Review article

Molecularly imprinted polymer composite membranes: From synthesis to diverse applications

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ABSTRACT

This review underscores the fundamentals of MIP-CMs and systematically summarizes their synthetic strategies and applications, and potential developments. MIP-CMs are widely acclaimed for their versatility, finding applications in separation, filtration, detection, and trace analysis, as well as serving as scaffolds in a range of analytical, biomedical and industrial contexts. Also characterized by extraordinary selectivity, remarkable sensitivity, and outstanding capability to bind molecules, those membranes are also cost-effective, highly stable, and configurable in terms of recognition and, therefore, inalienable in various application fields. Issues relating to the potential future for the paper are discussed in the last section with the focus on the improvement of resource practical application across different areas. Hence, this review can be seen as a kind of cookbook for the design and fabrication of MIP-CMs with an intention to expand the scope of their application.

1. Introduction

The advent of molecularly imprinted polymer (MIP) membranes has transformed the paradigm of selective separation and filtration in the membrane technology. These membranes are optimally structured and chemically composed to effectively captured and interact with target molecules. Significant advancements this vital area of both research and industry have been achieved through the deployment of varied and effective strategies [1,2]. To the pharmaceutical industry [3,4], they are very important for purification and trace detection of active compounds [5,6]. For environmental scientists, it is a form of pollution detection and is used for the selective sensing and removal of pollutants [7,8]. For the foods and beverages department, it is important for detecting the contaminants and assuring more safety standards of the commodities [9,10]. High levels of specificity, stability, and reusability in any particular application are a true benefit that will benefit both the scientific and industrial sectors.

Molecularly Imprinted Polymer membranes have been further enhanced by converting them into molecularly imprinted composite membranes (MIP-CMs) through the strategic incorporation of various advanced additives. These additives, including metal

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nanoparticles [11–14], graphene [15–17], carbon nanotubes [18,19], inorganic ceramics [20–22], titanium dioxide [23–26], gold nanoparticles [11,27], and metal-organic frameworks (MOFs) [28–31] significantly elevate the membranes' functional properties and overall performance. The integration of metal nanoparticles, for instance, markedly enhances the membranes' catalytic activity and electrical conductivity, making them highly efficient in catalytic and electronic applications [32–34]. Graphene and carbon nanotubes, known for their exceptional mechanical strength and electrical conductivity, contribute to the increased durability and functionality of the membranes [35,36]. Inorganic ceramics additives offer improved thermal stability and permeability, crucial for high-temperature applications and robust separation processes [37–39]. Titanium dioxide and gold nanoparticles improve the membranes' durability and chemical resistance, ensuring long-term stability in harsh chemical environments. Optical properties, make the membranes ideal for advanced sensing applications, enabling precise detection and monitoring [40–42].

General publication trends of applied research related to MIP-CMs from 2010 to 2024 can be seen in Fig. 1a, b (In all, 1200 publications have been retrieved from the Web of Science database between 2010 and May 2024). They may be absolutely normal, considering the intensity and focus of research for this particular period, 2010 to 2024. The steady upward growth up to 105 in 2022 shows that the interest of researchers towards the MIP technology is in a state of rapid increase for drug delivery, filtration, and environmental remediation applications. This does not represent the sum of effort but may represent the maturation of methodologies in the MIP community. It may account for a lower number of published articles after the peak witnessed because the consolidation period is visible, from which one can therefore infer that the researchers are basically synthesizing knowledge gained or laying emphasis on the commercialization practical application aspects. Such a general trend flags the dynamic and adaptive character of MIP research for the response to technological development and evolving scientific priorities over time, holding potential for saying something about future innovations and the potential rise of new exploratory opportunities in this field. Most publications relate to the section of analytical chemistry, polymer science and engineering fields of the Web of Science database.

These sophisticated MIP-CMs are synthesized through various cutting-edge techniques such as in situ polymerization [35], surface grafting [43,44], electro-polymerization [45,46], sol-gel processes [47], and layer-by-layer assembly [48]. Each method allows for meticulous control over the distribution and concentration of the additives within the polymer matrix. This precise control ensures that the functional properties of the composite membranes are finely tuned for their intended applications, leading to enhanced performance in a broad spectrum of fields [49,50]. In environmental sensing, the high sensitivity and specificity of these membranes enable the detection of trace pollutants and contaminants with remarkable accuracy [49,51]. In drug delivery, the well-designed release rates and improved stabilities of the membranes lead to better therapeutic results [52–54]. Also, in the more advanced filtration systems, these membranes have higher permeability and high selectivity and this enables the separation and purification of the targeted molecules from other compounds. Thus, the design and synthesis of MIP-CMs allow for the expansion of research and applications horizons, which lead to the great progress in a wide range of fields of science and industry. In doing so this technology not only fulfills the emerging need for accuracy in pharmaceutical and environmental industries but also paves new ways for high standards of performance in these sectors. Due to their ability to extend the limits of molecular recognition, MIP-CMs hold immense potential to change the conventional ways of dealing with molecular separation and targeted drug delivery [49,55,56].

In this review, the synthesis and application of MIP-CMs have been discussed based on the recent literature; emphasis has been laid on new developments and future trends. Thus, this review synthesizes current knowledge to advance the development of this
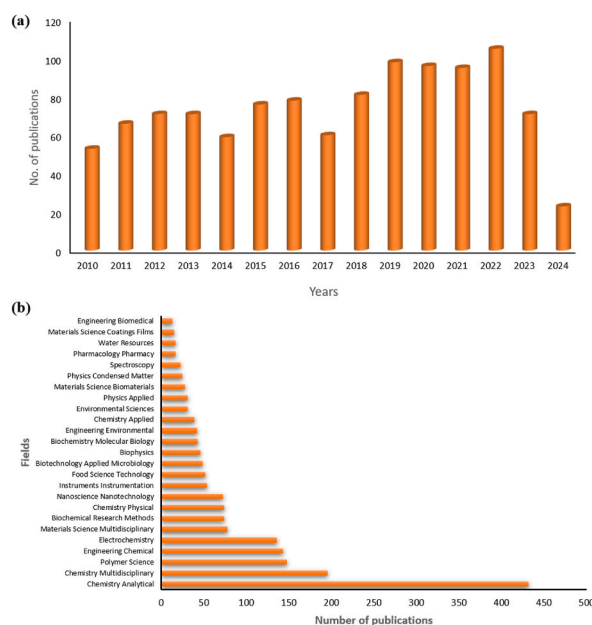

Fig. 1. (a) Number of publications by year (based on WOS Database by marking the time span from 2010 to 2024). (b) Number of publications by field (data retrieved from the WOS database through the selection of the period 2010–2024).

important area within the broader field of materials science.

2. Fundamentals of MIP-CMs

Molecularly imprinted polymer composite membranes (MIP-CMs) have a high degree of selectivity and specificity towards target molecules because they contain specific binding sites that imitate the natural recognition sites found in biological molecules [57,58]. The high specificity of MIP-CMs makes them suitable for use in sensor technologies [59], separation processes [60], and targeted drugs delivery [61]. Moreover, they possess a high level of durability, chemical stability, and the ability to operate well in challenging conditions that could potentially cause defects in biological systems [1,62,63]. However, there are some challenges that the process for developing MIP-CMs might be complicated and time-consuming, typically needing careful control of the polymerization conditions in order to attain the desired specificity [64]. Additionally, the ability of MIPs to bind may be limited by the availability of the imprinted sites, and the elimination of template molecules following polymerization can occasionally be insufficient, which can impact the effectiveness of the membranes. Moreover, the use of these membranes might be hampered by the inflexibility of the polymer matrix, leading to a delayed movement of bigger molecules and a decrease in the overall performance of the membrane [1,65,66].

Due to their predictable structure and specific recognition, MIP-CMs have varied applications in different fields [67]. MIP-CMs are crafted in the presence of a template, acting as a mold for the creation of template-matching binding sites. This enables them to discern a diverse range of target molecules with an affinity and selectivity akin to antibodies [68]. MIP-CMs demonstrate high compatibility for the template molecule in comparison with the other structurally analogous compounds. MIP-CMs are widely employed in chemical sensors [69–71] and separation media [72,73]. Nevertheless, conventional MIP-CMs have certain shortcomings [74], such as imperfect removal of the template, poor-affinity binding and lethargic mass transfer [75]. An array of microfiltration, ultrafiltration and reverse osmosis membranes comprised of various natural or synthetic polymers is now accessible and operated in a growing number of separation techniques in industries, environmental protection, life sciences, medicinal purposes and biotechnology [76]. Regrettably, selective separation of individual materials is not facilitated by the typical commercial membranes. Therefore, techniques for the development of membranes with well managed specificity for individual materials have grabbed a great attention in the last few years [77]. Membrane based chemical separation finds its usability in broad range of research areas and industrial technologies [78–80]. Anyhow, materials with high selectivity for the desired target molecules, incorporated into the geometry of membrane resulting in enhanced selectivity are required to further increase the process of separation [81,82].

3. Synthesis techniques for MIP-CMs

The fabrication of MIP-CMs includes a series of phases, starting with the choice of an appropriate template molecule and functional monomers. The monomers combine with the template to make a complex, which is subsequently polymerized with cross-linkers to produce an effective polymer network [83]. The initiation of polymerization can be achieved using thermal, photochemical, or chemical approaches, depending on the specific qualities sought for the membrane [84,85]. Following the process of polymerization, the template molecule is separated, resulting in the formation of cavities that possess a complementary structure, size, and functional groups to the template [55,86]. This precise imprinting technique imparts the membrane with a high level of selectivity and specificity.
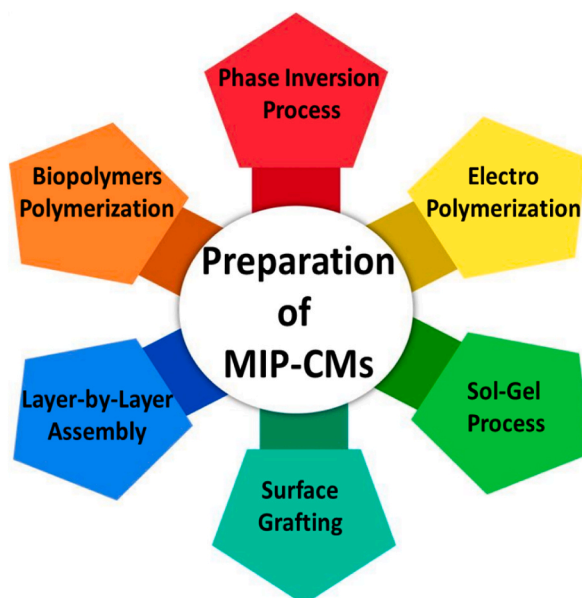

Fig. 2. Various methods of preparations of MIP-CMs.

Although MIP-CMs present many advantages, their preparation can frequently be complicated and time-consuming, demanding strict regulation of polymerization conditions [87,88]. In addition, the elimination of template molecules may be inadequate which could adversely impact the membrane's functionality. The polymer matrix's rigidity, although advantageous for stability, can impede the diffusion of bigger molecules and decrease the membrane's performance in specific applications [64,66,89]. According to Kochkodan et al. the molecularly imprinted membranes are prepared by various methods, among which the phase inversion method is most widely used. However, the membranes formed by this method suffer from low yield of recognition sites with random distribution. These drawbacks minimize the efficiency of membranes for selective separation. To overcome these problems, the formation of composite imprinted membranes with porous structures and high adsorption ability may be useful [90,91]. Different technologies have been employed to incorporate the pre synthesized MIP particles with or into the membranes. These include coating MIP particles onto the surface of membrane discs [92], encapsulation of MIP nanoparticles between two membrane layers [93], surface grafting of the MIP onto the membrane pores by photoionization polymerization [94], pore filling of the thin tracked etched membranes with MIP [95], and encapsulation of MIP nanoparticles into a composite nanofiber membrane by electrospinning [96]. The above strategies use the following processes for the synthesis of MIP-CMs as illustrated in Fig. 2.

