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Application of magnetic molecularly imprinted polymer as a versatile and highly selective tool in food and environmental analysis: Recent developments and trends

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ABSTRACT

Recently, combination of molecularly imprinted polymers (MIPs) with additional features, e.g. magnetic properties, has led to good results. Compared with conventional MIPs, magnetic molecularly imprinted polymers (MMIPs) have attracted considerable attentions attributing to the fact that they not only exhibit outstanding magnetism but also have specific selective binding for template molecules. However, use of MMIPs still faces challenges such as slow mass transfer, low binding capacity, and template leakage. In this article, we review several novel applications of MMIP-based approaches that were applied to food and environmental sample analysis and can overcome these problems. We discuss on brief descriptions of advantages/disadvantages and how these methods could improve the extraction and absorption process for a variety of analytes. Also, we compare each novel magnetic extraction technique with earlier studies in analytical parameters. Finally, some of the future MMIPs-based perspectives will be given.

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1. Introduction

Chemical contaminants such as pesticides, mycotoxins, veterinary drugs or other pollutants in food and environment pose a health concern. Therefore, one of the necessary parts of food and environmental safety testing programs is analysis of pertinent chemical contaminants to ensure consumer safety and compliance with set regulatory limits. Because of the large number of matrix

Abbreviations: 4-VP, 4-vinylpyridine; AHL, Gram-negative bacterial quorum signaling molecule; AM, Acrylamide; APS, Ammonium persulfate; APTES, 3-aminopropyltriethoxysilane; BET, Brumauer–Emmett–Teller; BPA, Bisphenol A; BT, Benzothiophene; CCD, Central composite design; CE, Capillary electrophoresis; CNT, Carbon nanotube; CPE, Cloud point extraction; CV, Cyclic voltammetry; DAD, Diode array detector; DBT, Dibenzothiophene; DES, Diethylstilbestrol; DIS, Dienestrol; DLLME, Dispersive liquid–liquid microextraction; DPV, Differential pulse voltammetry; DR-MIP, Dual-responsive MIP; DSMIP, Dummy-surface molecularly imprinted polymer; DSMMIP-GO, Double-sided MMIP modified graphene oxide; d-SPE, Dispersive solid phase extraction; ECL, Electrochemiluminescence; EDC, Endocrine disrupting chemical; EDX, Energy-dispersive X-ray spectroscopy; EGDMA, Ethylene glycol dimethacrylate; ELISA, Enzyme-linked immunosorbent assay; EMMIP, Electromagnetic molecularly imprinted polymer; ERY, Erythromycin; FAAS, Flame atomic absorption spectrometer; FI-CL, Flow-injection chemiluminescence; FLD, Fluorescence detection; GCE, Glassy carbon electrode; GC-MS/MS, Gas chromatography coupled with tandem mass spectrometry; GC-NPD, GC with Nitrogen Phosphorous Detector; GMA, Glycidilmethacrylate; GO, Graphene oxide; HES, Hexestrol; HMT, Hexamethylenetetramine; HPLC, High performance liquid chromatography; HPLC-MS, Liquid chromatography-mass spectrometry; HPMMIP, Hollow porous magnetic molecularly imprinted polymer; HQ, Hydroquinone; IARC, International agency for research on cancer; ILMBME, Ionic liquid magnetic bar microextraction; LC, Liquid chromatography; LLE, Liquid–liquid extraction; LMG, Leucomalachite green; LOD, Limit of detection; LOQ, Limit of quantification; MAA, Methacrylic acid; MCs, Microcystins; MDMINP, Magnetic dummy molecularly imprinted nanoparticle; MDMIP, Magnetic dummy molecularly imprinted polymer; MDSPME, Magnetic dispersive solid-phase microextraction; MG, Malachite green; MIP, Molecularly imprinted polymer; MIT, Molecular imprinting technology; MMINP-d-SPE, Magnetic molecularly imprinted nanoparticle with dispersive solid-phase extraction; MMIP, Magnetic molecularly imprinted polymer; MMISF, Magnetic molecularly imprinted sensing film; MMISPE, Magnetic molecularly imprinted solid phase extraction; MMMIP, Montmorillonite magnetic molecularly imprinted polymer; MNP, Magnetic nanoparticles; MRL, Maximum remnant limit; MUTDMIP, Magnetic ultra-thin dummy molecularly imprinted polymer; MWCNT, Multi-walled carbon nanotube; NIP, Non-imprinted polymer; PAE, Phthalate ester; PCA, Protocatechuic acid; PLE, Pressurized liquid extraction; PM-MIM, Photonic-magnetic responsive molecularly imprinted microsphere; PTOP, 4-tert-octylphenol; QuEChERS, Quick, easy, cheap, effective, rugged and safe; RAFT, Reversible addition fragmentation chain transfer; RAFTPP, Reversible addition fragmentation chain transfer precipitation polymerization; SBSE, Stir-bar sorptive extraction; SPE, Solid-phase extraction; SPME, Solid-phase microextraction; SR-MIP, Stimuli-responsive MIP; TBA, Tetra-n-butyl ammonium hydrogen sulfate; TEM, Transmission electron microscopy; TEOS, tetraethyl orthosilicate; TFA, Trifluoroacetic acid; TGA, Thermogravimetric analysis; TMIP, Thermosensitive MIP; TMMIP, Thermal-responsive and magnetic molecularly imprinted polymer; UHPLC, Ultra-performance liquid chromatography; US EPA, United States environmental protection agency; WC-TMMIP, Water-compatible temperature and magnetic dual-responsive MIP; XPS, X-ray photoelectron spectroscopy; XRD, X-ray diffraction; β-CD, β-cyclodextrin.

