electrode and reference electrode was measured which is equal to the concentration of analyte. Most widely used potentiometric sensor is the Lambda probe11, which is oxygen-sensor to control the combustion in internal combustion engines by maintaining the required oxygen-fuel ratio [14].

2.4 Field-effect sensors

Field-effect transistor (FET) consists of a p-type and two n-type semiconductors substrate (source and drain) and a top metal electrode referred as a gate. Silicon dioxide insulating layer cover the source and drain electrode, it also separates them from gate electrode. A chemically sensitive layer is applied to the gate electrode. By the variation of the voltage at the gate electrode, current flows from source to the drain. The signal from the FETs can be obtained when an analyte is adsorbed or dissociate on the metal surface. FETs can be used for the detection of ammonia, carbon dioxide, hydrogen in air, oxygen in blood and some explosive gases [15-16]

2.5 Conductometric sensors

Another parameter by which information can be obtained from an electrochemical system is conductance. These sensors are also called chemi-resistors because resistance is the reciprocal of conductance. The conductance or resistance of the sensor is altered when an analyte interacts with a selective layer applied to its surface. If the resistances at the electrode-selective layer interface, the selective layer bulk, the selective layer contact, and the selective layer surface are all measured, a sensor signal can be derived. This total resistance is known as equivalent resistance [17].

2.6 Acoustic Wave (or Mass) Sensors

A wide range of analytes can be detected by using chemical sensors that detect a change in mass at the surface of a chemically sensitive layer. Surface acoustic wave (SAW) and bulk acoustic wave (BAW) devices are common types of sensors in this category [14]. These gadgets rely on a mass shift that can be detected through other means, such a shift in frequency, amplitude, mechanical deformation, or phase.16 Absorption and adsorption are two mechanisms that might cause this shift in mass. An applied sensitive layer to the surface of a piezoelectric oscillator forms the basis of a mass sensor. When the analyte comes into contact with the layer, a quantifiable reaction occurs. Because of this, the analyte may be detected in extremely minute concentrations in air, soil, water, and complicated mixtures where many compounds are present; these instruments function like miniature, highly sensitive scales [18].

2.7 Bulk Acoustic Wave (BAW) Devices

The Quartz Crystal Microbalance (QCM), which is commonly referred to in academic literature, serves as a representative example of a Bulk Acoustic Wave (BAW) resonator operating in a thickness shear mode (TSM). The primary piezoelectric material utilised in chemical sensing within the field is comprised of an AT-cut quartz plate featuring round electrodes that have been imprinted on both sides (refer to figure 4). The generation of a thickness shear wave in the substrate is facilitated by the electrical characteristics and crystalline alignment of quartz when an electrical potential is applied between the electrodes.

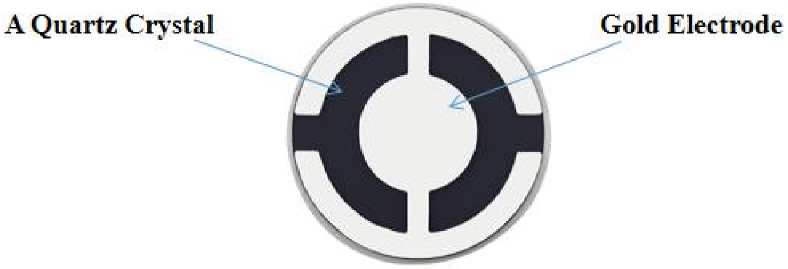

Figure 4: AT-cut quartz crystal with gold electrodes patterned on both sides.

The resonance frequency of the oscillating quartz is determined mainly by its thickness. QCMs are readily available up to 50MHz, but the plates become too thin for mechanical stability at higher frequencies. Quartz crystals with a frequency range of 5-10 MHz are frequently employed in sensor applications.

One way to impart selectivity upon a sensor is to coat the crystals in a chemical recognition layer. The selective film may really be the metallic electrode itself, when placed to the crystal. A circuit is set up to allow the coated QCM to oscillate at its fundamental frequency. The crystal's thickness, structure, shape, and mass all affect the frequency of its basic mode of vibration. Resonance frequency changes, which may be measured with great precision, as a result of analyte interactions with the surface of the sensitive layer [19].

2.8 Surface Acoustic Wave (SAW) Devices

SAW transducers are produced from ST-cut quartz plates on which a pair of inter-digital transducers (IDTs; the comb electrodes) is generated as shown in Figure 5. When an alternating current is applied, the piezoelectric material develops an electric field. A surface acoustic Rayleigh wave, formed when particles within the crystal dissipate, propagates from one of the comb electrodes, across the surface of the sensor, and into a second IDT.

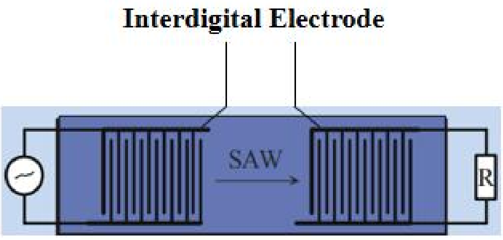

Figure 5: A surface Acoustic Wave Device

In response, a voltage fluctuation occurs, which can be measured electrically. SAWs have a far higher sensitivity to mass loading than QCMs since they can be operated in frequency ranges up to 2.5GHz (analytical applications have been reported up to 1.0GHz). However, conventional SAWs are inapplicable non the liquid phase due to the extreme surface-wave damping that occurs there [20].

3. Molecular Imprinting Technology

3.1 General Principal

Synthetic polymers can be imprinted with specific molecular information by co-polymerizing functional monomers and cross-linkers in the presence of the target analyte (the imprint molecule). The polymerization process begins with the formation of a complex between the template molecule and the functional molecule [21]. After polymerization highly cross-linked polymer is formed which held their functional groups in position. Template removal from polymer leaving behind empty spaces which is similar in shape and size to the template molecule. Thus molecular memory with very high specificity is introduced in to the highly crossed-linked polymer. Now this polymer has ability to bind with target molecule anions [21-23]. The synthesized MIP may be polymeric or oligomer or 2D surface assemblies. MIPs possess a diverse array of advantages as compared to their native biomolecular counterparts. This technology has many promising features and many benefits including easy synthesis, similar structure based recognition cavities, a highly stabilized and mechanical backbone of polymer matrix and large number of applications to different molecules, metalloids and metal ions.

Due to three distinguishing characteristics, MIPs have been the subject of extensive study:

i. Their selectivity and optimum affinity, is analogous to the natural receptors [24].
ii. Their exceptional stability, which is higher than that of natural biomolecules.
iii. Their simplest preparation and easy to use to different applications.

Small chemicals like medicines and big molecules like proteins and cells have both been effectively imprinted. Biological compounds with a molecular weight between 200 and 1200Da gave the best imprinting outcomes. The resulting molecular imprinted polymers are cost-effective, resilient, and exhibit high affinity and selectivity, making them ideal for use in industry [25]. MIPs are a possible alternative to enzymes, antibodies, and natural receptors in sensor technology due to their excellent specificity and stability [26].

Table 1.1: Sensor and MIPs natural biomolecule comparison

Molecular imprinted polymer (MIPs)	**Natural biomolecules**
Imprinted polymers are more stable at low pressure, pH and temperature i.e. <180◦C.	Less stable.
Easy to prepare, easy to handle, and inexpensive.	Enzymes and receptors are higher in price.
Imprinted polymers can work efficiently in organic solvents.	Natural biomolecules show commonly poor performance on non-aqueous medium.
Generally, MIPs of any chemical compound could be synthesized.	Enzymes and natural biomolecules may be used for limited number of analytes.
Imprinted polymers are highly compatible for micromachining	Enzymes and natural receptors have poor compatibility with micromachining technology and

technology.	miniaturization.

In molecular imprinting of synthetic polymers, the target analyte (the imprint molecule) serves as a molecular template and is co-polymerized with functional and cross-linking monomers. The functional monomers initially form a complex with the imprint molecule; after polymerization, the strongly cross-linked polymeric structure locks in place the functional groups of the functional monomers as dhown in figure 6. After the imprint molecule is removed, binding sites that are a good fit for the analyte are exposed. This imparts molecular memory onto the polymer, allowing it to rebind the analyte with extreme specificity [27-28]. The technique of molecular imprinting has different advantages such as it is used in purification, isolation, chiral separation, catalysis and in biosensor.