3.1. Phase inversion process

The method of phase inversion for the preparation of MIP-CMs involves the formation of a uniform solution of polymers, template molecules, functional monomers, and a solvent. Subsequently, the solution is transformed into a thin film, and the phase inversion is triggered by submerging the thin film in a non-solvent solution, leading to the precipitation of the polymer and the creation of a porous membrane structure. Afterwards, the template molecule is removed, leaving only the specific binding sites [8,55]. One advantage of this approach is the ability to regulate the porosity and thickness of the membrane, resulting in customized separation features and improved selectivity for the desired molecule. Furthermore, phase inversion is a highly adaptable and scalable method that is well-suited for use in industrial environments [97–99]. Nevertheless, the drawbacks include the possible complexity and challenge in attaining a consistent dispersion of the imprinted areas throughout the membrane. Furthermore, if the template molecules are not completely removed, it can negatively affect the function of the membrane. Additionally, the need for accurate control of the phase inversion parameters may render the process costly and challenging to optimize [98,99]. For instance, Niavarani et al. synthesized flat polyethersulfone membranes using a method called non-solvent induced phase inversion. These membranes were made with 14 % PES, 21 % N-methyl-2-pyrrolidone as the solvent, and 65 % polyethylene glycol as the agent for forming pores. To make composite membranes, they added molecularly imprinted or non-imprinted particles in amounts ranging from 0 to 4 %, mixed them for 30 min at 3500 revolutions per minute. The resulting mixture was then spread onto a glass plate and placed in a water vapor chamber for 5 min before being immersed in a cold-water tank. After this immersion, the membranes were subjected to electron beam irradiation under
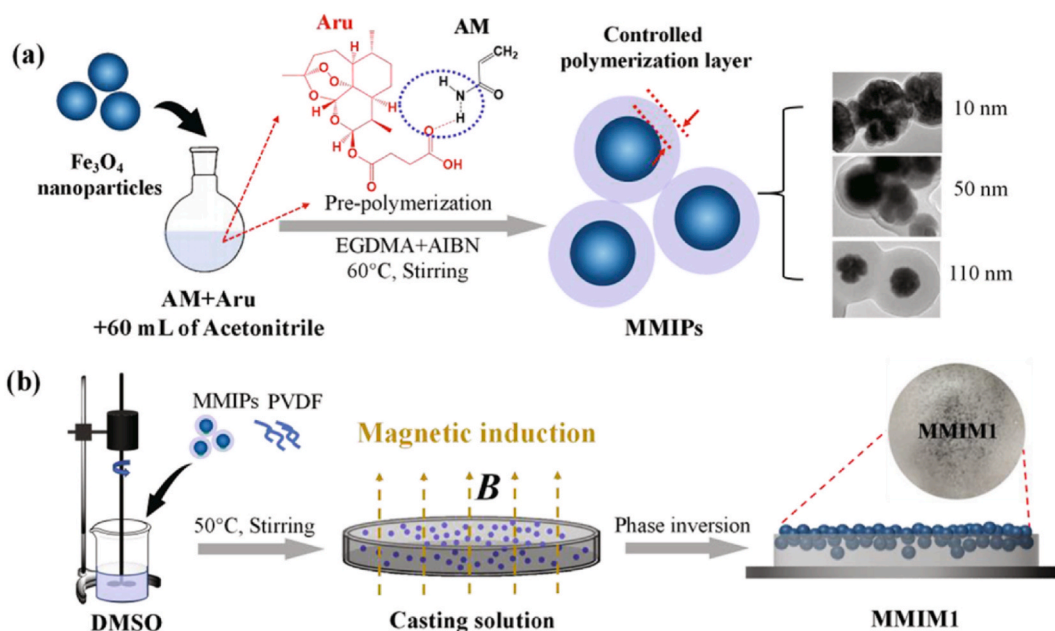

Fig. 3. (a) Shows the synthesis of MMIPs by precipitation polymerization. Fe₃O₄ nanoparticles and EGDMA were added to a mixture of ARU and AM in acetonitrile, followed by sonication and polymerization at 70 °C for 12 h (reproduced with permission) [103], (b) shows the fabrication of MMIM1 using phase inversion under a magnetic field to align MMIPs on the membrane surface. A PVDF and MMIP solution was dissolved in DMSO, poured into a Petri dish with a magnet above it, quickly frozen, and subjected to phase inversion in deionized water (reproduced with permission) [103].

nitrogen conditions (150 kGy, 160 kV, 10 mA) to embed the particles into the PES matrix [100]. Stoica et al. developed specialized membranes using copolymers of acrylonitrile and methacrylic acid through a method called wet phase inversion. These copolymers, which dissolve in dimethylformamide, allowed the creation of thin membranes. Acrylonitrile provided structural support with its polar cyano groups, while methacrylic acid was chosen for its ability to bind with polar molecules like ephedrine. The polymerization process utilized a redox initiator system comprising potassium persulfate and sodium metabisulfite at an acidic pH. Two specific copolymers were synthesized: one with 80 % acrylonitrile and 20 % methacrylic acid, and another with 75 % acrylonitrile and 25 % methacrylic acid. To prepare the membranes, 4 g of each copolymer was dissolved in 42 mL of dimethylformamide. For the molecularly imprinted membranes, ephedrine hydrochloride was added to the solution. The solutions were then spread onto glass supports and immediately immersed in distilled water at room temperature for phase inversion. This was followed by a 24-h stabilization in a coagulation bath. The membranes were dried at 60 °C for 48 h and then milled to ensure uniformity. Rebinding studies were conducted using the wet form of the membranes to prevent shrinkage of the imprinted cavities [101].

Huang al. reported a detailed synthesis methodology for the preparation of MIP-CMs with specific affinity for kaempferol by initially dissolving kaempferol and monomer 4-VP in acetonitrile and stirring for a while. After, crosslinker EDMA and initiator AIBN were added, and the resulting mixture was allowed polymerization in a 70°C-oil bath under nitrogen for 24 h. The resulting polymers, in turn, were submitted to a Soxhlex extraction with MeOH/acetic acid in order to remove kaempferol template and further washed until complete removal of acetic acid residues with MeOH. The polymers were dried under vacuum at 60 °C resulting in the MIP. The components of the MIP-CMs, the PPSU and kaempferol, were dissolved in the DMAC and homogenized at 60 °C and then cast on glass plates solidified through the phase inversion process. Membranes were treated likewise to the polymers for the removal of solvents. Nonimprinted versions were also synthesized by the same methods but without the kaempferol, and modifications were prepared, in which in the preparation of the membranes, kaempferol was replaced by either MIP or NIP [102]. Bai et al. prepared the magnetic molecularly imprinted polymers (MMIPs) by precipitation polymerization (see Fig. 3a). A solution of ARU and AM in acetonitrile was mixed with Fe_3O_4 nanoparticles as the core, where EGDMA was used to crosslink them. AIBN was added after sonication, and the mixture was polymerized at 70 °C for 12 h. The MMIPs were then separated and purified. Magnetic non-imprinted polymers (MNIPs) were synthesized using the same procedure without the ARU template. Meanwhile, MMIM1 was generated through magnetic field-aligned phase inversion to create a surface layer of MMIPs on the membrane (See Fig. 3b). Dissolved PVDF and MMIPs in DMSO
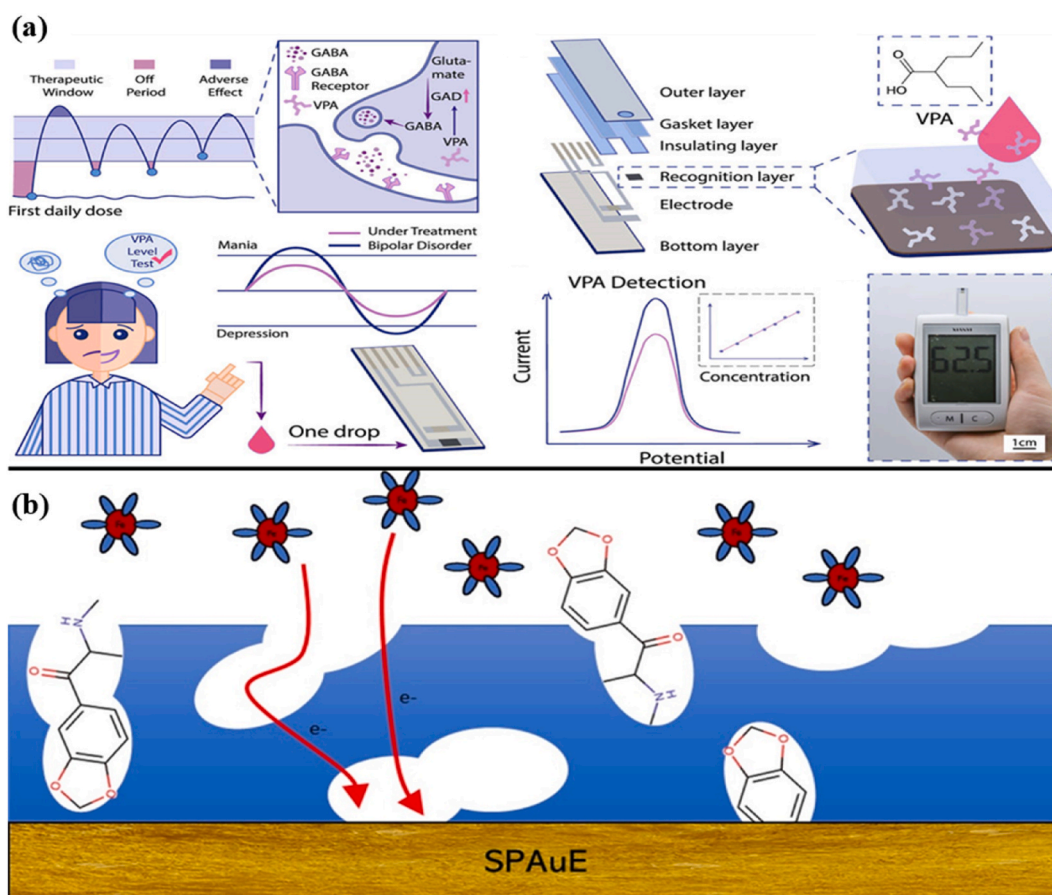

Fig. 4. (a) An electropolymerized biomimetic polypyrrole membrane-based electrochemical sensor for selective valproate detection (reproduced with permission) [46], (b) Screen-Printed Electrodes Modified with Electropolymerized MIPs for Methylone (reproduced with permission) [113].

were cast into a Petri dish with a magnet on top, followed by quick coagulation of the system using ice-cold water. The control membrane (MMIM0) was prepared without magnetic guidance [103].

3.2. Electro-polymerization technique

Electro-polymerization is a technique employed for fabricating MIP-CMs by subjecting a solution comprising the template molecule, functional monomers, along with electrolyte to an electric potential. This process initiates the formation of a polymer directly on the surface of an electrode, resulting in the formation of a thin and uniform polymer film. This method allows for specific control of the film's thickness and morphology. Afterwards, the template is removed in order to generate distinct binding sites within the membrane [104–107]. Electro-polymerization offers the advantage of producing MIP membranes that are extremely consistent and well defined, with perfect regulation of film thickness. This leads to improved selectivity and sensitivity of the membranes. This technique is also quite efficient and may be implemented under normal conditions [108–110]. Nevertheless, one limitation of this approach is the demand for conductive substrates, which entails limitations on the range of materials suitable for use as supports. In addition, the technique may not be well-suited for large-scale production since it requires specialized equipment and there is a risk of partial removal of template molecules, which might adversely affect the performance of the imprinted sites [111,112].

The electropolymerization imprinting technology described by Yuan et al. the imprinting of polymer films is done using cyclic voltammetry (see Fig. 4a). This process commences through the electrochemical deposition of gold nanoparticles on SPEs by immersing the SPEs in 10 mM HAuCl$_4$ and 5 mM HCl and applying a potential of 0 V for 100s. The electrode is then plunged into the de-aerated polymerization solution with 0. 025 M pyrrole. The patients enrolled had VPA as the only AED with mean plasma level of 1 mol L^{-1} and 0.1 M LiClO$_4$. The electrochemical polymerization is performed for seven cycles with the potential starting from −0. 6 and 0. With the applied potential of 8 V, the deposition process was performed using cyclic voltammetry in which the scan rate was 50 mV s^{-1} and this favored the formation of a polymer film that encapsulates the template molecules. After the post-polymerization, the VPA template is removed by applying +1. This paper was prepared using the following sample treatment: the sample was soaked at 3 V for 20 min. This was followed by washing the mixture with 2M K$_2$HPO$_4$ solution and at the end the only suitable shaped imprinted sites are left. This is due to the well-regulated voltage and current applied in the process which enhances the deposition of polymer and the
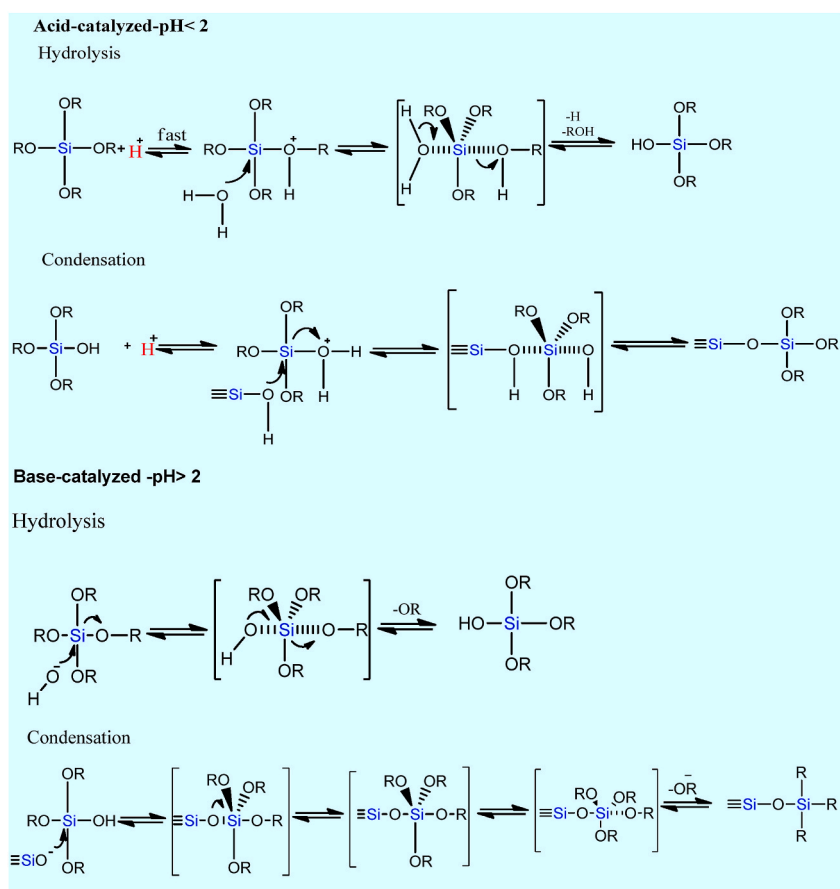