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compounds and the low-abundance of target components, the direct determination of analytes is difficult. Till now, several analytical methods have been described for food and environmental quantification to overcome the limitations. Proper isolation of target analytes from complex sample matrices relies on the use of very efficient extraction materials which able to interact with them in a selective way. Up to now, polymeric or silica-based sorbents and more recently nanostructured sorbents are usually applied in this step. However, some of them lack selectivity, especially when complex matrices are processed, leading to the co-extraction of various interferences.

In the recent years, molecular imprinting technology (MIT) has been considered as an attractive method to produce artificial receptors obtained with the memory of size, shape, and functional groups of the template molecules [1]. Molecularly imprinted polymers (MIPs) have been widely used as biomimetic molecular recognition receptors with recognition sites for a given molecule structure [2]. The specific recognition property of MIPs was based on the formation of complexes between the appropriate functional monomers with the templates which mainly could bind covalently or non-covalently. Non-covalent interactions between the functional monomers and the template are probably the most flexible regarding the selection of the possible template molecules and the functional monomers [3]. These complexes were then immobilized by copolymerization with a high concentration of cross-linkers. After the polymerization was complete, the templates were removed, providing binding sites in the MIPs that had complementary shapes, sizes, and functionalities toward the target template. With these tailor-made binding sites, MIPs not only recognize the shape and size of a given template but also respond to the functional groups of the molecule [4]. Compared with traditional media used for separation and analysis, MIPs, featuring good mechanical/chemical stability, high specificity, low cost, ease of preparation and reversible adsorption/release of the target molecule which are receiving significant attention as robust and smart materials for separation and sample preconcentration [5]. The schematic image of the MIP preparation is summarized in Fig. 1. The advantages of MIPs caused broad applications in the analytical chemistry where they are used for the separation or detection of many compounds. MIPs possess an extensive application potential in separation science [6,7], food analysis [8], biological and environment analysis [9], and other fields [10,11]. However, some drawbacks to MIPs have restricted their widespread application, such as slow mass transfer, irregular shape, incomplete template removal, poor site accessibility, or heterogeneous distribution of binding sites [12]. Many efforts have been made to solve the above issues. One of the promising alternatives is imparting magnetism to the MIPs and then using magnetic separation.