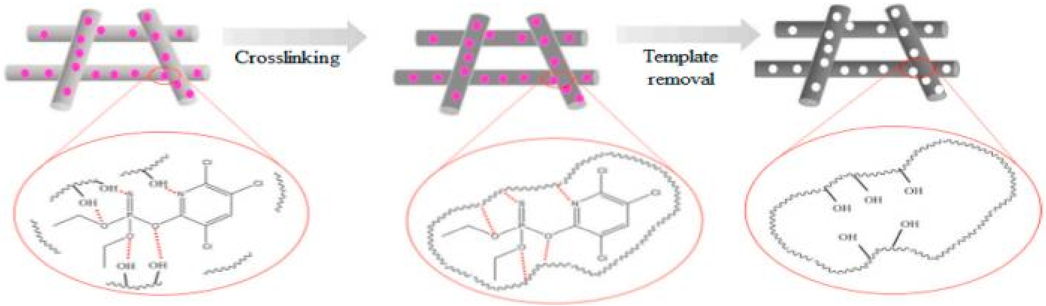

Figure 6: Schematic illustration of the preparation of molecularly imprinted polymers [29].

Imprinting technique can be divided into two main types i.e. bulk imprinting and surface imprinting.

3.2 Bulk Imprinting

The technique of bulk imprinting is successful for low molecular weight compounds. For imprinting of macromolecule, MIPs are the simplest method. For the whole protein 3D binding sites are formed in this method [30]. The target molecule is wholly imprinted in the molecular imprinted polymer and it must be removed wholly from the polymer matrix after polymerization as shown in figure 7. The next step is the formation of small particles from the bulk polymers. The polymer is mechanically crushed to form small particles. In this way molecular imprinted polymer is formed having 3D interaction sites specific to the template molecule. Usually bulk imprinting is chosen to imprint small molecules. The advantage of this method is that the imprint molecules are easily adsorbed and release and this process is reversible and faster due to which imprinted support can be utilized for several rounds of analyses [29].

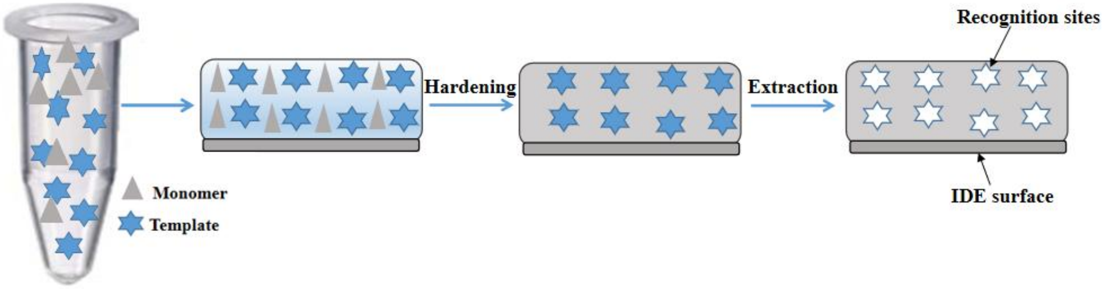

Figure 7: Schematic presentation of bulk imprinting

Bulk imprinting is used for various organic compounds such as volatile organic compounds, polycyclic aromatic compounds, environmental contaminants, pharmaceutically active compounds etc. The desired template is added to functional monomer during polymerization [31-32]. The binding sites formed by molecular imprinting are not limited to the surface of synthesized receptors; rather, they are dispersed throughout the whole polymer matrix, hence the phrase "bulk imprinting." Since a significant number of cavities have been incorporated into the newly designed recognition layer, it is possible that the polymer matrix beneath it can be tested by diffusion to determine their usefulness. In the case of small analyte molecules, this guarantees the required sensitivity, and the presence of interaction sites may aid to boost the sensor response (depending on layer height) [33]. Figure 6 depicts a schematic of a bulk imprinting procedure.

3.3 Surface Imprinting

In surface imprinting, surface of the solid substrate is required for the imprinting of MIP is representing in figure 8. In this technique, obtained MIPs have abundant cavities and sites [34-35]. In this technique binding sites are present only at the surface instead of whole matrix. Surface imprinting is used for macromolecule [36-37].

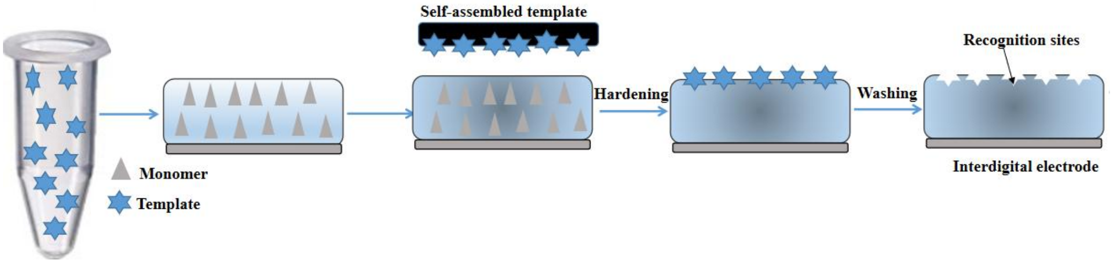

Figure 8: Schematic presentation of surface imprinting

Since binding sites are exclusively surface-level, this approach has the major drawback of having fewer binding sites. Surface imprinting methodology is mostly used for large template including microorganisms, proteins and cells, which is the major drawback of this method [38].

4. Basic Feature of Molecularly Imprinted polymers (MIPs)

4.1 Covalent Approach

Covalent imprinting was the first approach for realizing the affinity of molecule in organic polymer networks. In this imprint technique, template is covalently bonded with polymerizable groups in bond formation and the process of rebinding [38]. After polymerization, the target molecule is no longer present in the matrix, but binding sites that are structurally identical to the target molecule remain, ready to form a new covalent bond. One benefit of this approach is that only the functional groups are linked to the template sites [39]. However, difficulties in cleaving the template and a limited set of imprintable compounds (alcohols (diols), aldehydes, ketones, amines, and carboxylic acids) prompted the development of other imprinting methods [40].

4.2 Non-Covalent Approach

The development of molecularly imprinted binding sites has shifted towards a non-covalent approach. This technique involves the non-covalent interaction of a functional monomer with a template molecule prior to polymerization, followed by polymerization using free radicals and a cross-linking monomer. Hydrogen bonding, Vander Waals forces, and - interactions are the non-covalent interactions. This strategy likewise employs non-covalent contact to accomplish the rebinding of the template. This method has very simple and cheap method of MIP preparation sites [41]. For this approach to work, the non-covalent contact between the functional monomer and the template must be stabilized, which will lead to the binding of functional groups into the sites of the template. Binding sites with only a few convergent positioned functional groups can bind a substrate with very high binding energies, as demonstrated by biological recognition elements. The imprinted materials can be prepared quickly in any modern laboratory. Some of the MIPs prepared show high selectivity and binding affinities in the order of those showed by antibodies for their antigens [42]. The most widely utilised functional monomer, methyl methacrylic acid, bonds to the template via hydrogen bonding and other non-covalent interactions.

4.3 Semi-covalent Imprinting

Covalent and non-covalent approaches are combined to form the semi-covalent approach. This technique combines non-covalent interaction during rebinding with covalent interaction by employing a template containing polymerizable groups. The benefit of this approach is that additional kinetic limits do not apply and the binding sites have a more restricted distribution [43].

This technique was used to eliminate the shortcomings of non-covalent interactions [44]. The semi-covalent method bonds template to monomer covalently and hydrolyzes it. Later, the imprinted sites can interact non-covalently with the template.

The typical advantages of semi-covalent interactions are

I. Monomers with functional groups connect to the template molecule.
II. Functional groups are not randomly distributed due to stoichiometric ratios.
III. Template rebinding is easy with binding sites.

Due to the scarcity of suitable monomers, however, the semi-covalent strategy has limited practical uses. Metal ion coordination interaction is a new method proposed in MIP synthesis, particularly for bio-molecules. Any MIP synthesis technique will need a functional monomer, a template, a cross-linker, an initiator, and a porogen solvent [45].