Fig. 5. Common pathways of converting alkoxysilane precursors to silicon dioxide and its polymerization in acidic environment and basic environment catalyzed conditions. Condensation of a liquid can result in the formation of either water or alcohol [116].

production of binding sites on the electrode [46]. For instance, Couto et al. designed methylone sensors for detection as shown in Fig. 4b. The detection scheme started from the fabrication of a screen-printed gold electrode (SPAuE) from a 10-mm-diameter gold working electrode that was washed and dried and then activated electrochemically by cyclic voltammetry in sulfuric acid. A thin film of target analyte methylone molecularly imprinted polymer was synthesized on the electrode by electropolymerization of a 2-MBI monomer solution of 100 mM concentration and, also, a 5 mM target analyte methylone was added. The electrode was then cycled through a series of CV scan numbers after which the analyte methylone was cleaned and extracted. A non-imprinted polymer (NIP) sensor was fabricated in an identical manner, but the target analyte was not included to ensure a suitable control. The sensors were characterized by CV and electrochemical impedance spectroscopy (EIS) in an electrolyte of phosphate-buffered saline (PBS) containing a $[Fe(CN)_6)]^{3-/4-}$ redox probe. The sensing behavior of the sensor has been compared with a number of other techniques by LC-MSn and GC-MS as shown in their reference table. All measurements were recorded at room temperature [113].

3.3. Sol-gel process

The sol-gel production of MIP-CMs involve the conversion of a solution containing monomers, template molecules, and a Sol-gel precursor (usually metal alkoxides) into a gel by means of hydrolysis and polycondensation processes. This leads to the formation of a permeable, three-dimensional structure that includes the imprinted sites inside the inorganic-organic hybrid. Subsequently, the template molecules are removed, resulting in the retention of distinct binding sites [49,114,115]. In hydrolysis, metal alkoxides react with water to replace alkoxide groups with hydroxyl groups as has been illustrated in equation (1).

$$nSi(OR)_4 + 2nH_2O \rightarrow nSiO_2 + 4nROH \tag{1}$$

The general formula for its representation for compounds with amino group is $+4nROH$, where R stands for an alkyl radical. The condensation reactions that follow these chain hydrolyzed products connect the products by Si–O–Si bonds and the release of small molecules like water or alcohol which explain the porosity of aerogels and xerogels. The structure, porosity, and surface area of the resultant material are also affected by the pH of the solution; the rate of these reactions varies with this parameter. Acid catalysis whereby the pH of the solution is below 2 while that of base catalysis whereby the pH is above 2 influence the gelation point and some characteristics of the gel (See Fig. 5). Moreover, the size and steric effects of the alkyl groups of the alkoxide precursor will also hinder the rate of hydrolysis and condensation thus affecting the denseness and mechanical strength of the gel. By these means, the selection of metal alkoxides determines the gel microstructure which is pervasively essential when producing materials for diverse applications say, for sensing and insulation [116].

Sol-gel synthesis offers several advantages, such as its ability to construct membranes that possess exceptional thermal and chemical stability, customizable porosity, and flexible surface features. These characteristics make sol-gel membranes appropriate for a wide range of applications, including sensor technology and separation processes. Furthermore, the moderate synthesis conditions maintain the functionality of both the template and monomers. Nevertheless, there are drawbacks to consider, such as the challenge in producing a consistent distribution of the imprinted sites and the risk of incomplete removal of the template, which might negatively impact the performance of the membrane. The procedure can also be intricate and time-consuming, requiring fine regulation of reaction conditions to attain desirable membrane features [117–119]. For instance, Wu et al. synthesized the GO/TiO₂-loaded nanocomposite fibrous membranes using polydopamine/gel based imprinting processes which were named as GT-DIMs. These membranes showed reasonably higher enhancements and possessed a fairly good rebinding capacity of about 70.63 mg g^{-1} for both and, reaches the adsorption equilibrium within 30 min. The permselectivity factors were approximately 5.0, demonstrating highly selective and efficient recognition and separation in multichannel systems and in the vidal realistic and laboratory-simulated wastewater matrices. The sol-gel method helped to develop new GO/TiO₂-based channels for the continuous improvement of the structure's stability and separation performance in actual uses [120]. Kalogiouri et al. prepared the molecular imprinted sol-gel silica-based hybrid polymeric sorbent for BPA using phenyl triethoxysilane and 3-aminopropyl triethoxysilane for the selectivity and pre structure of tetraethyl orthosilicate for efficient template to analyte transfer. This method gave relatively higher percentage extraction of BPA from the water samples with HPR/MIP-SE as compared to the HPR with a recovery of 93 % as analyzed using molecular imprinted solid phase extraction followed by high pressure liquid chromatography photodiode array analysis of 4 % and LOD of 0. 015 to 20 000 ng μL^{-1}. The Linearity of the method was from 0. 015–50 ng μL^{-1} and the LOD in this study was found to be 0. 045 ng μL^{-1} (LOQ) [118]. Wu et al. fabricated rich-featured molecularly imprinted nanocomposite membranes on basswood by using surfaces with TiO₂@PDA and the sol-gel process. Thus, the resulting 3D mesoporous wood-based membranes possessed a high rebinding capacity of 150. Five milligrams per gram, fast adsorption and high permselectivity coefficients being more than 9.0. It was concluded that PDA-modification of the layers led to a significant increase in TC-imprinted sites [121]. Zhi et al. developed a quercetin-detecting MIP-CM surface through sol-gel technology. The process was initiated by activating silica (SiO₂) through refluxing in methane sulfonic acid. After the treatment, the SiO₂ was separated, dried, and neutrally washed. Quercetin and APTES were stirred in acetone to form a monomer-templated complex. The activated SiO₂ was mixed with the above complex and further treated with TEOS and acetic acid to form a rigid polymeric network in the cross-linking on the surface of SiO₂. This creates an MIP by anchoring the complex to the SiO₂ surface, and after the extraction of the template molecule (quercetin), the template cavities are left behind, which are complementary both in shape and size to that of quercetin. In this way, the MIP is able to selectively rebind quercetin in order to discriminate it from its analogues in mixed samples [122]. Cheng et al. have reported the preparation of molecularly imprinted composite membranes (MICM) for evodiamine detection using sol-gel technology. A polysulfone (PSF) membrane was first synthesized by dissolving PSF and polyvinylpyrrolidone (PVP) in dimethylacetamide (DMAc) - a membrane casting solvent. The synthesized solution was then deaerated and

finally formed into membranes by phase inversion. Evodiamine was first dissolved in a mixture of methanol-tetrahydrofuran and then mixed with 3-aminopropyl triethoxysilane (APTES) to yield a complex. After complexing, the evodiamine-APTES complex was cross-linked with tetraethyl orthosilicate (TEOS). A prepolymer solution was prepared in a mixture solution of methanol-tetrahydrofuran (3:1 V:V) and the PSF membrane was immersed in the solution. After the addition of ammonia, poly-merization was initiated to form the polysulfone-evodiamine MICM. The membrane was then subjected to post-curing and washing to remove the template molecule, leaving specific binding sites for evodiamine. A non-imprinted version (NICM) was also prepared for comparison, excluding the evodiamine template [123].

Overall fabrication of boronate-affinity sol–gel molecularly imprinted membranes: performance A schematic representation is shown in Fig. 6. Effective addition of polydopamine into the MOFs mixed-matrix membranes using a rotary evaporation procedure is shown to enhance both the selectivity and the adsorption V-Value for shikimic acid to >5.0 with an excellent adsorption capacity to 107.87 mg g^{-1}. This approach provides a significant platform for the generation of advanced, selective separation membranes [47].

3.4. Surface grafting technique

There are a number of ways given above to impart the above characteristics to the membrane application. The most common use is surficial grafting of a functional hydrogel layer to the pore surface of the base membrane [124–126]. A technique to prepare such a membrane is the two-step surface grafting technique, which results in the production of an antibody-imprinted membrane [127,128]. In the research work of Zeng et al. a silica layer was grafted onto the surface of a ceramic membrane (CM) using a two-step, in situ hydrolysis deposition approach. Briefly, a disc of CM was prepared as follows. It was first treated with a dilute solution of HCl (5 wt%) for 24 h. The disc was then immersed in a TEOS/ethanol mixture (volume ratio = 1:15) and reacted at 70 °C for 30 h. The resultant was further calcined at 550 °C for 3 h and then cooled, washed by ultrasound, and re-immersed in a 1 wt% HCl solution for a further 24 h to activate the silanol groups. Once it was neutralized by extensive rinsing, the SiO$_2$-CM was dried in a vacuum oven at 80 °C for 8 h resulting in an activated silanol group-coated SiO$_2$-CM. In a second preparation, SiO$_2$-CM was modified with MPS at 110 °C for 5 h in a nitrogen atmosphere in a sealed reactor. After washing and drying, this resulted in the production of MPS-CM. With the use of a surface-initiated graft-polymerization approach, VI and AIBN were reacted with MPS-CM in DMF under nitrogen at 70 °C for 8 h. The resultant was washed and dried, thus obtaining the grafted PVI-CM. In preparing the Mo(VI) anion surface imprinted material, designated as the IIP-PVI/CM, the PVI/CM was incubated in an 8 mmol/L Mo(VI) at pH 4.0 at 30 °C for 2 h. The second cross-linking
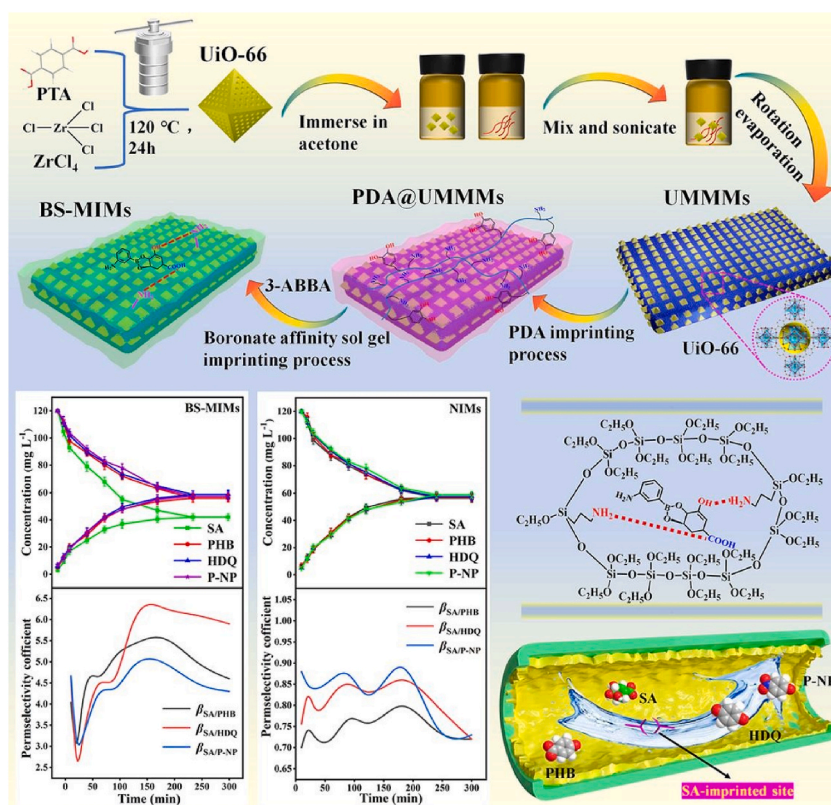

