Talanta 167 (2017) 470–485

Contents lists available at ScienceDirect

Talanta

journal homepage: www.elsevier.com/locate/talanta

Recent configurations and progressive uses of magnetic molecularly imprinted polymers for drug analysis

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ARTICLE INFO

Keywords:
Drug analysis
Magnetic molecularly imprinted polymer
Magnetic particles
Magnetic solid-phase extraction
Synthesis of MMIP

ABSTRACT

Since the introduction of the molecularly imprinting technology (MIT) in the 1970s, it becomes an emerging technology with the potential for wide-ranging applications in drug determination. With the rise of green chemistry, many researchers began to focus on the application and development of green materials which led to the breakthrough of molecularly imprinted polymers (MIPs) in the green chemistry. Because of the low concentration levels in the human matrices, almost adequate analytical methods should be used for quantification of drugs at the trace levels. In recent years there have been reported benefits of combining MIPs with additional features, e.g. magnetic properties, through the build-up of this type of material on magnetite particles. Magnetic molecularly imprinted polymer (MMIP) is a new material which is composed of magnetic material and non-magnetic polymer material and shares the characteristics of high adsorption capacity to template molecule, special selective recognition ability, and the magnetic adsorption property. These materials have been widely used in the different fields such as chemical, biological and medical science. This review describes the novel configurations and progressive applications of magnetic molecularly imprinted polymers to the drug analysis. Also, the advantages and drawbacks of each methodology, as well as the future expected trends, are evaluated.

Abbreviations: 17β-E2, 17β-Estradiol; 4-VP, 4-vinylpyridine; 5-DTAF, 5-(4,6-dichlorotriazinyl) aminofluorescein; AIBN, 2,2azobisisobutyronitrile; AMPS, Acrylamido-2-methyl-1-propanesulfonic acid; APTS, 3-aminopropyltrimethoxysilane; ATRP, Atom transfer radical polymerization; BHb, Bovine hemoglobin; BZE, Benzoylecgonine; CAP, Chloramphenicol; CE-FD, Capillary electrophoresis with fluorescence detection; COC, Cocaine; Core-shell MMIP, Core-shell Magnetic molecularly imprinted polymer; DES, Deep eutectic solvent; DFC, Diclofenac; DI, Dihydrocodeine; DIS, Dienestrol; DLLME, Dispersive liquid-liquid microextraction; DMMIP, Dimetridazole-magnetic molecular imprinted polymer; DMZ, Dimetridazole; DVB, Divinylbenzene; ECL, Electrochemiluminescence; EDS, Energy dispersive X-ray spectroscopy; EE, Environmental estrogen; EGDMA, Ethyleneglycol dimethacrylate; EGMRA, Ethylene glycol maleic rosinate acrylate; EME, Ecgonine methyl ester; EMMIP, Electromagnetic molecularly imprinted polymer; EMMIP-GCE, Electromagnetic molecularly imprinted polymer with glassy carbon electrode; FD, Fluorescence detection; FF, Florfenicol; GC-ECNI-MS, Gas chromatography-electron capture negative ionization mass spectrometry; GC-MS, Gas chromatography-mass spectrometry; GO, Graphene oxide; GSH, Glutathione; HPLC-UV, High performance liquid chromatography with ultraviolet detection; HPMMIP, Hollow porous Magnetic molecularly imprinted polymer; HPTLC, High performance thin layer chromatography; HAS, Human serum albumin; IP-CPE, Ion-pair cloud-point extraction; LC-DAD, Liquid chromatography-diode array detector; LC-MS, Liquid chromatography-mass spectrometry; LOD, Limit of detections; LOQ, Limit of quantification; LPME, Liquid-phase microextraction; MAA, Methacrylic acid; MAC, Methacrylamide; MBA, N,N′-methylenebisacrylamide; MC, Methacryloyl chloride; MDMINP, Magnetic dummy molecularly imprinted nanoparticle; MDMIP, Magnetic dummy molecularly imprinted polymer; MGCE, Magnetic glassy carbon electrode; MIP, Molecularly imprinted polymer; MIP-HFT, Molecularly imprinted polymer-coated polypropylene hollow fiber tube; MIT, Molecularly imprinting technology; MMINP-d-SPE, Magnetic molecularly imprinted nanoparticle with dispersive solid-phase extraction; MMIP, Magnetic molecularly imprinted polymer; MMIP-CNT, Magnetic molecularly imprinted polymer with carbon nanotube; MMIP-DES, Magnetic molecularly imprinted polymer with deep eutectic solvents; MMIP-d-SPME, Magnetic molecularly imprinted polymer with dispersive solid-phase microextraction; MMIP-GCE, Magnetic molecularly imprinted polymer with glassy carbon electrode; MMIP-GO, Magnetic molecularly imprinted polymer-graphene oxide; MMIP-MWCNT, Magnetic molecularly imprinted polymer with multi-walled carbon nanotube; MMIP-SBSE, Magnetic molecularly imprinted polymer with stir-bar sorptive extraction; MMIP-μ-SPE, Magnetic molecularly imprinted-micro-solid phase extraction; MMISF, Magnetic molecularly imprinted sensing film; MMISF-GCE, Magnetic molecularly imprinted sensing film with glassy carbon electrode; MMISPE, Magnetic molecularly imprinted solid phase extraction; MMMIP, Montmorillonite magnetic molecularly imprinted polymer; MMT, Montmorillonite; MNIP, Magnetic molecularly non-imprinted polymers; MNP, Magnetic nanoparticles; MNZ, Metronidazole; MO, Morphine; MP, Magnetic particles; MPS, 3-methacryloxypropyltrimethoxy-silane; MRI, Magnetic resonance imaging; MSPD, Matrix solid-phase dispersion; MSPE, Magnetic solid phase extraction; MUTDMIP, Magnetic ultra-thin dummy molecularly imprinted polymer; NIP, Non-imprinted polymer; NIPAM, N-isopropylacrylamide; OA, Oleic acid; PDA, Photodiode array detector; PM-MIM, Photonic-magnetic responsive molecularly imprinted microsphere; PNIPAM, Poly (N-isopropylacrylamide); PPZ, Perphenazine; RAC, Ractopamine; RAFT, Reversible addition-fragmentation chain transfer polymerization; RIV, Rivastigmine; SBSE, Stir-bar sorptive extraction; SPE, Solid-phase extraction; SPME, Solid-phase microextraction; Surface-MMIP, Surface magnetic molecularly imprinted polymer; TAP, Thiamphenicol; TEOS, Tetraethyl orthosilicate; TMMIP, Thermal-responsive and magnetic molecularly imprinted polymer; TRIM, Trimethylolpropane trimethacrylate; VSM, Vibrating sample magnetometer; WC-TMMIP, Water-compatible temperature and magnetic dual-responsive MIP; μ-SPE, Micro-solid phase extraction