5.Chemicals and Constituents Required for MIPs

5.1 Template

Template and functional monomer are backbone of the molecularly imprinting process. The interaction between monomer and template determines the success of any MIPs. Ideally strong interactions enhance the recognition mechanism of synthetic receptors [46]. Appropriate selected species as template have the capability to produce maximum recognition sites [47]. Analyte and functional monomer must have non-covalent approach. For the selection of template, stability, capability of hydrogen bonding establishment and absence of polymerized groups prevent the newly synthesized free radicals [48]. In this proposed research work, the template molecules will be the mono and disaccharides.

5.2 Functional Monomer

Functional monomer imparts vital role in cavities generation for the analyte of interest [49]. The interactions between monomer and template may be covalent or noncovalent. Most commonly used monomer are methacrylic acid (MAA), acrylic acid (AA), 2- or 4-vinylpyridine, sulphonic acids (2-acrylamido-2-methylpropane sulphonic acid), carboxylic acids (acrylic acid, methacrylic acid, vinylbenzoic acid), hetero aromatic bases (vinylpyridine, vinylimidazole) [50]. Methacrylic acid, most frequently used due to covalent and noncovalent interaction with template. Monomer to template ratio is significant because analyte decides the exact number of monomer molecule to be attached. Template functionality and monomer molecule matching functionality are critical for enhancing complex formation and the imprinting effect [51].

5.3 Cross-linker

The chemical nature and amount of cross-linker used in imprint polymer synthesis play an important role in selectivity of a sensor. The responsibility of cross-linker is to maintain morphology of polymer matrix (gel-type, macrosporous or a microgel powder), stabilization of molecular binding sites and imparts the mechanical stability to the polymer imprinting binding sites in the polymer matrix [52]. Ethylene glycol dimethacrylate (EGDMA), trimethylolpropane trimethacrylate (TRIM) and divinlybenzene (DVB) are most commonly used cross-linkers in MIPs. For PCPs analysis, EGDMA is a frequently used crosslinker. For triclosan analysis, DVB and TRIM have been used to prepare imprinted nanobeads. For polymerization process, high cross-linker to functional monomer ratio is preferred for the material with suitable mechanical strength and access permanently porous materials to maintain the stability of recognition sites. After the removal of template, it is the responsibility of high cross-linker to maintain three dimensional structure (chemical functionality and shape to that of template cavities). Most commonly, polymers with cross-link ratios above 80% will be employed [53].

5.4 Porogenic Solvents

For the synthesis of macrosporous polymer, porogenic solvents have significant role for porous structure of MIPs. Strength of non-covalent interactions is determined by the nature and amount of solvents. It also helps to influence the morphology of polymer, which is directly related to the efficiency of MIPs. Following points are important to consider during the selection of solvent, firstly, all the polymer ingredients such as template, free radical initiator, functional monomer and cross-linker should be soluble in the selected porogenic solvents. Secondly, porogenic solvents have property to produce large sized pores for good flow through properties of resulting polymer. Thirdly, solvents should be of relatively lower polarity to lower the interference during complex synthesis between the monomer and the imprint molecule [54].

5.5 Initiators

Free radical polymerization can use a variety of free radical sources, each of which has its own unique chemical characteristics. Usually, initiators are used at a relatively low level with the concentration of functional monomer. The decomposition rate of initiator can be controlled and triggered by different methods such as heat, light and electrochemical and chemical ways, based on its chemical nature. Most commonly used initiator is azobisisobutyronitrile (AIBN). Initiation of the development of a wide variety of vinyl monomers is possible when AIBN initiator has been appropriately degraded either photolysis (UV) or thermolysis (heat) [55].

5.6 Polymerization Conditions

Molecular imprinted polymer (MIPs) of higher selectivity are made at low temperature as compared to polymerization at increased temperature. Usually 60 °C is referred as appropriate polymerization temperature. Elevated temperature reduces MIP stability, which reduces monolithic stationary phase reproducibility and increases column pressure decreases. To create repeatable polymerization, a low temperature and extended reaction time will be chosen [56].

6. Sensor Fabrication MIP Immobilization on Transducer Surfaces

MIPs will be immobilized on transducers to make chemical sensors. MIP-based recognition elements will be deposited or grafted onto transducer platforms as thin films or layers. The film or layer coated onto the transducer is important for useful response of a sensor (Rebeloet al., 2021). Electro-synthesis of MIPs at the transducer surface is the best immobilization method, in-situ polymerization. In-situ polymerization automates mass manufacturing by incorporating the immobilization phase, and it will be used on suitable template molecules.

Immobilization by surface coating is another important method of coating and immobilizing molecular imprinting polymer (MIPs) onto the selected transducer (QCM, IDEs) surface. Spin and spray coating will help in the synthesis of thin film of MIPs (if prepared in a suitable dissolved solvent). MIPs deposition can be performed manually at different times to prepare sensors.

Another important method is entrapment of MIP particles into gels or membranes will also be used for electrochemical or mass sensitive transducers for immobilization. Then these polymerized material will be deposited onto the surface of transducer

7. Types of transduction

MIP-based electrochemical sensors have been synthesized using Conductometry, impedance spectroscopy, chemical field effect, Amperometry, potentiometry, and voltammetry. Some approaches require vigorous and continual shaking to achieve a stationary regime, therefore the sensitive layer should be robust to prevent bonding site distortion during operating conditions [57].

7.1 Conductometry

Conductometry uses an electric field to produce current by moving ions between electrodes in an electrolyte solution. Based on MIP membrane development, a molecular imprinted polymer sensor was synthesized. Early studies supported polymerization with glass filters [58]. Conductivity is an additive, so this type of conduction is least sensitive to the electrochemical techniques.

7.2 Capacitance/Impedance

System impedance or capacitance variations measure interfacial process. A thin, pore-free, dielectric coating primarily deposited on the gold electrode is needed. The ultimate objective is a thin, pore-free dielectric film on gold substrates [59]. Electro-polymerized polymers like phenol (non-conductive) are a good alternative since film growth is straightforward to control, but pores must be filled with alkyl thiols on a gold substrate [60].

7.3 Amperometry

Amperometry measures current linearly with electroactive species concentration at constant voltage. Electrochemical displacement can include non-electro-active species [61]. Template molecule diffusion into the working electrode and reaction product diffusion outward are needed to establish a current; otherwise, the surface is passivated. Thus, the recognition layer's porosity (regardless of the recognition element) determines the pores needed for analyte adsorption and electron transfer reactions at a bare transducer surface. Selective template extraction and electron exchange with the electrode surface follow measurement.

8. Analytical characteristics of chemical sensors

8.1 Limit of Detection (LODs)

It has been reported that acrylic MIPs is very difficult to obtain low LODs [62]. This improved method of synthesis and imprinted binding site concentration. Electrochemical sensors should use the most sensitive transduction type, such as amperometric or DPV. In any case, the LOD depends on transduction type and binding site basics.

8.2 Selectivity

Mostly selectivity is used with the receptors by batch binding assays, thus simply ignoring the essential interventions but not with sensors [63]. An important feature of voltammetry is to improve the detection by optimizing the binding media and measurement medium [64]. Electrochemical transduction also uses the similar medium for the detection and binding. Electrochemical potential also identifies structurally related interferents [65].

8.3 Reproducibility and reversibility

Polymer-based molecular imprint sensors that can be reactivated through repeated washing and binding would be ideal. Full template extraction occasionally necessitates lengthy washing times, say 12 hours. Additionally, molecular imprinted polymer based sensors can be integrated into devices that consistently and cheaply replace a stale and worn out components [66]. Another method for creating highly repeatable, simple-to-prepare sensors is electro-polymerization.

8.4 Response time and Stability

Generally, response time of molecular imprinted polymer based sensors have been longer than thin films. The smallest response time was obtained from grafting polymerization [59]. Sensors fabricated using acrylic or vinyl as monomers have demonstrated remarkable durability during extended periods of storage, often exceeding 6 months. This outcome aligns with expectations for a polymer with a high degree of cross-linking.

The oscillator surface's mass transfer and/or film deposit govern the time constant. If diffusion controls transport, the time constant is usually (d2 /D), where (d) is layer thickness and (D) is apparent diffusion coefficient. Online monitoring of processes, such as the monitoring of a chemical reaction, benefits greatly from rapid responses. Operating at greater temperatures results in a quicker response time. Other parameters that affect the response time include type of layer material (e.g.; polymer or metal oxide), layer thickness, type of the analyte, interaction between the analyte and the surface etc.