Fig. 6. Illustrates that membrane-based separation reduces environmental impact and energy consumption. The developed molecularly imprinted nanocomposite membranes, combined with the boronate-affinity sol-gel imprinting technique, demonstrate high selectivity and adsorption capacity for shikimic acid (reproduced with permission) [47].

and imprinting reactions were performed using a mixed solution kept at 55 °C for 6 h. The membrane was washed with NaOH/NaCl solution, then ethanol, and finally dried. The following product was obtained: IIP-PVI/CM. A control non-imprinted membrane (NIP-PVI/CM) was also synthesized by the same procedure, with the exception of the Mo(VI) template [37]. Yin et al. utilized a new two-step imprinting approach, which was surface grafting-oriented, to design immunoglobulin G (IgG)-targeted molecularly imprinted polymer. The process started with track-etched polyethylene terephthalate (PET) membranes with aliphatic C–Br initiator groups on their pore surfaces, allowing the generation of a functional polymer scaffold via surface-initiated atom transfer radical polymerization (SI-ATRP). The scaffold was then employed for the attachment of the IgG template. The UV-initiated crosslinking copolymerization of acrylamide and methylene bisacrylamide (MBAA) obtained by grafted MIP directly to the surface resulted in layer growth of the MIP hydrogel. This grafted MIP process was optimized for the scaffold chain length, the cross-linking degree, the content of MBAA used, and the irradiation time as a function of the layer thickness. In this way, a significant increase was achieved for the selectively bound amount of IgG and its purity when extracted from HSA mixtures, thereby proving the approach to be an extension of molecular imprinting to larger bionanoparticles and to a class of other base membranes that have a higher specific surface area [127]. Xing et al. developed a technique based on the chemical functionalization by anchoring functional monomers on the surfaces of nanofibers with a high level of hierarchical porosity (Fig. 7), allowing one to reach molecular selectivity and high selectivity in targeting pollutants such as atrazine [129].

3.5. Layer-by-layer assembly method

The layer-by-layer assembly of MIP-CMs is a technique that comprises the step-by-step deposition of several layers of polymers with opposite charges [130]. These layers include functional monomers and template molecules in specific regions. The introduction of each layer is accomplished using methods such as dipping, spraying, or spin-coating, which enable fine adjustment of the membrane's thickness and composition. After reaching the required number of layers, the template molecules are removed, which results in the formation of extremely precise binding sites inside the multilayered structure [48,131]. The advantages of layer-by-layer assembly lie in its simplicity, adaptability, and the capacity to precisely adjust the parameters of the membrane, such as its thickness, porosity, and distribution of functional groups. This optimizes the selectivity and performance of the MIP membrane. Moreover, this technique enables the integration of several functional elements and can be performed under mild environments. Yet, the primary drawback is the time-consuming aspect of the procedure, since each layer needs individual integration and drying, rendering it less appropriate for large-scale production. In addition, the removal of template molecules may occasionally be inadequate which might potentially impact the effectiveness of the membrane. Furthermore, the mechanical stability of the multilayered structure may be weaker in comparison to alternative synthesis methods [71,132–134]. For instance, Wang and his colleagues designed a new molecular imprinted adsorbent with rapid magnetic property for the selective extraction of adenosine 5′-monophosphate (AMP). This adsorbent was prepared by the attachment of AcrU and AM as functional monomers onto Fe_3O_4 nanoparticles in the presence of AMP as a template with the help of glutaraldehyde cross-linking. The Fe_3O_4@MIPs-LBL2, that has two layers of MIPs, had a better performance with imprint factor of 2.50 and high adsorption capacity is 105.72 $\mu mol\ g^{-1}$, which allows for easy AMP extraction and separation from complex samples like human urine while also enabling easy recovery due to the material's magnetism [130]. Wang et al. developed a Cu-IIM for the selective separation of copper ions through layer-by-layer assembly (Fig. 8). The polyethersulfone substrates were modified and added to multi-layers of the ion-imprinted material by electrostatic interactions. The template ions were removed by photo-crosslinking, using 4,4′-diazostilbene-2,2′-disulfonic acid disodium salt, through an acetic acid solution; this technique provided a way to control the
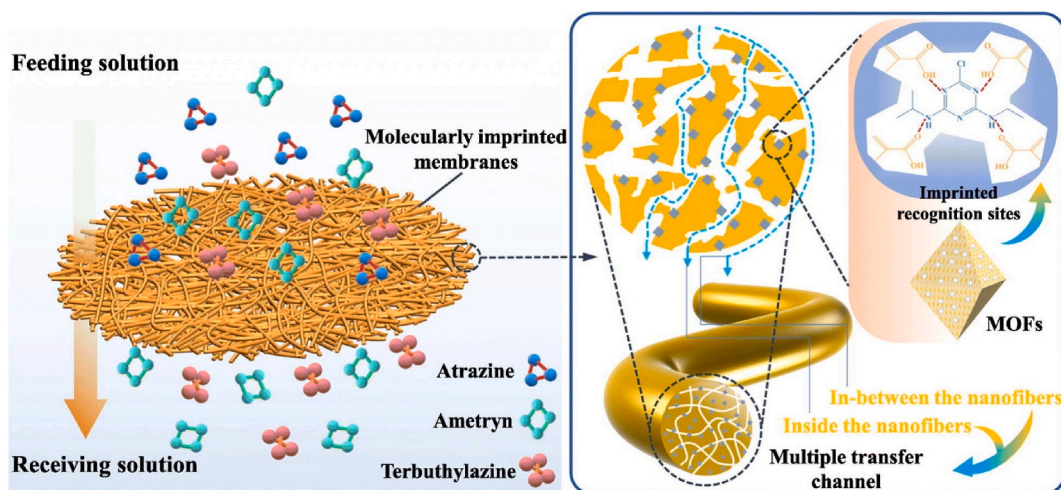

Fig. 7. Shows that the hierarchically porous MOF-based molecularly imprinted nanofiber membranes (HP-MINMs) efficiently enhance selective atrazine (ATZ) removal through multiple mass transfer channels, achieving superior permeation flux and permselectivity compared to traditional MIMs (reproduced with permission) [129].
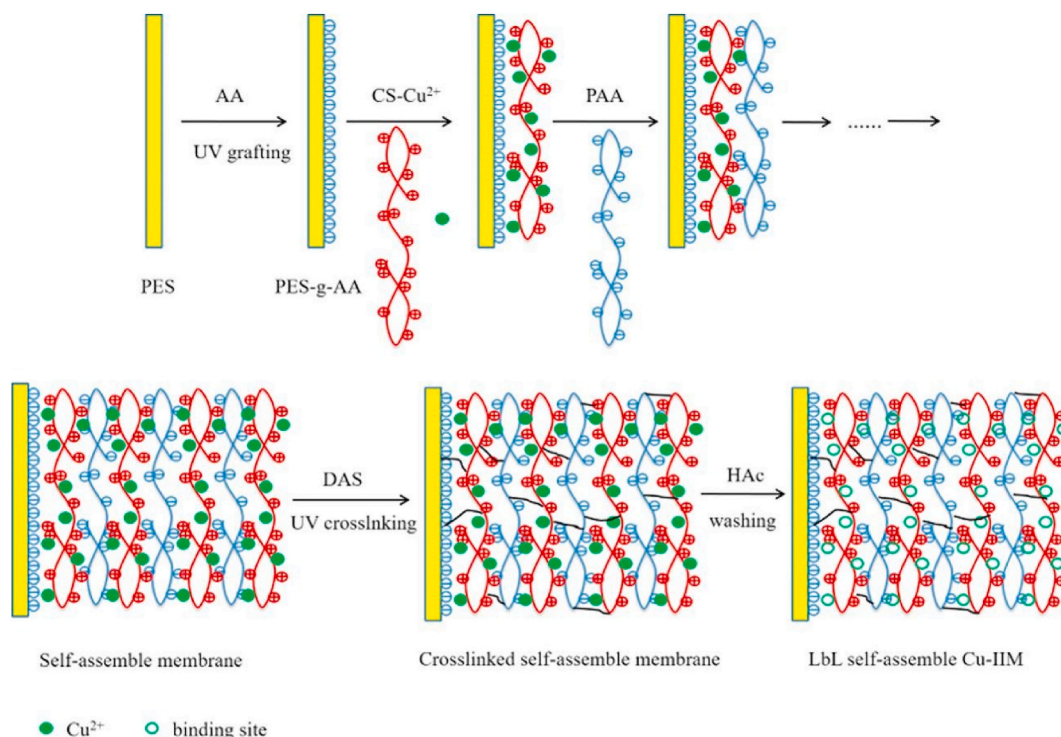
Fig. 8. Shows layer by layer self-assembled surface copper ion-imprinted membrane (Cu-IIM) demonstrates fast adsorption kinetics, high capacity, and selectivity for Cu^{2+} detection (reproduced with permission) [135].

membrane characteristics quite precisely, such as the number of assembly layers, which has strong effects on the adsorption capacity and selectivity of the membrane [135]. Xu et al. developed a molecularly imprinted sensor using a screen-printed carbon electrode that was modified with graphene and gold nanoparticles. In the first step, the electrode was patterned, and after that, layers of conductive inks were used and defined the working, reference, and counter electrodes. After the pretreatment of the electrode using PBS, the surface was modified with glutaraldehyde and then amino-functionalized graphene and gold nanoparticles with electrochemical reduction. In the next step, for the development of selective recognition capability, the surface was modified with electropolymerized MIP developed with 2,4,6-trichlorophenol and ortho-phenylenediamine by direct deposition on the modified electrode. The imprinting process was done by running cyclic voltammetry, which created a polymer membrane from the electropolymerization process that was specifically imprinted with the target molecule, and the electrically-driven deposition also washed out the target molecule; the specific binding sites were left. A similar NIP electrode was also made but in the absence of the target molecule, so the device was more capable of correctly identifying specific analytes [136].

3.6. Biopolymers polymerization technique

Although indeed MIP-CMs prove enormously useful for a plethora of applications, their non-biodegradability serves as a limiting factor. Contrarily, their polarity has resulted in the development of microbial polymers based on-not restricted to BC, cellulose, starch, alginate, carrageenan, and hyaluronic acid in the making of MIP-CMs. They are derived from novel composites have been discovered to possess unique nanostructures, high biocompatibility, crystallinity, biodegradability, and water holding capacity. For instance, Pavaloiu et al. designed both these mono- and multilayered composite membranes based on PVC, chitosan, and BC of maintaining the release of Ibu sodium. Dima et al. designed MIMs based on BC nanofibers, incorporating these with the natural biodegradable polymer xanthan to increase efficiency in controlled drug release and selective separation. In this regard, Liu et al. developed a chlorogenic acid molecularly imprinted chitosan membrane, showing high affinity sites reaching a high adsorption capacity. Similarly, Xu et al. designed MIMs based upon biomass bacterial cellulose, which has a film-like shape composed of three-dimensional hollow nano fibrous structures with high surface hydrophilicity; such kind of membrane shows high adsorption capacity for o-cresol, m-cresol, and p-cresol and fast adsorption kinetics [49,137,138].

4. Applications of MIP-CMs

MIP-CMs are widely used in several fields owing to their exceptional selectivity and specificity towards certain molecules. They are widely utilized in separation and purification treatments, where they selectively separate certain substances from complex mixtures,

such as in water treatment, environmental monitoring, and pharmaceutical purification [38,49,139]. MIP membranes are utilized as recognition components in chemical and biosensors within the field of sensor technology. They offer exceptional sensitivity and selectivity for the detection of pollutants, hazardous substances, and biomarkers [140–142]. Within the field of drug delivery, these membranes can be engineered to release therapeutic substances in a controlled manner, with the aim of selectively affecting particular cells or tissues [139,143]. This approach enhances the efficiency of the treatment while minimizing any adverse reactions. In addition, MIP membranes are employed in catalysis, functioning as catalysts or catalyst supports with exceptional selectivity for certain processes [144,145]. Their durability, resistance to chemical changes, and capacity to operate in challenging environments enhance their value in a wide range of industrial, environmental, and biomedical areas. Various applications of MIP-CMs are shown in Fig. 9. A brief overview of the various kinds of MIP-CMs prepared for different functions is provided below.