Magnetic separation technology, in which polymers are prepared using fabricating the MIP on the surface of a magnetic substrate, has received remarkable attention in recent years for its potential application in separation and extraction [13]. Recently, magnetic molecularly imprinted polymers (MMIPs) have attracted significant attention attributing to the fact that they not only exhibit specific selective binding for the template molecule but also have outstanding magnetism [14]. Thus, the MMIPs can be isolated easily from samples with an external magnet without filtration or centrifugation step [15]. The resulting MMIPs are added to a solution or suspension containing the target analyte and stirred to permit the analyte to adsorb onto the magnetic polymers. Then, the polymers with captured analyte are recovered from the suspension with a suitable magnetic separator. Finally, the target analyte is eluted from the polymers and analyzed. This technique provides a relatively easy and rapid way to remove magnetic polymers from sample matrices by applying a magnetic field without the need for tedious filtration or centrifugation [16]. The MMIPs, compared with conventional MIPs, show many superior characteristics involving fast and effective binding to target analytes, magnetically susceptible characteristic, and shorter pretreatment time [17]. Currently, magnetic molecularly imprinted solid-phase extraction (MMISPE) is one of the most accepted sample pretreatment approaches for the preconcentration and extraction of analytes in the field of food, environmental, and biological samples because of its simplicity, convenience, and time-saving [18]. Furthermore, MISPE coupled with chromatographic detection technology easily detects traces of endocrine disrupting chemicals in complex matrices [19]. However, one of the MIPs limitations in their applications as the sorbent in SPE or SPME is their small particle sizes, which makes their separation process from aqueous samples difficult. Because they can be quickly separated by an external magnetic field after they are dispersed in the solution, magnetic nanoparticles (MNPs) are appropriate sorbents for SPE [20]. The selectivity of MNPs has been

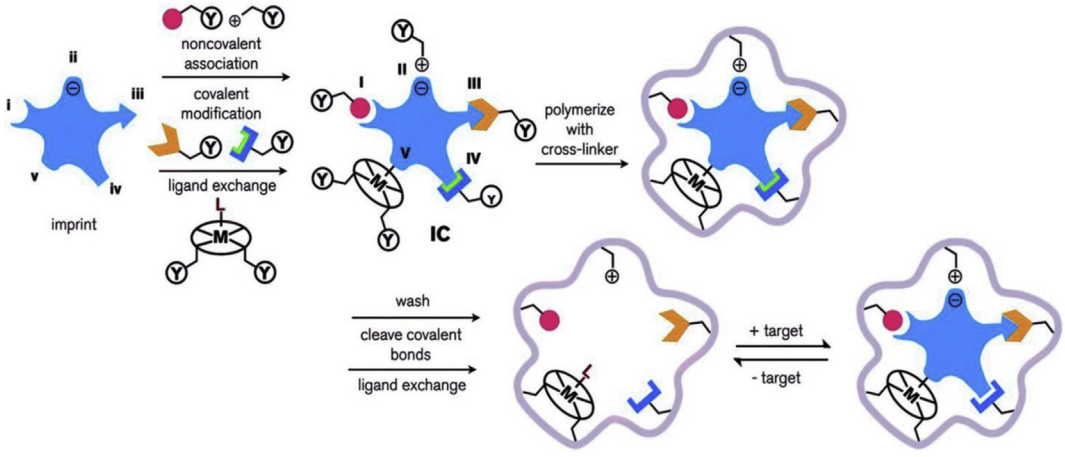

Fig. 1. Schematic representation of the synthesis, recognition, and separation of an MIP. There are five main types of molecular imprinting process: (i) non-covalent; (ii) electrostatic/ionic; (iii) covalent; (iv) semi-covalent; and (v) metal center coordination. Template molecules and functional monomers, containing a functional group, Y, combine via cross-linkers. A three-dimensional polymer network form after the polymerization progress. Then, the template molecules remove using washing, ligand exchange or cleaving covalent bonds, leaving cavities similar to template molecules in shape, size, and molecular interactions. By the addition of chiral compounds, the enantiomer similar to the template molecule will have a stronger interaction with the surrounding cavities. Reproduced with permission [6]. Copyright 2014 The Royal Society of Chemistry.

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