8.5 Drift

Drift is a slow, non-random change of the sensor response of a piezoelectric sensing device under unaltered experimental conditions. In the simplest case, drift is determined by fitting a linear function to the data set collected in a given period of time. The slope of the line of the measured variable i.e.; drift is the variation in frequency with respect to time. If the layer's adhesion to the quartz crystal or electrodes is poor, or if there are electrical issues, this can be detected over time [67].

8.6 Hysteresis

Hysteresis can be defined as the irreversibility of the sensor signal i.e.; change in frequency due to change in the ambient conditions e.g.; concentration, is not reversible when the ambient conditions are changed to original. Usually, hysteresis has kinetic origins. Therefore, reversible values of the measured signals are expected, if sufficient time is allowed for the system to return to its initial condition [68].

8.7 Level of Noise

A sensor's desirable characteristics are high sensitivity and low intrinsic noise. This quality allows the sensor to respond to extremely low concentrations and to detect extremely subtle shifts in analyte concentration. While working in the gas phase, this becomes even more significant as the concentrations of the analytes are very small [69].

9. Applications of molecularly imprinted polymer based chemical sensing

9.1 General process for the synthesis of MIPs based chemical sensors

Chemical sensors known as molecularly imprinted polymer (MIP) sensors use synthetic materials with molecular imprints to detect and measure particular target molecules with great selectivity and sensitivity. MIPs have been developed to replicate the recognition capabilities of organic receptors, such antibodies or enzymes, but because they are wholly synthetic, they are more reliable and affordable [70]. The target analyte serves as a template molecule for the polymerization of functional monomers to form MIPs. Following polymerization, the template is eliminated, leaving voids in the MIP with forms that are compatible with the target molecule. The analyte binds specifically to these holes, enabling the MIP to distinguish it from a complicated mixture of other molecules and collect it [71]. Several stages are needed to synthesize MIP sensors. The procedure varies by application and analyte. These crucial features:

Monomers Selection: Select functional monomers that bind to the target analyte. Monomers ought to comply with polymerization conditions.

Template Molecule Selection: Determine the specific analyte that the molecularly imprinted polymer (MIP) sensor is intended to detect. The template molecule needs to possess same dimensions, configuration, and chemical attributes as the target analyte.

Polymerization: Use an appropriate polymerization process to encapsulate the template molecule in a network of functional monomers. Bulk polymerization, precipitation polymerization, and surface imprinting are just a few of the more common techniques. Polymerization can be started with the use of heat, light, or a chemical catalyst.

Removal of Template: Remove the template molecule from the MIP after polymerization to form cracks that recognize the target analyte. Solvent extraction or thermal degradation remove templates.

Transduction of Sensors: Use the MIP to create a sensor substrate that can measure molecular identification signals. The application determines the transduction method: optical, electrochemical, piezoelectric, or others.

Characterization of Sensors: Characterize the MIP sensor thoroughly to evaluate its performance. Assess sensitivity, selectivity, dynamic range, and response time. Experimentally calibrate the sensor with specified target analyte amounts.

Optimization: Based on the outcomes of the characterization, repeat and improve the MIP synthesis and sensor design. To enhance sensor performance, change the functional monomer type and ratio, template concentration, polymerization conditions, and sensor set-up as necessary.

Testing for Appropriate Applications: To make sure the MIP sensor is functional and appropriate for the intended use, test it in real-world scenarios. Utilize intricate sample matrices that may contain interfering compounds to test the sensor's performance.

Long Term Durability: Examine the MIP sensor's long-term stability and shelf life to guarantee its dependability throughout time.

10. Molecularly imprinted polymers for water and wastewater

Water is an essential resource that supports the existence of all organisms on Earth, ranging from the most basic to the most intricate. It is considered a valuable commodity and plays a crucial role in sustaining life. Water is a vital resource utilised by all organisms, encompassing both plants and animals. It plays a crucial role in several biological processes, commencing with the photosynthetic activities of plants and extending to the reliance of humans and animals on these plants for sustenance. Therefore, it can be said that this particular element has significant importance and plays a crucial role within various biological interactions, since it is utilised by all organisms for distinct and necessary functions [72].

With population growth, technical and industrial improvements produce water pollutants such heavy metals, dyes, pesticides, EDCs, and pharmaceuticals. [73]. Therefore, the treatment of water pollution, including contaminants from wastewater, freshwater, human and environmental pollution, is the biggest challenge to the environment globally. Physical adsorption, oxidation, sedimentation, coagulation, and bioremediation are just some of the conventional methods used, although they are rarely effective enough.

Many of the stated traditional methods are neither efficient or sensitive enough for the removal and detection of contaminants since most pollutants are concentrated in parts per billion (micrograms per liter) or parts per trillion (nanograms per liter). As a result, there is a requirement for improved methods of adsorption and pre-concentration. Because of their versatile, target-specific manufacturing, molecularly imprinted polymers (MIPs) are a promising material class with applications in environmental science. It has been stated that MIPs have excellent features, such as being simple to prepare, environmentally friendly, cost-effective, and sensitive to several kinds of pollutants that are likely to be present in wastewater [72, 74].

10.1 Heavy Metals

Heavy metals are among the most harmful chemical contaminants that can leach into water supplies as a result of human activity, and they are just one of many causes of water pollution. Because of their low cost, stability, and, in many circumstances, acceptable selectivity compared to more conventional extraction methods, molecularly imprinted polymers are a promising alternative to more established chemical and physical processes [75]. Two recent reviews examined molecularly imprinted polymer methods for harmful heavy metal detection [72, 76].

10.1.1 Mercury

Mercury is mostly produced by Earth's geothermal processes. Mercury toxicity can be assessed by protein precipitation, enzyme inhibition, and corrosive effect. The most toxic form of mercury, methylmercury, has a high affinity for sulfuryl ligands in amino acids and may affect protein structure and function. Mercury contamination is one of the most researched environmental heavy metal contaminants due to its persistent buildup in aquatic environments (figure 9) [75]. IIPs were evaluated for selective detection and extraction Hg(II) from water samples.

A novel Hg(II) electrochemical sensor, based on a reduced graphene oxide (RGO) surface imprinted modified glassy carbon electrode (GCE), uses methacrylic acid (MAA) as the functional monomer, ethylene glycol dimethacrylate (EGDMA) as the cross-linker,2,20–((9E,10E)–1,4–dihydroxyanthracene–9,10-diylidene)bis(hydrazine–1 carbothioaide) (DDBHCT) as the chelating agent, and APS as initiator Accumulation of Hg(II) ions on a modified glassy carbon electrode allowed for electrochemical Hg content tracking from 0.07 to 80 µg L^{-1}. The LOD was 0.02 µg L^{-1} (S/N = 3), below the WHO recommendation threshold. [77].