4.1. MIP-CMs for separation

Since the separation process is directly responsible for determining both the quality and the cost of goods, it is the most important component in the chemical industry [146,147]. The following are the approaches that are often utilized for separation: distillation systems [148], extraction procedures [130], adsorption based separation processes [149,150], crystallization techniques [151], chromatographic approaches [152], and membrane based separation methods [153]. Membrane separation technologies are environmentally friendly as they use low energy and produce manners waste relative to distillation. These work at lower pressure and temperatures, consume less energy and many of them run 24 h a day making the processes even more efficient. Membranes are selective and more efficient because it means the usage of less chemical and generation of less wastes [154–156]. It is essential in water treatment and its reuse hence reducing water consumption and reduction of greenhouse gas emissions. Furthermore, it is possible to recover valuable materials in the process, integrate membrane systems with renewable energy, and due to the size and modularity of the equipment, its overall environmental impact is small [55,157]. Following the tests conducted by Abble Nollet in the year 1748, researchers have made significant advancements in the construction of artificial membranes, which has led to the development of a variety of membrane technologies. Membrane-separation material functions as a kind of filtering media that enables separation at the molecular level. For the purpose of achieving selectivity, it is necessary to pick a membrane that has pores of an adequate size. Membranes are similar to separating screens. Through the integration of molecular imprinting with membrane-separation technology, MIPs membranes have been able to accomplish important improvements in the areas of molecular-specific identification, bio-macromolecule separations, and chiral-compound separations [157].

Bakhshizadeh et al. designed a new membrane made of polyester fabric with a coating of polyvinylidene fluoride and a MOF molecularly imprinted polymer for 3-aminophenol. To increase the hydrophilicity of the membrane sodium dodecyl sulfate was incorporated into the membrane. Al doped via a phase inversion technique; this membrane performs well in the selective deposition of Trypan blue dye with a removal rate of up to 94 %. System parameters like pH, dye concentration and pressure were tuned to improve the filtration efficiency which points to the possibility of the membrane's application in removal of dye from water [158]. Chai et al. created a molecularly imprinted membrane based on tannic acid and polyimide for the removal of bisphenol A from water. This biodegradable membrane also has antibacterial properties and this is because there is reduction of silver nitrate by tannic acid. The contact angle of the optimized membrane is reduced and the adsorption capacity is 33. The adsorption capacity was determined to be
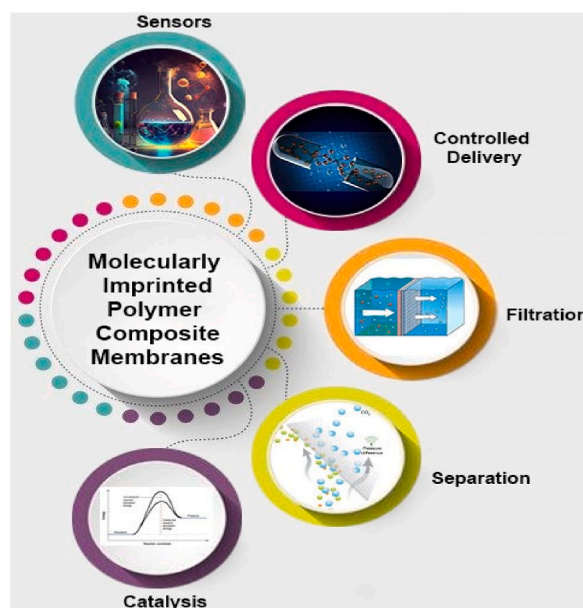
Fig. 9. Schematic for the applications of MIP-CMs.

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41 mg g^{-1} for BPA, the material was stable over six usage cycles with negligible adsorption loss. Such innovation presents a possibility of being used in many environmental and industrial settings for selective removal and purification of contaminants [159]. Zheng et al. synthesized a new UiO-66@Nylon based MIPs@UiO-66@Nylon for the selective extraction of SAs which are a class of broad-spectrum antibacterial agents. This new membrane with tannic acid and UiO-66-NH$_2$ on the nylon substrate has high hydrophilicity and good adsorption performance. The membrane has a good rate of recovery which is established to be between 83.2 and 98 %. The response of the developed PLSR model was very stable and provided 4 % over multiple cycles with minimal adsorption loss, which indicated that it was highly efficient and long-lasting for SA detection in areas concerning human health and environmental protection [160]. Wu et al. designed a novel boronic acid functionalized sol–gel basswood-based molecular imprinting membrane (BSISM) via top-down approach for the efficient adsorption and separation of shikimic acid. Basswood pores with in-situ grown MOFs and nanofluidic layers with molecular imprinting increase the adsorption capacity and selectivity of the membrane. It solves the problem of the tradeoff between membrane flux and selectivity and provides ultra-high-water permeability and environmental friendliness. This development offers great promise for use in the selective separation in the chemical industry, environmental science and biomedicine [161]. In a recent study, MIMs with excellent performance were synthesized for selective adsorption of ATSN, using a UiO-66@PDA shell with SiO$_2$ nanoparticles. This design incorporated polydopamine-coated MOF and SiO$_2$ as the support material improving the stability of the membrane and increasing the density of functional groups required for the binding of ATSN. The k-SUPCP-IMs membranes were synthesized by click chemistry and showed high selectivity and rebinding to ATSN, therefore, they have promising future in detecting and separating various biomolecules in biomedical and environmental fields [162]. A new type of dual selective imprinted membrane
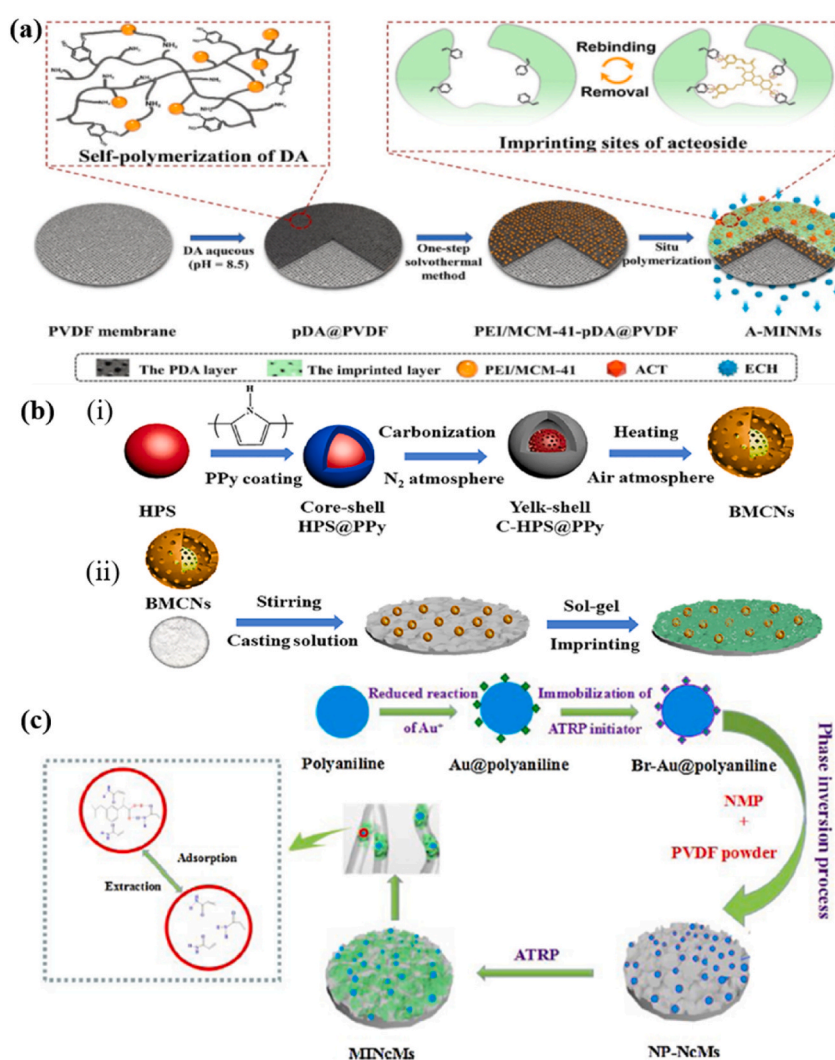
Fig. 10. (a) Schematic diagram of Multilayer-functionalized molecularly imprinted nanocomposite membranes for efficient acteoside separation (reproduced with permission) [169], (b) Creating a bilayer-mesoporous structure in molecularly imprinted nanocomposite membranes to enhance the efficiency of acteoside separation (reproduced with permission) [170], (c) Schematic illustration for the synthesis pathway of molecularly imprinted nanocomposite membranes modified by Au@polyaniline (reproduced with permission) [171].

12

has been prepared for the adsorption of pollutants such as Tetrabromobisphenol A and hexavalent chromium ion (Cr^{6+}). This membrane has a special surface that has different specific sites for each of the pollutants and is created through delayed phase inversion and surface modification. The DSIM shows very high adsorption efficiency and selectivity coefficients for TBBPA and Cr6+ and is superior to most of the existing materials. It also features very efficient and fast separation in both monocomponent and multicomponent mixtures; this has been confirmed by Density Functional Theory to be due to specific interactions at the recognition sites [163].

Ashrafian et al. developed a novel composite membrane embedded with MIP porous structures for the phenol removal. Polymeric nanospheres with a mean diameter of 210–250 nm and an average pore diameter of 8 nm were added to the dope solution. The HMIP-2 membrane with 10 % MIP showed excellent molecular recognition for phenol, with a 3.5 times greater selective recognition compared to catechol. The maximal separation factor of phenol is was found to be 2.19 relative catechol [164]. A new multilayered molecularly imprinted composite membrane was developed using carbon nanosphere and polydopamine to increase phenol selective adsorption. The membrane has a porous structure with numerous hydroxyl and amino groups that increase its hydrophilicity and imprinting activity, promoting mass transfer and adsorption. The m-MIM has high absorption capacity and permselectivity coefficients due to its many imprinted cavities. It can be used for water treatment and selective separation [165]. Cholesterol-imprinted PHEMA-MTrp membrane particles were tested for adsorption using methanol or intestinal-mimicking solutions. The imprinted membranes were highly selective for cholesterol compared to stigmasterol and estradiol, and had no adsorption capability of 23.43 mg g^{-1} at 2 mg mL^{-1} cholesterol concentration. The MIP particle-embedded composite membranes retained their cholesterol adsorption ability even after 10 cycles [166].

According to Wu et al. a bio-adhesive platform technique has been developed for thermally responsive molecularly imprinted composite membranes (TMICMs) with good temperature-dependent switching modes for selective recognition and separation of specific molecules. The method involves a two-step surface manufacturing process inspired by mussels using a self-polymerized polydopamine coating and constructing TMICMs in situ photoinitiated ATRP using the thermo-responsive backbone monomer N-isopropylacrylamide (NIPAm). TMICMs have potential applications in fluid management, thermo-responsive sensors, membrane-based medication delivery, and separation [167]. A new method using melamine-imprinted nanospheres on a cellulose acetate membrane is developed for detecting melamine in dry milk. The method is found to have better binding than nonimprinted membranes and has a linear calibration curve with limits of 0.007 and 0.020 μg/mL for detection and quantification, respectively. Melamine recoveries were 88.7–94.8 % [168].

Zhang et al. [169] described the multilayer-functionalized molecularly imprinted nanocomposite membranes (A-MINMs) with a schematic diagram as shown in Fig. 10a for the efficient separation of acteoside (ACT). These membranes consist of a base layer of PDA for mechanical support and adhesion, a middle layer of PEI functionalized mesoporous silica (PEI/MCM-41) for controlling pore size and providing amino groups for imprinting, and an outer layer that is ACT imprinted. From the results presented in the paper, A-MINMs exhibit a high rebinding capacity of 108. 74 mg g^{-1} and permselectivity with the value of βACT/ECH of 9. These figures are 43 indicating the possibilities of using them for natural product separation. Similarly, Chen et al. [170] demostrated a schematic representation for the bilayer mesoporous structure of molecularly imprinted nanocomposite membranes (A-MINMs) that are applied in the efficient extraction of acteoside (ACT) from Cistanche tubulosa as shown in Fig. 10b. These membranes are prepared by immobilizing bilayer-mesoporous carbon nanospheres (BMCNs) on polyvinylidene fluoride (PVDF) with phase inversion method. Consequently, the ACT-imprinted layer is developed on these membranes through the sol–gel process. Thus, the A-MINMs-400 obtained exhibit high performance with a rebinding capacity of 114.94 mgg^{-1}, and a permselectivity of 7. It has a turns of 15 and a selectivity of 4.48 for ACT. This optimization is credited to the one-of-a-kind bilayer mesoporous character of BMCNs, which allows for better passage and larger regions for ACT molecules, thus increasing the rebinding capabilities as well as selectivity. The incorporation of BMCNs as nanofillers also creates a large number of imprinted sites to enhance the membrane's preference for ACT. This new method has great possibility for the isolation of bioactive compounds from natural sources.