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http://dx.doi.org/10.1016/j.talanta.2017.02.049
Received 5 January 2017; Received in revised form 20 February 2017; Accepted 21 February 2017
Available online 22 February 2017

1. Introduction

In recent years, a significant progress has been attained in the field of drug analysis devices. In most cases, the complex matrix and the low concentration of analyte strongly limit and lower the characteristic performance of most analytical techniques and induce the researchers to introduce newly separation and/or preconcentration methods. Till now, several methods for the drugs quantification in biological samples [1,2] and pharmaceutical preparations [3,4] such as solid-phase extraction (SPE) [5], solid-phase microextraction (SPME) [6], stir-bar sorptive extraction (SBSE) [7], matrix solid-phase dispersion (MSPD) [8], micro solid-phase extraction (MSPE) [9], liquid-phase microextraction (LPME) [10], dispersive liquid-liquid microextraction (DLLME) [11], and ion-pair cloud-point extraction (IP-CPE) [12] have been described utilizing high performance liquid chromatography with ultraviolet detection (HPLC/UV) [13,14], fluorescence detection (FD) [15], gas chromatography-mass spectrometry (GC-MS) [16], liquid chromatography-mass spectrometry (LC-MS) [17,18], high performance thin layer chromatography (HPTLC) [19] and spectrofluorometric [15] to overcome the limitations. Hence, a sample pretreatment technique should be used for determination of organic pollutants that presenting at trace levels in the complex matrices.

At this regard, nowadays sample preparation is considered as a bottleneck of the whole analytical process. The removal of potential interferents, analyte preconcentration, converting (if needed) the analyte into a more appropriate form for separation or detection, and providing a robust and reproducible technique independent of variations in the sample matrix are the main objectives of sample preparation. More recently, new objectives have been set such as using smaller initial sample sizes, improvement of selectivity in extraction, to facilitate the automation, and to minimize the amount of organic solvents and glassware to be used [20]. However, the main drawback associated with them is the lack of selectivity of the sorbents used making necessary an extensive optimization of the typical steps involved in the clean-up and extraction of target analytes. Unfortunately, even after careful optimization, some matrix components are co-eluted with target analytes making difficult to reach detection limits according to the nowadays accurate regulations.