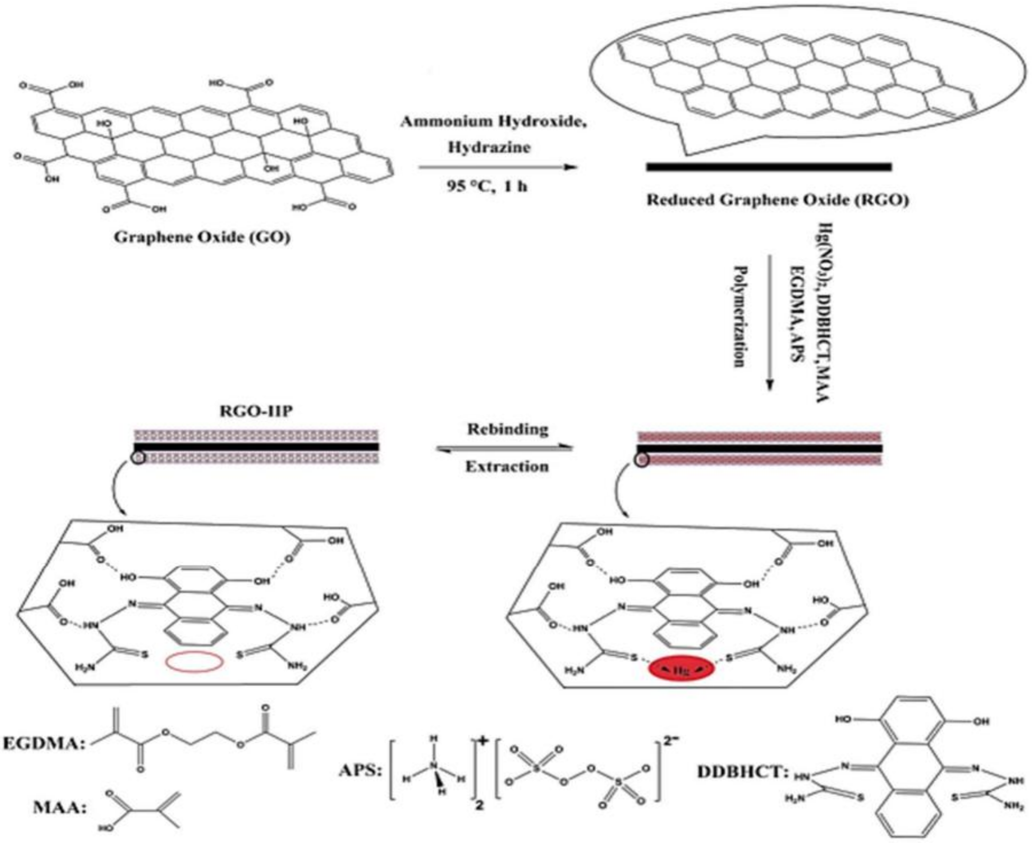

Figure 9: Preparation of reduced graphene oxide-ion-imprinted polymer modified glassy carbon electrode [77].

Another method used sulfur-containing carboxymethyl ion-imprinted polymers. Electro-polymerizing pyrrole and drop coating with S-IIPs modified a glassy carbon electrode. The ion-imprinted polymer electrochemical sensor detected Hg(II) in water. A detection limit of 0.1 mg L^{-1} was determined within a concentration range of 20-800 mg L^{-1}, lower than the WHO criterion of 0.001 mgL^{-1}. The results in real sample compared to inductively coupled plasma-optical emission spectrometry show its potential for Hg determination [78].

10.1.2 Lead

Lead is the most poisonous heavy metal to people and animals. It is poisonous to living things and can be spread from soil to plants to animals and humans through the food chain, causing serious pollution. Lead accumulates by adsorption, complexation, or chemical interactions with inorganic and organic substances due to its non-biodegradability [79]. Paints, wires, pipes, and insecticides all contain lead, and the largest human input comes from fossil fuel combustion engines [73, 75]. An 8-

hydroxyquinoline-grafted gelatin and chitosan solid phase extraction technique with genipin as the cross linker and lead ions as the monomer. With a high adsorption capacity of 235.7 mg g^{-1} and a detection limit of 0.2 ng mL^{-1}, three-dimensional IIPs were developed. Spectrophotometric detection at 560 nm was used to follow the adsorption/desorption interactions throughout a linear range of 1-100 ng mL^{-1} [80].

10.1.3 Copper

Copper is detrimental to the environment and creatures in excessive amounts. Copper pollution is known to harm urban ecosystems and aquatic species' chemoreception and chemosensory [81].

The WHO recommends a safe dose of 2 mg L^{-1} to prevent health risks such as hypochromic anemia, osteoporosis, liver and kidney failures, and immune-toxicity [55, 60]. Graphite oxide/imprinted polymer composite was used to construct a copper (II)-selective potentiometric sensor. 5-methyl-2-thiozylmethacrylamide was a functional monomer and Cu IIP/multi-walled carbon nanotubes/graphene oxide was a carbon paste electrode modification. A linear response range of 1×10^{-6}, 1×10^{-1} M was observed, with a detection limit of 4×10^{-7} M. Inductively coupled plasma–mass spectrometry (ICP-MS) and electrode analyses of tap, river, and dam water samples were comparable [82]. Chitosan, gelatin, and 8-hydroxyquinoline (HQ) monomers were utilised to make green imprinted Cu(II) IIPs. Figure 4 shows multi-point interactions from complementary functional monomers that the ideal G:HQ mass ratio was 2:1 and the optimal G-HQ:C: Cu(II) ratio was 150:150:5. At ambient temperature and pH 6, G-HQ-C IIPs had a maximum adsorption capacity of 111.81 mg g^{-1} for Cu(II). G-HQ-CIIPS selectively removes Cu(II) from water samples. [83].

10.1.4 Chromium

Leather tanning, electroplating, metal finishing, chromate preparation, dye manufacture, and aluminum built-up employ chromium. Hexavalent Cr(VI) and trivalent Cr(III) are environmental chromium oxidations. More hazardous than Cr(III), Cr(VI) is carcinogenic and fatal to human health. A photoelectrochemical (PEC) sensor based on nanostructured hybrid formate anion inserted in graphitic carbon nitride (F-g-C_3N_4) was intelligently coupled with a Cr(VI) ion-imprinted polymer (IIP) (IIP@F-g-C_3N_4) to measure Cr(VI) in water samples (figure 10). With a correlation coefficient of 0.9998, and a detection limit of 0.006 g L1 (6 ppt), we found a linear response to Cr(VI) concentrations from 0.01 to 100 g L1. Electro-catalytic results (5.2 ppb) and FAAS (0.8 ppb) are both more sensitive than this. In order to identify Cr(VI), Cr(III), and total chromium in aqueous solution, Cr(III) was oxidized to Cr(VI) and then measured [84].

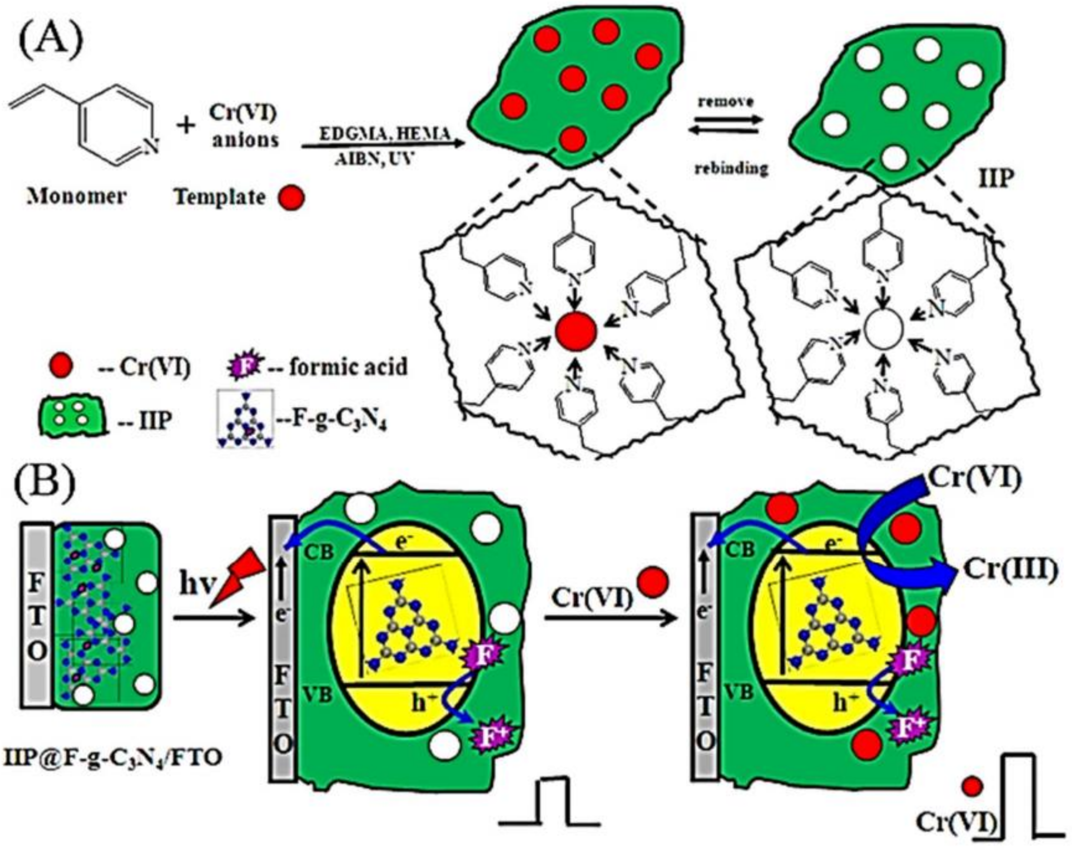

Figure 10: Schematic illustration of the formation process of (A) Figure 5. Schematic illustration of the formation process of (A) the IIP and (rinci ple of PEC determination of B) the principle of PEC determina VI usin IIP@ ra hitic carbon nitride F- -C3N4 tion of Cr (VI) using IIP@ graphitic carbon nitride (F-g-C) 3 53 . Re roduced under ermission from Elsevi N4 [77].