Wu et al. [171] describe the synthesis pathway for MINcMs with the Au@polyaniline nanostructure in Fig. 10c. This new synthesis method greatly improves the adsorption capacity, perm-selectivity of the membranes, and their ability to be regenerated. The prepared MINcMs possess high ibuprofen adsorption capacity where the highest adsorption capacity was 22.02 mg g^{-1}, which is four times higher than that of the non-imprinted nanocomposite membranes (NINcMs). Also, the selectivity factor of ibuprofen in MINcMs is 4. This was at 67 with a perm-selectivity factor (β) of 8. Specifically, the results of the present study for the 74th section confirmed their good selectivity in separation processes. This new strategy not only contributes to the creation of new molecular imprinting membrane materials but also provides a rational and efficient method for the selective separation of ibuprofen.

4.2. MIP-CMs as sensing material

One of the key components within many sectors are sensors used to identify the presence of chemical and biological substances, from clinical diagnostics, to environmental surveillance, to food safety assessments [172–174]. They share the characteristics of high specificity, sensitivity, and responsiveness. In general, a sensor consists of recognition and transduction elements, the former being biological. This part can be either biocatalytic, accelerating enzymatic processes, or affinity-based, coordinating selective binding [50, 51,175]. Chemical and biosensors produce a signal when analyte binds to the recognition element. A transducer converts this signal into a quantitative output. MIPs can replace biomolecules. General characteristics or physical changes can identify analytes. Reporter groups can improve sensor response and unique characteristics like fluorescence or electrochemical activity can be employed for detection [176,177].

A novel potentiometric sensor was designed for the detection of lindane, using a modified –HCCH imprinted polymer film on Cu

electrode and glycidyl methacrylate-grafted multi-walled carbon nanotube. The sensor has high selectivity and sensitivity, with a detection limit of 1 × 10-10 M, responding to –HCCH in the range of 1×10^{-1} to 1×10^{-3} M [178]. Silica gel was grafted with imprinted polymers using atom-transfer radical polymerization, resulting in MIP-SG with higher binding capacity and mass-transfer efficiency than MIPs synthesized conventionally. High-performance liquid chromatography column packed with MIP-SG showed improved column efficiency and resolution. The selectivity order for Boc-L-Trp and Boc-D-Trp depended on the template molecule used during MIP-SG synthesis [179]. New sensors were developed to evaluate dopamine and detect phloxine B in coffee, using graphene-chitosan MIPs and GO-MISPE, respectively [180]. A new technique for detecting phloxine B in coffee beans has been developed using molecularly imprinted polymers (MIPs) loaded graphene oxide (GO) solid-phase extraction (GO-MISPE) with high-efficiency liquid chromatography and laser-induced fluorescence detection. It accurately identifies phloxine B in coffee beans with high recovery rates and low detection limit [181].

Gold nanoparticles on a thin gold film form a sensitive and selective sensor for atrazine detection. The sensor detects 5pM atrazine in acetonitrile using an atrazine-imprinted polymer as a synthetic receptor [182]. Some other hybrid imprinted composite membranes were developed that efficiently capture BPA and improved separation capabilities for BPA and HPA using the hybrid membranes [183]. Cellulose/silica composites were used to produce a molecularly imprinted paper for caffeine. The MIP displayed improved binding for caffeine and good selectivity towards theophylline. A cellulose:silica ratio of 4:2 was found to be optimal [184]. A molecularly imprinted electrochemical sensor was developed by Wenjing et al. to detect chlortetracycline. The sensor was constructed on a gold electrode decorated with CD-MWCNTs, Au-PAMAM, and CSDT. The sensor showed high selectivity and stability towards CTC and had a linear range of 9.00×10^{-8} to 5.00×10^{-5} mol L^{-1} with a LOD of 4.954×10^{-8} mol L^{-1}. The sensor's quantitative analysis was validated by real sample analysis [185].

A novel electrochemical sensor made of MIP, nitrogen doped graphene nanoribbons (NGNRs), and ionic liquid (IL) was used to measure 4-Nonyl-phenol. The MIP/NGNRs-IL/GCE sensor performed well, with the peak current linearly related to NP concentration and a detection limit of 8 nM. The sensor provided excellent results when used to measure NP in actual samples [185]. AuNPs@-SiO$_2$-MIPs composite was used to build an electrochemical sensor for dopamine measurement. The sensor shows high selectivity towards DA and a broad linear your range of detection. Its successfully detected DA in human urine sample and dopamine hydrochloric injection [186]. A pH sensitive MIP nanospheres/hydrogel composite with controlled-release DXP has been developed for implantable biosensors biocompatibility enhancement. An electrochemical sensor based on MIP film has been prepared for detecting aspirin, which shown a detection limit of nmolL^{-1} [187]. A new electrochemical sensor that uses MIP film has been developed to detect aspirin. The sensor comprises a conductive hybrid membrane that imprints acetylsalicylic acid. The MIP polymer electrode offers high sensitivity, selectivity and reproducibility, with a detection limit of 0.3 nmol L^{-1} [188].

A glucose biosensor was prepared using molecularly imprinted polymers. The sensitive layer was produced by electropolymerizing O-phenylenediamine on a gold electrode with glucose as the template. The modified electrode surface was washed with distilled water to remove the template molecules. The biosensor showed a stronger response to glucose than to ascorbic acid or fructose. Investigations were done on the stability and reusability of the biosensor [189]. A colorimetric test was developed to detect produce inine in aqueous samples. It uses composite MIP-CMs as synthetic receptor sites for produce inine recognition. The produce inine concentration is determined by selective adsorption in the composite MIP-CM, followed by a color reaction. The test is easy to use, affordable, and has a 0.25 mM detection limit with a 0.25–2.5 mM linear dynamic range. It has at least one-year storage stability at room temperature [190].

Bompart et al. developed a submicrometer-sized sensor particle with a composite core-shell structure coated with a thin MIP shell. Gold colloids serve as antennas and detect bound target molecules, while the MIP shell provides analyte selectivity and inhibits signal interference. The manufacturing method for polymer-gold composite particles by Prasad and coworkers was modified to surround the polymeric core particle before adding the MIP (S)-propranolol shell [191]. A new electrochemical sensor has been developed for detecting tetracycline in honey. The sensor is based on a MIP microporous-metal-organic framework-based gold electrode surface and has a linear range of 224 fM to 22.4 nM, with a limit of detection of 0.22 fM. Tetracycline recoveries using this method range from 101.8 % to 106.0 %, with an RSD of less than 8.3 % [192]. An imprinted electrochemical sensor based on PPy-SG/HA-MWCNTs was used to detect tryptamine with high sensitivity. MIPs were produced by electropolymerizing tryptamine with pABA. The sensor demonstrated high selectivity and detected tryptamine in a linear range of 9.0×10^{-8} mol L^{-1} to 7.0×10^{-5} mol L^{-1} under optimum parameters, with a detection limit of 7.41×10^{-8} mol L^{-1}. Real samples were successfully detected using this imprinted electrochemical sensor [193]. An electropolymerized MIP integrated with single-wall carbon nanotubes accurately measured brucine in human serum. The imprinted sensor had higher recognition ability for brucine than non-imprinted polymer. The sensor detected brucine in human serum samples with recoveries ranging from 99.5 to 103.2 %. The imprinted sensor's current response was linear to brucine concentrations between 6.2×10^7 and 1.2×10^5 M under ideal experimental circumstances, with a detection limit of 2.1107 M [194].

An electrochemical sensor using chitosan-platinum and graphene-gold nanoparticles was prepared to measure erythromycin quickly and accurately. It has a linear range of 7.0×10^{-8} mol L^{-1} to 9.0×10^{-5} mol L^{-1} with a detection limit of 2.3×10^{-8} mol L^{-1} (S/N = 3). The sensor is highly selective, stable, and repeatable in actual spiked samples [195]. A new electrochemical sensor for neomycin detection was designed using chitosan-silver nanoparticles/graphene-multiwalled carbon nanotubes composites decorated gold electrode and electropolymerization process to synthesize molecularly imprinted polymers. The linear range was 9×10^{-9} mol L^{-1} to 7×10^{-6} mol L^{-1} with LOD of 7.63×10^{-9} mol L^{-1}. It exhibited high binding affinity, good reproducibility, and stability. The sensor was successfully used to detect neomycin in milk and honey samples with satisfactory recovery [196]. Molecularly imprinted nanoparticles were electrospun into polymer nanofibers to produce a novel molecular recognition material. The nanofibers contained theophylline and 17â-estradiol imprinted nanoparticles and were made of poly(ethylene terephthalate). The composite nanofibers had

a defined shape and were selective in target identification. This electrospinning method has advantages and offers new possibilities for using both electrospun nanofibers and molecularly imprinted nanoparticles [197]. A molecularly imprinted electrochemical sensor for glutathione detection was prepared using Fe_3O_4@PANI/rGO nanocomposites via magnetic field directed self-assembly. The ternary nanocomposites offered functionalized sites that could form hydrogen bonds and electrostatic interactions with GSH in the matrix, as well as a network that facilitated electron transfer. The tunable sensor was highly reproducible and stable, with a detection limit of 3 nmol L^{-1}. Additionally, the sensor was effective in the clinical detection of GSH in biological samples [198].

MIP-CMs were prepared using photopolymerization with tri(ethylene glycol) dimethacrylate as a cross-linker, methacrylic acid as a functional monomer and atrazine as a template. The imprinted membranes were tested as a recognition component of an atrazine-sensitive conductomeric sensor, which showed high selectivity and sensitivity with a detection limit of 5 nM. The sensors reaction time varied from 6 to 15 min depending on membrane thickness [199]. A fluorescence switch sensor was developed using a dye-doped MIP and silver nanofilm amplification to detect the fungicide fenaminosulf (FM). The new method for FM detection had a detection limit of $1.6 \times 10\text{-}11$ mol L^{-1} and was used to quantify residual FM in vegetables with recoveries ranging from 92.0 % to 110 % [200]. Fe_3O_4@C-based thermal-responsive and magnetic molecularly imprinted polymers (TMMIPs) bind and release 2,4,5-TCP from water. TMMIPs are stable, have multilayer structure, and thermo-responsive properties. Selective adsorption and controlled release of 2,4,5-TCP were examined at 25 °C, 45 °C, and 60 °C [201].

An electrochemical sensor was developed to detect herbicide simazine using MIP-CMs and o-aminothiophenol functionalized gold nanoparticles. The sensor has high electrocatalytic activity and quick rebinding kinetics for simazine reduction. It has a linear dependence on simazine concentrations ranging from 0.03 to 140 M, with a detection limit of 0.013 M. The suggested approach was successfully employed to assess simazine in real-world samples with good potential for practical application [40]. A microfluidic paper-based colorimetric sensor was developed using $ZnFe_2O_4$ magnetic nanoparticles and MIP-CMs. The sensor showed high reproducibility, selectivity, and regeneration, and is ideal for portable detection of Bisphenol A in environmental monitoring, security inspection, and complex matrices [142]. A Study conducted by Fu et al. used a molecularly imprinted electrochemical sensor detect determine selectively in complicated criminal samples. The imprinted sensor showed good sensitivity, selectivity and long-term
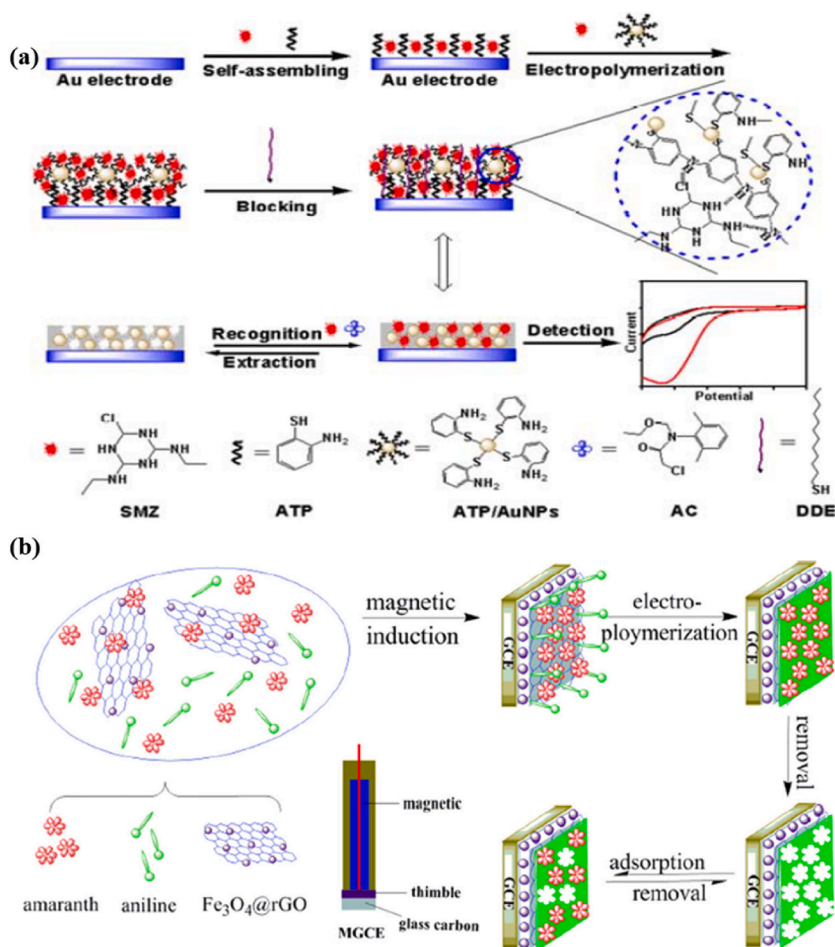

Fig. 11. (a) Schematic representation for the construction process of herbicide simazine imprinted sensor (reproduced with permission) [40]. (b) Schematic representation for the preparation of Fe_3O_4@rGO doped MIP-CM (reproduced with permission) [207].

stability. It has a low detection limit and successfully measured ketamine in urine and saliva samples [202]. A new electrochemical sensor was developed by Wang et al. for measuring tinidazol using a carbon paste electrode modified with multi-walled carbon nanotube and boron-embedded MIP-CMs. The sensor showed high sensitivity and selectivity with a reduced detection limit and excellent accuracy when tested with pharmaceutical and biological materials [203].