Molecularly imprinted polymers (MIPs) are the class of synthetic polymeric materials with special molecular recognition abilities that provide a high selectivity towards the selected molecules. In these tailor-made polymers, the recognition sites are imprinted in the polymer matrix by the presence of a template during their synthesis which is formed according to the shape, size, and the functional groups of the template molecule [21]. MIPs are used in the chemical analysis in different forms: spherical particles, crushed monolith or a polymer layer coated on another medium (e.g. magnetite or silica) [6]. They offer high mechanical, chemical, and thermal stabilities. These materials are obtained by the polymerizing cross-linking agent and functional monomers with large amount of porogenic solvents around a template molecule, leading to a highly cross-linked three-dimensional network polymer. The functional monomers are chosen considering their capability to interact with the functional groups of the template molecule. Once polymerization has taken place, template molecules are extracted and binding sites with size, shape, and functionalities complementary to the target analyte are established. The resulting imprinted polymers are robust, stable, and resistant to a wide range of pH, temperature, and solvents. Therefore, the behavior of MIPs emulates the interactions established by natural receptors to selectively retain a target molecule (i.e. antibody-antigen) but without stability limitations. In addition, it is important to point out that synthesis of MIPs is also relatively easy and cheap, making them a clear alternative to the use of natural receptors. The schematic image of the MIP preparation is summarized in Fig. 1.

The MIPs advantages caused broad applications in the analytical chemistry such as foods, drugs, biological and environmental samples [22] where they are used for the detection or separation of many compounds. In drug analysis field, we require the quantification of drugs and its metabolic compounds in trace levels from biological samples such as plasma, serum, and urine. Due to their outstanding molecular recognition characteristics, application and combination of such materials with liquid chromatography [23], capillary electrophoresis [24], capillary electrochromatography [25], SPE [26,27], SPME [28,29], sensors [30,31], enantiomeric separations [32], catalysis [33,34] and etc. lead to simplifying, avoiding matrices effect, and also significant improvement. Nevertheless, the main challenges have limited their widespread and successful use, such as time-consuming separated, slow mass transfer, and template leakage. Moreover, traditional MIPs in which molecules imprint layer are very thick having the disadvantages of hard eluting, large diffusion resistance, deeply embedded template in the internal, and low binding rate. In recent years, MIPs have been combined with additional properties of the support, e.g. magnetic properties. By solving this problem, the promising strategy of magnetic molecularly imprinted polymer (MMIP) has been developed. MMIP can be easily collected /isolated and recycled by an external magnetic field without additional centrifugation or filtration [35,36].

For the very first time, with the use of a magnetic iron oxide, in 1998, an MMIP with a mean diameter of 13 μm produced by the polymerization of monomers in liquid perfloro chlorine [37]. In

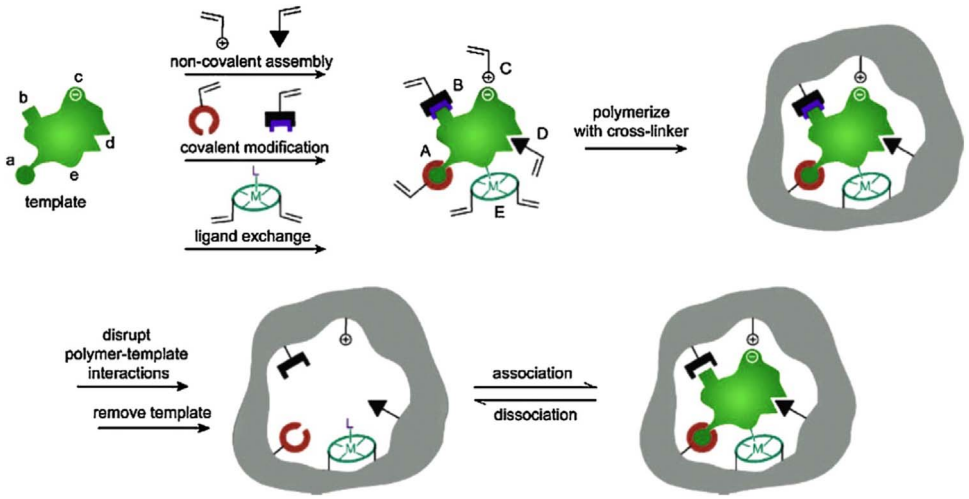

Fig. 1. Schematic representation of the synthesis and molecular imprinting process. Reproduced with permission [22].