A fluorescence sensor was developed to detect Cr(VI) from 20 g L^{-1} to 1 mg L^{-1}, with a threshold for detection of 5.48 g L^{-1}, by using ion-imprinted polymer (IIP) as the recognition element and Mn-doped ZnS QDs as the fluorophores. In comparison to Cr(III), Cl, SO$_4^{2-}$, PO$_4^{3-}$, and MoO$_4^{2-}$, the QDs-IIP sensor demonstrated high selectivity for Cr(VI). [85].

10.1.5 Nickel

Nickel, one of the most common metals, has numerous applications including electroplating, battery production, and electronic components [65]. Nickel is necessary for hormonal action and for lipid metabolism. However, excessive nickel exposure can cause genotoxicity, hepatotoxicity, and carcinogenicity. Magnetically imprinted polymer was prepared by precipitated polymers of amine-functionalized silica-capped Fe_2O_3 nanoparticles, 4-vinyl pyridine as complex agent, and meth-acrylic acid as well-designed monomer. ICP-OES was utilised to identify the polymer during all adsorption studies, and pseudo-second-order kinetics and Langmuir isotherm showed that it was very sensitive to Ni^{2+}. The adsorption capability was 16.72 mg g^{-1}, with a detection limit of 0.58, quantification limit of 1.93, and percent relative standard deviation of 3.4% [67].

10.1.6 Manganese

Manganese is used in ferrous metallurgy, electrochemical, chemical, food, and pharmaceutical sectors. Although important for many human processes, the maximum permissible amount in water should not exceed 0.1 mg L^{-1} due to its harmful impact on the neurological system [86].

For manganese ion detection and extraction, MWCNT-IIP was developed. Vinyl functionalized MWCNTs were free radical polymerized with methacrylic acid to create the ion imprinted layer. The polymer was immobilized in nafion to create a modified platinum electrode. Under optimized conditions, differential pulse voltammetry could sense Mn(II) ions from 1 to 5 ppm with a LOD of 0.0138 mM [87].

10.1.7 Cadmium

Cadmium is a poisonous metal that can cause lung cancer, renal, bone, and hemorrhoids. The WHO recommends a drinking water level of 10 µg L^{-1} [65]. For selective cadmium interaction, a poly (N-isopropyl acrylamide) grafted and modified g-C_3N_4 magnetic Fe_3O_4 ion-imprinted polymer (IIP) was developed. The adsorption of Cd^{+2} is 2.86 times higher than that of Cu^{+2}, Pb^{+2}, and Zn^{+2}. Pseudo-second-order and Langmuir isotherm models capture the experimental data well [88].

10.1.8 Dyes

Industries like textiles, paper, plastics, leather, and paints use synthetic dyes. Even a tiny amount of dye (10-20 mgL^{-1}) negatively impacts water transparency, gas solubility, and photosynthesis in aquatic environments by blocking light and oxygen penetration. For water sample analysis polluted with different colors, molecularly imprinted polymers seem promising.

Disazo dye Congo Red (CR) metabolizes to benzidine, a carcinogen. Sulphonic acid groups increase hydrophilicity, stability, and low biodegradability. A single-step expansion polymerization was employed to create CR-MIP microspheres in water using meth-acrylic acid as the active monomer after dispersion polymerization for polystyrene microspheres with CR as the template molecule. The stated absorption capacity of various adsorbents is comparable to 27.0 mg g^{-1}. The MIP-based method yields polymers that are more durable and recyclable. The polymers' average CR removal rates from river and wastewater were 95.63% and 91.73%, respectively, demonstrating their excellent applicability [89].

The triphenylmethane dye Acid Green 16 (AG16) is used to colour nylon, wool, cotton, and silk. Genotoxic and mutagenic, it has three aryl radicals linked to a central carbon atom. For Acid Green 16 bulk polymerized imprinted polymer, 1-vinylimidazole was used as a functional monomer and ethylene glycol dimethacrylate as a cross-linker. The HPLC data demonstrated 83% polymer rebinding to the template. Dye Triphenylmethane dyes include Acid Violet 19. 1-vinyl imidazole was chosen as a functional monomer after computational simulation. After extracting with the polymer, Acid Violet 19 was found in complex actual samples with recoveries of 85–99% [90].

10.1.9 Pesticides

The usage of pesticides worldwide to guard against insects, viruses, exotic plants, fungi, and weeds has expanded dramatically in recent decades. Despite their protection, they collect in soils and can reach water level [91]. Thus, their presence and permissible WHO levels should be checked, especially the use of dangerous reagents. Due of their chemical qualities and durability, they can harm the ecosystem as well as target organisms [92].

Since many chemicals in this class are either UV- or electro-inactive, the use of conventional HPLC-UV detection methods is limited, and the use of solid-phase extraction techniques is limited to those that utilize Gas Chromatography (GC) or mass detection (MS). In contrast, molecularly imprinted polymers have been widely used for electrochemical detection in recent years, allowing for the use of an electroactive probe like ferrocyanide even when working with inactive analytes. Many industries have found use for MIPs because of their ability to detect herbicides, insecticides, and fungicides, the three most common types of pesticides.

10.1.10. Insecticides

The type of pesticide known as organophosphates is commonly used, but it can accumulate in water for days or weeks, causing problems for aquatic ecosystems and disrupting marine life [93].

11. Molecular imprinted polymers for Food Applications

11.1 MIPs as sensors

The extremely low LODs attained with MIP-based sensors make them ideal for detecting trace chemicals in food. When the target molecule interacts with the MIPs' precise bonding sites, the resulting physicochemical signal can be recognized and quantified by a transducer [94]. Based on the nature of the signal they generate; MIP-based sensors can be divided into two groups: MIP-based sensors can be classified into two categories according to the type of signal produced:

1) ***Electrochemical MIP sensors:*** MIP-target molecule interactions generate electrical signals, and these signals can be translated by electrochemical MIP sensors [95]. It is possible to obtain a variety of electrochemical MIP-based sensors by varying the current density generated by the potential sweep technique. The most common electrochemical MIP sensor detection techniques include cyclic voltammetry, differential pulse voltammetry, linear sweep voltammetry, and square wave voltammetry. [96], and

2) When a MIP interacts with a target molecule, a spectrophotometric optical signal (such as fluorescence emission or UV-Vis absorption) is generated and can be detected with an optical MIP sensor. This has been made possible with the advent of

revolutionary optical MIP sensors based on quantum cascade lasers (QCM) [97, 98], which make use of surface-enhanced Raman scattering (SERS) and surface plasmon resonance (SPR). QCM-based MIP sensors, in particular, are well-suited to large molecular-size analytes because they can create a substantial mass difference, allowing for the determination of a large signal change when the MIP-target molecule interactions occur [100]. That's why these gadgets can be so sensitive that they can detect molecules at the pico-molar level [101]. For instance, MIP sensors based on hollow-shell type QCM NPs produced by surface imprinting can be used to quantify methimazole (MMZ) (Fig. 12). According to [102], this type of sensor can detect methimazole in beef, milk, and pig samples with a linear range of 5 to 70 ppb, a LOD of 3 ppb, and RSDs of 88.32 to 107.96%. Optical MIP sensors have been the subject of far more research than their electrochemical counterparts.

11.2 MIPs for food contamination sensing

For the food business to ensure food quality and safety, the quick, inexpensive, and accurate detection of food pollutants is essential. In this regard, a number of MIP-based sensors have thus far shown themselves to be effective tools for the identification of a number of food pollutants [103].

11.3 Quantifying dietary nutrients and nutraceuticals with MIPs

Quercetin [104] and sialic acid [105-106] can be extracted from broccoli and onion wastes using MIPs, whereas konjac galactomannan can be used to make polysaccharide-based MIPs.