Fang et al. developed a novel 3D MIP quart crystal microbalance sensor for detecting trace levels of Citrinin. The sensor utilized a composite modified Au electrode surface and showed a linear response to Citrinin concentrations over a wide range. It achieved a low detection limit and exhibited excellent selectivity, anti-interference capability, reproducibility, and long-term stability. This holds promise for detecting trace Citrinin in food samples [204]. MIP-CMs were prepared using ECB separated from Euphorbia fischeriana as template, acrylamide as a functional monomer, and ethylene glycol dimethacrylate as a cross-linker. The membranes selectively identified and transported ECB with a maximum adsorption quantity of 3.39 µmolcm^{-2} and 38.71 % ECB permeation. The membranes were effective for selectively separating ECB from Euphorbia fischeriana [205]. Fan et al. synthesized a novel MIP composite membrane i.e. NH$_2$-POSS-pDA@PVDF membranes ACT-imprinted layers in A-MICMs for ACT adsorption. A-MICMs showed a high selectivity of 4.63 and a maximum rebinding capacity of 98.37 mg g^{-1}. The interleaved imprinted network structure of A-MICMs can be used to selectively separate bioactive components [206].

Zhang et al. illustrates Schematic representation for the construction process of SMZ imprinted sensor shown in Fig. 11a and b.

4.3. MIP-CMs in catalysis

Catalysis appears as one of the most valuable tools in industry and a powerful tool for environmental protection as well, as it significantly enhances the effectiveness of chemical transformations, and at the same time reduces energy consumption and the formation of pollutants [208,209]. MIP-CMs have been employed extensively in catalysis because they can provide active sites that are extremely specific and selective, specifically designed for certain processes. These membranes are designed with imprinted cavities that precisely match the form, size, and functional groups of the target molecules [210–212]. This allows for highly precise catalytic activities to take place. MIP membranes have the ability to function as both catalysts and supports for immobilizing catalytic species in catalytic applications. This dual role enhances the efficiency and selectivity of the reaction [213–215]. Due to their durability and
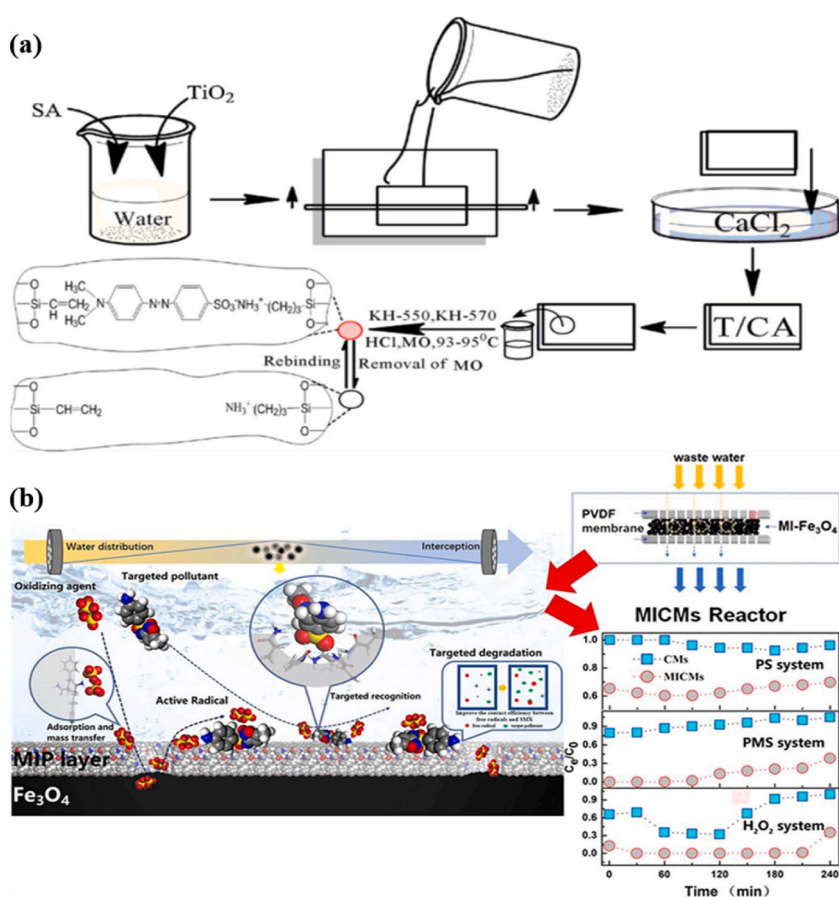

Fig. 12. (a) representation for the preparation of TiO2/Alginate membrane and MO MIP-CMs (reproduced with permission) [34], (b) MIP-CM for improved oxidation degradation efficiency of low concentration pollutants (reproduced with permission) [212].

chemical stability, MIPs may function effectively in harsh environments, which makes them ideal for use in industrial catalytic processes [216–219]. In addition, the simplicity of recovering and reusing MIP membranes enhances the sustainability and cost-efficiency of catalytic systems. Yet, achieving the uniform distribution and availability of active sites in MIP membranes can be challenging, and it is crucial to thoroughly remove template molecules to avoid contamination and maintain the best possible catalytic performance [213,220,221].

For instance, the effectiveness of TiO_2 photocatalysis in the removal of low-level organic pollutants may be improved by the use of MIP coating. The removal of the target pollutants 2-nitrophenol and 4-nitrophenol was accomplished with great activity and selectivity using photocatalysts made of TiO_2 that had been coated with MIP. One of the primary factors that contributed to the increased selectivity was the interaction between the target molecules and the MIP coating via the use of functional groups [222,223]. A novel mass spectrometric method was developed by Cheng-Tai et al., and they used a thin layer of TiO_2 sol-gel as the sample substrate. In order to conduct an investigation based on molecular recognition, they synthesized molecularly imprinted Titanium dioxide sol-gels. The modified glass slide has the ability to selectively extract r-CD from a sample solution, and it was immediately identified by mass spectrometry. This methodology introduces a novel detection method for analysis that is based on molecular recognition [224].

Zhao et al. synthesized a MIP-CM of TiO_2/calcium alginate using T/CA as the supporting matrix, KH-570 and KH-550 as functional monomers, and MO as the template. Their findings revealed that the MIP exhibited superior adsorption and faster degradation rates for MO compared to the NIP. Moreover, it displayed excellent selectively for the photodegradation of MO when competing with MR as a competitive molecule. The results highlight the potential of MIP-CMs for effective pollutant removal in water treatment application (Fig. 12a) [34]. Ulbricht et al. investigate the use of MIP-CM as catalysts for a dehydrofluorination process. Membrane reactors were used to test two distinct membrane types. The findings demonstrated that PVA membranes with imprinted sites displayed distinct catalytic effects when compared to PVA membranes with non-imprinted polymer [95].

A schematic representation of preparation is shown in Fig. 12b. Another group reported that the use of a molecularly imprinted catalytic membrane for treating water has dramatically increased its efficiency in oxidation and the removal of pollutants. The oxidation rate constants on the MICM are 3.5–33 times of the Fe_3O_4 in different systems and the observed rate constants are as high as $0.142 \ min^{-1}$. More than 90 % of sulfamethoxazole was removed within 150 min, which makes effective use of the targeted catalysis and oxidant adsorption in enhancing the reaction efficiency in non-free radical processes [212].

4.4. MIP-CMs for controlled release

Specific molecular recognition is an essential component of biological systems that support processes such as neural transmission, respiration, immunological defense, cellular differentiation, and food reliance. As a result, it is not surprising that scholars have dedicated a significant amount of time and effort to analyzing and, more recently, modelling biological function. Molecular imprinting is the most promising of the several synthetic approaches explored so far. Molecular imprinted polymers are widely used as dependable and effective synthetic molecular sensors in a variety of applications. However, the most promising future possibilities may be found in the area of medication delivery, particularly in intelligent drug release and magic bullet drug targeting [225–227]. MIP hybrid cellulose membranes were created by integrating MIP beads specific for S-Omeprazole in cellulose and were utilized for enantioselective control administration of S-Omeprazole [228]. As previously stated, cellulose and propranolol imprinted beads form a composite membrane. Suedee et al. created a reservoir-type transdermal enantioselective control administration device for racemic propranolol employing a MIP composite membrane. They created a MIP composite cellulose membrane by reactively filing a bacterial cellulose membrane with a MIP thin layer and a silanized coupler as an extra anchor for the MIP. These composite membranes were employed in an enantiomer-controlled releasing mechanism. Because of the microporous nature of the cellulose, they are unsuitable for UF, and the use of a costly technique makes them uneconomic. Furthermore, the separation ability was not adequate [229]. MIP-CMs show potential for creating synthetic molecular sensors and rugs delivery systems. However, addressing challenges like cost-effectiveness and improving separation abilities are crucial for maximizing their practical application.

In summary, molecular imprinting shows potential for creating synthetic molecular sensors and smart drug delivery systems. However, addressing challenges like cost-effectiveness and improving separation abilities is crucial for maximizing their practical application.

4.5. MIP-CMs in enhancing filtration dynamics

MIP-CMs exhibit remarkable selectivity and specificity for target molecules, making them particularly efficient in filtration applications. These membranes are designed with imprinted spots that precisely match the size, shape, and chemical properties of specific pollutants [157]. This enables them to selectively remove unwanted chemicals from complex mixtures. MIP membranes in water treatment have the ability to selectively eliminate contaminants, including heavy metals, drugs, and organic molecules, thereby improving water quality and safety [230,231]. Additionally, they are employed in the process of purifying pharmaceuticals and bioproducts, ensuring the elimination of contaminants and improving the overall purity of the final product. MIP membranes are highly durable and chemically stable, making them ideally suited for utilization in challenging environmental conditions and industrial applications [45,230]. Fabrication of these membranes can be intricate and time-consuming, and attaining a homogeneous distribution of imprinted sites is an enormous challenge. Possibility of improper removal of template molecules could affect filtration efficiency and selectivity. Although there are certain challenges, MIP composite membranes provide an outstanding and flexible solution for advanced filtration purposes [232–234]. For instance, Szekly et al. employed organic solvent nanofiltration followed by polishing step with MIPs to remove 1,3-diisopropylurea from the active pharmaceutical ingredients. This combination reduced 1,

3-diisopropylurea from 100 mgg^{-1} to 2 mg g^{-1} of pharmaceutical ingredients [235]. 4-vinyl-pyrididne and ethylene glycol dimethylacrylate MIP-CM showed enhanced S-Naproxen permeation (binding constant: 13.8) [236]. The "gate effect" is the change in MIP's morphology and solute diffusive permeability due to templates. Koji Hattori et al. used ultraviolet irradiation to graft a copolymer onto a cellulose membrane and studied the relationship between graft copolymer content, gate effect strength, and UV exposure duration. The irradiation time affected the variation caused by the template or analogue and the selectivity of the permeability. Synthesized MIP has a "living nature" and can regulate the gate effect through irradiation time, making it a promising technique for producing complex MIP membrane architecture with self-controllable permeability [94].