11.4 Active food packaging MIPs

Food science and technology are interested in active food packaging because it can extend food shelf life and prevent losses [107-108]. Despite this, MIPs have been poorly investigated in this field. Two pioneering research [109] and [110] controlled the release of ferulic acid from MIP-based food nano-packaging to limit lipid oxidation on chicken and butter, respectively.

11.5 Sample fabrication MIPs: disposal, pre-concentration, measurement

Food samples are challenging to analyze because the sample material or insufficient concentrations of analyte can prevent CE, GC, and HPLC detection. [101]. Analyte extraction, purification, and concentration utilised MIPs as absorbing materials since 1994 [111]. MIP-based sorbents have various advantages over conventional ones, including:

1. Selectively concentrating MIP analytes,
2. Thermal resistance makes MIPs flexible.,
3. MIPs are convenient to synthesize.
4. MIPs enhance target molecule recovery and LOD [101].

MIPs have been effective sorbents for many extraction methods, including DSPE, MSPE, MSPDE, μ-SPE, and SBSE [112]. These points are best explained below:

11.6 MIPs for regulated food ingredients release

Recently, MIP has been proposed for use in the food industry for the regulated and prolonged release of active compounds including cosmetic and medicinal ingredients. Several active compounds, including anti-cancer chemicals, can be released slowly in a human body-like environment using MIPs [113]. Riboflavin (vitamin B2) from MIPs was reported to be released under simulated dietary conditions [114].

11.7 MIPs for Pesticides

Assessing pesticides involves several methodologies. Biosensors [116], nanomaterials-based sensors [117], and electrophoretic chips [118-119] have been used to improve HPLC linked to mass spectroscopy [115]. Pesticide sensing with high selectivity and sensitivity helps assess food safety [120-121]. MIP coupled to sensors for pesticide detection can overcome the constraints of classic analytical procedures, especially high selectivity, providing an excellent chance to develop practical, low-cost, and speedy detection methods [118,122-123]. Malathion (MAL) in olive fruits and oils was monitored using a sensitive and selective MIP-modified screen-printed gold electrode (Au-SPE). The MIP sensor was made using acrylamide, bisacrylamide, and MAL. MAL binding was linear in the concentration range of 3×10^{-16} to 3×10^{-12} mol L^{-1}. A 2×10^{-16} mol L^{-1} LOD was established using DPV. [124].

Methyl parathion was quantified using a MIP-IL-Gr/GCE ionic liquid–graphene composite film-coated glassy carbon electrode. The MIP was first synthesized via free radical polymerization with methacrylic acid (MAA) as a functional monomer, ethylene glycol dimethacrylate (EGDMA) as a cross-linker, and 2,20-azobis (isobutyronitrile) (AIBN) as an initiator. CV enabled selective and sensitive MP detection with a LOD of 6×10^{-9} mol L^{-1}. The MIP was successfully used to monitor cabbage and apple peel quality. [121]. Magnetic NPs (MNPs) are commonly employed to synthesize electrochemical sensor-based MIPs. Magnetic fields can modify this substance in pre-concentration to detect targets in complex samples. MNPs can also be integrated on magneto-electrodes for electrochemical reaction output utilizing permanent magnets. Li et al. [125] suggested a field-effect transistor-based MIP sensor for chlortoluron detection. On magnetic nickel (II) oxide (NiO) nanoparticles, MIP was synthesized. The decrease in H_2O_2 oxidation current on NiO nanoparticles modified GCE caused by access obstruction after re-binding can indirectly detect chlortoluron. The significant catalytic action of NiO nanoparticles on H2O2 oxidation led to good sensitivity, with a LOD of 2×10^{-9} mol L^{-1}. MIP sensor analysis yielded 97%–105% recoveries from water samples. Magnetic nickel hexa-cyanoferrate (NiHCF) nanoparticle-coated MIP sensors for chlortoluron measurement were proposed by another research group [126]. This indirect chlorotoluron determination used the change in current from hydrazine hydrate oxidation on the modified electrode. Due to the gate-controlled electro-catalytic amplification effect and magnetic particle enrichment, this approach is suited for trace chlorotoluron analysis. Developed sensor indicated many reuses. Chlorotoluron spiked water samples yielded 97%-105% recoveries with a low LOD of 9×10^{-10} mol L^{-1}. AuNPs, graphene, and imprinted polymer nanocomposites have multiple imprinted cavities, a larger surface area, catalytic oxidation for target analytic, and better physical properties. A carbamate pesticide MIP composite containing rGO-AuNPs was recently developed. The MIP suspension was drop-cast onto a rGO-AuNPS/GCE sensor using carbo-furan as a template, methyl acrylic acid as a monomer, ethylene glycol-maleic acrylate as a cross-linker, and toluene as a progeny solvent. Carbo-furan sensor showed high selectivity and LOD of $2 \times 10-8$ mol L^{-1} [127]. Tan et al. [128] suggested sensitive molecularly imprinted sensors for phoxim measurement using graphene and a novel cross-linker (ethylene glycol maleic resinate acrylate). A graphene-modified GCE was used to generate a MIP film for free radical polymerization phoxim determination. DPV electrochemical approach yielded LOD of 2 × 10^{-8} mol L^{-1}. The imprinted electrochemical sensor recovered 98%–101% phoxim from cucumber samples. An easy flow injection system with a MIP-coated carbon nanotube amperometric sensor was proposed by Amatatongchai et al. [129] to selectively measure profenofos (PFF). The carboxylated CNTs were coated with SiO_2 and vinyl end groups and terminated with MIP shells to make MIP-CNTs. MAA monomer, EGDMA cross-linker, and AIBN initiator grafted MIP onto CNT cores. The template was extracted from a GCE after covering it with 3D-CNTs@MIP to make the PFF sensor. The imprinted sensor detected PFF in vegetable samples with a LOD of 2 × 10^{-9} mol L^{-1}. Zhang et al. [130] presented a simple graphene (GN) imprinting method synthesis to make an electrochemical sensor for sensitive and selective imidacloprid (IDP) residue detection. The polymerization reaction lasted 24 hours at 60 ◦C. The GN-VBA-MIP was drop-cast onto GCE using chitosan.

Electrochemical CV removal of molecules from matrix till imidacloprid signal was absent. An actual sample of brown rice yielded a LOD of 1 × 10^{-7} mol L^{-1}. MIP was prepared using 2-vinyl pyridine, MAA, and acrylamide, three theoretical functional monomers. The template hexazinone herbicide is highly selectively bound by 2-vinyl pyridine-based MIP. [131].

11.8 MIPS for Veterinary Drugs

In 2010–2021, MIPs were used more for veterinary medication determination than for other pollutants, as shown in Figure 1B. Veterinary medications, like other drugs, have biological effects at low doses, making them chemical pollutants. Besides the active molecule or prodrug (inactive precursor that is transformed into an active form by normal metabolic processes), they contain its bioactive metabolites and transformation products. Antimicrobials, antiparasitics, hormones, antifungals, anti-inflammatory drugs (steroidal and non-steroidal), anaesthetics, euthanasia products, tranquillizers, sedatives, bronchodilators, antacids, diuretics, emetics, and emulsifiers are veterinary pharmaceuticals [132-133]. There are seven primary classes of antibiotics: tetracycline, macrolide, aminoglycoside, peptide, lincosamides, streptogramins, and β-lactam. Even at low amounts, antibiotics in the food chain can harm human health. Due to intensive farming and axenic use, antibiotic misuse has increased resistance genes, decreasing illness treatment efficacy. Even Europe and other nations' non-standardized use laws are critical. The development of sensitive, selective, and simple technologies for quick antibiotic screening in food samples is very desirable [134].

Numerous research has used MIP and nanoparticles to increase antibiotic sensitivity and speed. Surya et al. [135] proposed an electrochemical biomimetic ciprofloxacin sensor. An AuNPs-chitosan decorated MIP (Ch-AuMIP) modified GCE for sensor preparation. AuNPs and Ch- MIP synergistically determine CIP (LOD = 2×10^{-7} mol L^{-1}) in various samples, including water, mineral water, milk, and pharmaceutical formulation, with a recovery rate of 94-106%, even with Norfloxacin and Ofloxacin present. Liu et al. [136] presented a self-supported Fe_3N-Co_2N nano-array with good electrical conductivity and extensive surface area growth on MIPs to create a sensitive and stable ampicillin electrochemical sensor. Fe_3N-Co_2N/carbon cloth was hydrothermally produced first. MIPs were produced by in situ polymerizations on the Fe_3N-Co_2N/carbon cloth electrode. The electrochemical sensor for MIPs detected ampicillin with a low LOD (4×10^{-10} mol L^{-1}) and shown good reproducibility and stability. Wie et al. 2021 [137] suggested a sulfanilamide electrochemical sensor using a GCE modified with MIP and GO. The MIP/GO material was made by precipitation polymerization at 60 ◦C for 24 h using sulfanilamide as a template and MAA as a monomer. SWV electrochemical approach quantified sulfanilamide in milk with a LOD of 6×10^{-10} mol L^{-1}. Sulfamethoxazole (SMX) as the template and MAA as the monomer were employed to generate free radicals via hemin/graphene hybrid nanosheets (H-GNs) to start imprinting polymerization by Guo et al. [138]. MIPs employing sulfamethoxazole as the template were directly produced on H-GNs without film modification. Most notably, the template may be adsorbed on H-GNs to increase imprinted sites and MIP film selectivity. The LOD was 5×10^{-12} mol L^{-1}. This sensor was free from sulfamethoxazole analogue interference, which is a novel insight for MIPs-based food safety monitoring sensors.

Estradiol is commonly utilised in veterinary medicine [139]. The magnetic molecularly imprinted sensing film (MMISF) by Han et al. [140] measured estradiol (E2). The MagMIPs were synthesized via in situ polymerizations of aniline and glutathione (GSH)-functionalized gold (Au)-coated Fe_3O_4 nanocomposites. A magnetic GCE (MGCE) was softly modified with MagMIPs to create the MMISF or freely installed a magnet to remove it. The MMISF method detected E2 in milk powder with high sensitivity, selectivity, and repeatability, with a LOD of 3×10^{-9} mol L^{-1}.

Bai et al. [141] created an AuNPs/MWCNTs-Chitosan electrochemical sensor for diethylstilbestrol (synthetic oestrogen and growth promoter). The sol-gel process produced MIP. A GCE was modified step-by-step with AuNPs and MWCNTs-Chitosan. These two nanomaterials increased electrode surface area to boost electron transport and sensor signal. MIP was electrodeposited on the electrode. The DPV measurements showed a very low LOD of 9×10^{-17} mol L^{-1}.

11.9 MIPs for Processing and Packaging Food Toxins

Chemical reactions between food elements and ingredients can produce undesirable chemical compounds during preparation. Materials, machinery, containers, and packaging might release them. Heat processing food (baking, frying, etc.) causes reactions between its components, some of which might generate unwanted chemicals like acrylamide. For instance, plastic food storage can leak harmful chemicals. Phthalates and bis-phenol A are the most common plasticizers used in toys, food packaging, and pharmaceuticals. Published research has linked these synthetic chemicals' teratogenic and carcinogenic impacts to the health of all living things on Earth [142-143]. However, the methods for detecting phthalates include costly machinery, lengthy processing times, and convoluted processes. A GCE MIP-based sensor for Diisononyl phthalate (DINP) was proposed by Zhao et al. [144]. Electrode construction involved combining MIPs and agarose. Without any prior sample pretreatment, the MIPs-modified electrode is able to detect the analyte. Direct detection by CV yielded a LOD of $3 \text{ x } 10^{10}$ mol L^{-1} in a white liquor sample. For the analysis of dibutyl phthalate, Li et al. [145] presented a composite of Magnetic Graphene Oxide Gold Nanoparticles- (MGO-AuNPs) and MIPs. It was through the use of co-precipitation and self-assembly procedures that the MGO@AuNPs composite was initially synthesized. Due to MGO AuNPs' high affinity, the template molecules (DBP) were readily taken up by their surface. Using DPV as the electrochemical approach, the electrochemical sensor for DBP demonstrated good reproducibility for 30 repeated tests and a low LOD ($8 \text{ x } 10^{10}$ mol L^{-1}). For the detection of bisphenol A (BPA) in plastic water bottles and metal cans, Deng et al.

[146] presented a graphene flakes doped chitosan film as an imprinted sensing nanocomposite on an acetylene black paste electrode (ABPE). Using derivative voltammetry, the LOD was determined to be 6 x 10^{9} mol L^{-1}.

The GCE modified with amino-functionalized graphene oxide and MIP was created by Dadkhah et al. [147] for electrochemical sensing of BPA. Adding amino groups to GO was a breeze because of the use of 3-aminopropyltriethoxysilane (APTES). The recognition performance of MIP-based sensors was then enhanced by immobilizing the template onto amino-functionalized GO. Michael addition reaction was used to graft ethylene glycol dimethacrylate onto APTES-coated graphene oxide sheets before polymerization. This process resulted in the formation of several uniform imprinting sites on the GO sheets. To get a direct DPV reading of BPA, the resultant composite was mounted on a GCE. Without any pre-treatment or matrix interfering effects, a LOD of 3 x 10^{-9} mol L^{-1} was found in both milk and mineralized water. Graphene (Gr) modified acetylene black was proposed as a component in MIP synthesis by Xu et al. [148]. The solvent was removed from the MIPs by drop casting them onto Gr/ABPE and then heating the substrate with an IR light. Using DPV as an electrochemical probe, we determined that the LOD for BPA on the constructed electrode was 2 x 10^{-13} mol L^{-1}.

11.10 MIPs for Biogenic Amines

Health risks have been linked to eating food that has high quantities of biogenic amines (BAs). The eight biogenic amines that are regarded to be the most significant in foods are histamine, putrescine, tryptamine, cadaverine, tyramine, b-phenylethylamine, spermidine, and spermine. For their toxicity and use as gauges of the level of food freshness or decomposition, BAs must undergo analysis with high precision and selectivity [149-150].

MIP linked electrochemical sensors have recently been studied and effectively used to authentic food matrices for the determination of BAs in foodstuff. By decarboxylation of tyrosine, tyramine is a BA created. In addition to meat, fish, and other dairy products, it is extensively present in fermented foods and drinks. MIP was created on a sol-gel matrix by Huang et al. [151] for the detection of tyramine in yoghurt samples using a mixture of MWCNT-gold nanoparticle (MWCNT-AuNP) composites and chitosan polymer.

Tyramine served as an initial molecule for the MIP's synthesis, and silicic acid tetracthyl ester, triethoxyphenylsilane, and EGDMA were used as cross- linkage agents. The surface of the MWCNT-AuNP/chitosan/GCE was dropped-cast with the tyramine-MIP solution. Chitosan serves as a bridge for the imprinted layer, while MWCNT-AuNP composites were added to improve conductance and subsequently the sensitization of the electrode. With the help of direct amperometric measurement, a LOD of 6 x 10^8 mol. L^{-1} had been reported.

The enzyme L-histidine decarboxylase catalyzes the decarboxylation of the amino acid histidine to produce histamine, another natural amine. [152]. Foodstuffs and bodily fluids contain histamine when exposed to extreme conditions (such as acidic or basic conditions, chemical solvents, etc.). Thus, it is essential to create a strong technique for analysis that can withstand extreme pH situation. An impedimetric MIP-based sensor to identify low levels of histamine was studied by Bongaers et al. [153] MIP was created using bulk polymerization. The created MIP-sensor for histamine showed remarkable sensitivity, a LOD of 2×10^9 mol/L, and no histidine interference responses while demonstrating its stability over a wide pH range of 5 to 12.

Conclusions and Future Perspectives

Wearable sensors and healthcare systems can supply real-time medical services efficiently. As the population ages and health costs rise, preventive care and lifestyle changes are becoming increasingly important. Wearable health coaching gadgets will be needed to monitor patients accurately and thoroughly to improve healthcare quality, availability, and affordability worldwide. Wearable devices scan and transmit data, including body and activity signs. Wearable health coaching technologies raise new research questions and satisfy old ones. Privateness, sensor accuracy, and medical ecosystem compatibility are a few. Research has important healthcare responsibilities in addition to these challenges. Due to this, wearables will take longer to hit shop shelves. The issues that need to be addressed include the basics of electrochemistry in wearable conditions, software algorithms to treat signal artefacts in real-time wearable detection, sensors for long-term wearable sensing in the body fluid, platforms for testing newly developed sensors, validation of test results, reproducible and standardized sampling methods, and the clinical significance of wearable testing data.

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