Membrane-based chemical separations are gaining popularity in research and industry. Selective composite membranes are desired to separate target molecules from feed solutions. Jae-Min et al. proposed a novel method of photopolymerizing a "molecularly-imprinted polymer film" on a microporous substrate to synthesize ultrathin film composite membranes. They used polymers imprinted with theophylline to build composite membranes, and analyzed their rate and selectivity for theophylline transport [81]. Niavarani et al. developed a plyethersulfone microfiltration membrane with MIP for selective adsorption of 17β-estradiol from water (maximum adsorption capacity 21.9 mg g^{-1}) which was found higher than commercial nano-filtration [100]. MINMs employed selective separation mechanisms categorized as retarded permeation and facilitated permeation. Fig. 13 shows the schematic illustration of permselectivity mechanism of A-MINMs. In A-MINMs, ECH molecules penetrated directly, while ACT molecules are intercepted. Abundant imprinted sites and cavities in A-MINMs capture ACT molecules, binding them securely through hydrogen bonding. This combination, known as retarded permeation facilitates the separation process [170]. A detailed list of various MIP-CMs along with their analytical applications is shown in Table 1.

5. Our group's breakthroughs in MIP-CMs

Our research team has contributed several breakthroughs in the field of MIP-CMs since 2010. We have made significant strides in developing innovative solutions for various applications, including chiral separation and selective adoptions. Our notable contribution includes the development of a novel MIP composite membrane for chiral separation of phenylalanine racemate solution [250–252]. The MIP-CM incorporated the MIP beads and MIP membranes where both showed their contribution in selectivity. It was very first study where two MIP formats were merged for racemate resolution. These membranes have demonstrated remarkable chiral separation capabilities, with high permselectivity and swelling ratio. Additionally, our exploration in adsorptive MIP-CMs has further advanced chiral separation techniques, with composite membrane displaying improved adsorption capacity and selectivity compared to control membrane [253–256]. Furthermore, our ongoing unpublished projects, undertaken by dedicated researchers, aim to push boundaries of MIP technology, addressing the emerging challenges and exploring new opportunities in the field. Similarly, we are in continuation of our journey and presently we reached to utilizing magnetic core-shell MIP beads that will be further incorporated in developing MIP-CMs. Our contribution in the field of MIPs is also reflected in the form of our published review papers [254,256]. Through our commitment to excellence and collaboration, we strive to continue contributing impactful research to scientific
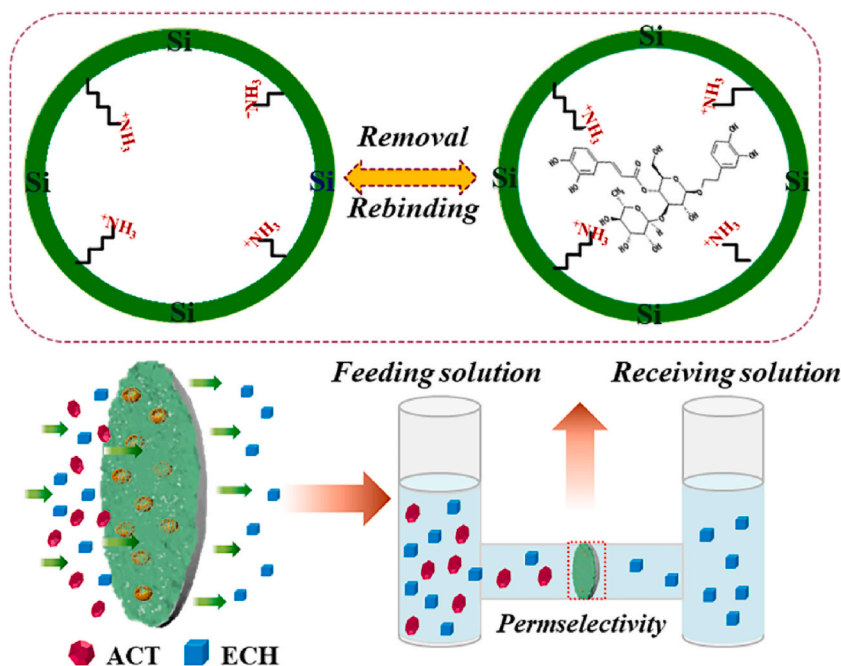

Fig. 13. Schematic illustration provides a clear visualization of the key mechanism responsible for the separation, emphasizing the process of retarded permeation (reproduced with permission) [170].

Table 1
Various MIP-CMs and their analytical performance.

MIP-CMs	Template	Separation by	Separation factor (Max.)	Adsorption capacity (Max.)		Adsorption selectivity (Max.)	Rejection selectivity (Aver.)	Ref.
				Template	Counter molecule			
CMLIBDIM	D-Phe	UF	α (D/L) = 2.99	1.92 mg g^{-1}	1.90 mg g^{-1}	αads(D/L) = 1.44	αrej(D/L) = 0.644	[237]
CMDIBLIM	D-Phe	UF	α (D/L) = 3.090	1.892	1.900	1.510	αrej(D/L) = 0.589	[237]
CMDIBDIM	L-Phe	UF	α (D/L) = 0.435	1.970	1.781	2.722	αrej(D/L) = 0.535	[237]
CMLIBLIM	L-Phe	UF	α (D/L) = 0.438	1.805	2.077	2.983	αrej(D/L) = 0.437	[237]
(MAA[a]/EGDMA[b]) MIP nanoparticles/PA[c] support	tert-Butoxycarb-onyl)-L-Phenylalani-neanilide(BFA)	Filtration	–	50.00 µmol g^{-1}	–	–	–	[238]
(MQN[d]/EGDMA) MIP beads/C[e]	S-Omeprazole	Diffusion	α (R/S) = 1.29	–	–	–	–	[239]
(MQD[f]/EGDMA) MIP beads/C	S-Omeprazole	Diffusion	α (R/S) = 1.18	–	–	–	–	[239]
(MAA/EGDMA) MIP microspheres/C	Template S-propranolol	Diffusion	α (S/R) = 1.33	–	–	–	–	[90]
	Structural analogues Cyclopropr-anoyl-propranolol		α (S/R) = 1.36	–	–	–	–	
	Valeryl-propranolol		α (S/R) = 1.27	–	–	–	–	
	Oxprenolol		α (S/R) = 1.16	–	–	–	–	
	Pindolol		α (S/R) = 1.16	–	–	–	–	
(MMA[g]/MAA) MIP nanospheres/ (MMA/AA[h]) membrane	Theophyline	Adsorption	–	0.95 mg g^{-1}	0.095 mg g^{-1}	13.51	–	[240]
(MMA/MAA) MIP nanoparticles/ (MMA/AA) membrane	Theophyline	Adsorption	–	5.2 µmol g^{-1}	–	10	–	[241]
	Caffeine	Adsorption	–	~0.57 µmol g^{-1}	–	3.75	–	
(BADM[i]/DVB[j]) MIP particles/PS[k]	Bisphenol A	Adsorption	–	38 µmol g^{-1}	–	–	–	[242]
(BADM/DVB) MIP particles/CA[l]	Bisphenol A	Adsorption	–	148 µmol g^{-1}	–	–	–	[242]
(BADM/DVB) MIP paricles/Ny[m]	Bisphenol A	Adsorption	–	142 µmol g^{-1}	–	–	–	[242]
(BADM/DVB) MIP paricles/PSf[n]	Bisphenol A	Adsorption	–	158 µmol g^{-1}	–	–	–	[242]
(α-TMA[O]/DVB) MIP powder/PSf	α-Tocopherol	Adsorption	–	70.2 µmol g^{-1}	–	–	–	[243]
(α-TMA/DVB) MIP powder/CA	α-Tocopherol	Adsorption	–	58.1 µmol g^{-1}	–	–	–	[243]
(α-TMA/DVB) MIP powder/Ny	α-Tocopherol	Adsorption	–	42.5	–	–	–	[243]
(MAA/MAM[p]) MIP/ commercial filter paper support	Curcumin (Cu)	Adsorption	–	234.99 µmol g^{-1}	167.22 µmol g^{-1}	αads(CuI/ CuII) = 1.50	–	[244]
(MAA/EGDMA) MIP ultrathin film/ alumina support	Theophiline	Filtration	α (Th/Cf) = 2.60	–	–	–	–	[245]
	Caffeine (Cf)	Filtration	α (Cf/Th) = 3.00	–	–	–	–	
(AMPS[t]/MBAA) MIP/ PP[u] support	Desmetryn	Filtration	–	12.10 nmolcm^{-2}	–	–	–	[246]
(AMPS/MBAA) MIP thin layer/PVDF[v]-phob support	Desmetryn	Filtration	–	10.20 µgcm^{-2}	–	–	–	[247]
(AMPS/MBAA) MIP thin layer/PVDF-phil support	Desmetryn	Filtration	–	8.6 µgcm^{-2}	–	–	–	[247]

Table 1 (*continued*)

MIP-CMs	Template	Separation by	Separation factor (Max.)	Adsorption capacity (Max.)		Adsorption selectivity (Max.)	Rejection selectivity (Aver.)	Ref.
				Template	Counter molecule			
(MAA/EDMA) MIP thin membrane/C support	*S*-propranolol	Diffusion	α (S/R) = 2.00	–	–	–	–	[248]
(γ-APTS^w/TEOS^x) MIP/ alumina support	Luteolin (Leu)	Diffusion	–	1.87 Mmolg^{-1}	0.21 Mmolg^{-1}	αads(Leu/Lu) = 14.12	–	[249]
(4-VPy^y/EGDMA) MIP/ PVDF support	*S*-Naproxen	Adsorption	–	1.90 μmolg^{-1}	–	–	–	[157]

community, there by underscoring the vast potential of MIP-CMs based technologies for various applications.

6. Challenges and future perspective

MIP-CMs may pose several potential challenges and offer some promise for future advances in the field. The most relevant difficulties and directions of possible breakthrough for these new membrane technologies are listed below:

- High selectivity often results in low flux, which can make practical throughput impossible to achieve for industries requiring high efficiency.
- The membranes need to be operated under various conditions, including exposure to extreme temperature, pH, and harsh chemicals.
- Many MIP-CMs do not fulfill the set criteria for high demand in long-term industrial applications.
- The molecular imprints on the membranes need to be an exact match with the target molecules to show efficient separation performance.
- Developments in scalable advanced material production, e.g. MOF, which are a new candidate but complex to produce with high consistency and at competitive costs.

Being motivation behind ongoing research and development, the practical applications of MIP-CMs are issues that have to be pushed notwithstanding these. Approaches with respect to the said challenges ought to be multidisciplinary and should relate to the state of knowledge in polymer science, material engineering, and, at times, computational modeling. It is only when these challenges are overcome that the full potential of MIP-CMs in these applications can become real.

In the future, the challenges to MIP-CMs development in technological and material innovation shall be thus realized.

- New polymer matrices and imprinting technologies are needed that will yield a proper balance between selectivity and permeability.
- Advances in membrane designs with specific binding sites and better mass transfer features are expected with innovations in nanotechnology and material science.
- The prediction and optimization of target molecule/imprinted site interactions can occur by the application of machine learning algorithms and computational models.
- The development of new membranes containing hybrid materials, organic and inorganic components, for strength and durability.
- Scalability and cost of production of MIP-CMs, including by electrospinning and layer-by-layer fabrication.
- The inclusion of materials such as MXenes, perovskites, quantum dots, photonic crystals etc. To increase the surface area and conductivity, respectively.
- Design of cost-effective use of materials and green chemistry principles for a minimal environmental footprint and enhanced sustainability.
- Developing protocols for ensuring that the final MIP-CMs produced at scale maintain the desired properties.
- Establish guidelines for MIP-CM production and use via academia-industry-regulatory bodies partnership.

7. Conclusion

In conclusion, this comprehensive review provides in-depth exploration of the diverse applications of MIP-CMs in sample preparation. The discussion encompasses five distinct methods for crafting MIP-CMs: coating membrane disc with MIP particle's encapsulating MIP Nanoparticles between membranes layers grafting MIP onto membrane pores via photoionization polymerization filling thin tracked etched membrane pores with MIP, and encapsulating MIP NPs in composite nanofibers membrane through electrospinning.These techniques serve a multitude of purpose including selective separation controlled release adsorption and purification of crucial molecule's with endeavors made towards enantioseparation.

Data availability statement

No data used for this article.

CRediT authorship contribution statement

Nasrullah Shah: Writing – review & editing, Supervision, Methodology, Investigation. **Muffarih Shah:** Writing – original draft, Validation, Investigation, Conceptualization. **Touseef Rehan:** Writing – original draft, Validation, Conceptualization. **Abbas Khan:** Writing – review & editing, Conceptualization. **Noor Majeed:** Writing – original draft, Validation, Investigation. **Abdul Hameed:** Writing – original draft, Validation, Investigation. **Mohamed Bououdina:** Writing – review & editing. **Rasha A. Abumousa:** Writing – review & editing, Conceptualization. **Muhammad Humayun:** Writing – review & editing, Writing – original draft, Validation, Funding acquisition, Conceptualization.

Declaration of competing interest

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

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