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Functionalized Magnetic MOF-Based Sorbents for Sample Preparation in Food Contaminant Analysis: Advances, Mechanistic Evidence, and Application Boundaries

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Abstract

Food contaminants often occur at trace levels in complex matrices rich in proteins, lipids, carbohydrates, pigments, and inorganic salts, making sample preparation a critical determinant of analytical sensitivity, accuracy, and robustness. Metal-organic frameworks (MOFs) provide tunable pore architectures and tailorable surface chemistry, while integration with magnetic components such as Fe_3O_4 enables rapid magnetic recovery. This mechanism-oriented narrative review examines the structure–performance relationships and application boundaries of functionalized magnetic MOF-based sorbents, focusing on interfacial engineering, multicomponent hybridization, surface chemical tailoring, and specific recognition. Although the available evidence remains limited, it indicates that specific surface area alone cannot explain practical extraction performance; analyte–site complementarity, pore accessibility, magnetic separation efficiency, and competition from matrix constituents are also critical. The review therefore emphasizes matrix validation and mechanistic evidence rather than direct comparison of detection limits obtained under heterogeneous conditions.

Keywords

functionalized magnetic MOFs, food contaminants, sample preparation, magnetic solid-phase extraction, structure–performance relationship, food safety analysis

1. Introduction and Application Context

1.1 Application Context

Foods may be exposed to pesticides, veterinary drugs, mycotoxins, heavy metals, per- and polyfluoroalkyl substances (PFAS), plasticizers, and processing contaminants during production, farming, processing, storage, transportation, and packaging. These analytes differ markedly in molecular size, aromaticity, polarity, acid-base dissociation behavior, and coordination capability, and they are generally present at low concentrations. Proteins and lipids in dairy products, starches and pigments in cereals, fatty acids in edible oils, and high salt contents in soy sauce and preserved foods can cause co-extraction, chromatographic contamination, ion suppression or enhancement, and quantitative bias. Sample preparation in food analysis must therefore achieve

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analyte extraction and enrichment while also addressing matrix cleanup, processing speed, solvent consumption, operational reproducibility, and applicability across diverse matrices.

Conventional liquid-liquid extraction, cartridge-based solid-phase extraction, and immunoaffinity cleanup are well established, but in some applications they remain limited by solvent consumption, backpressure or clogging, disposable-consumable costs, and insufficient selectivity. In magnetic solid-phase extraction (MSPE), the sorbent is dispersed directly in the sample solution and recovered after adsorption with an external magnetic field, reducing the need for cartridge packing, filtration, and high-speed centrifugation. MOFs consist of metal ions or clusters coordinated with organic linkers and can provide tunable pores, aromatic frameworks, open metal sites, and interfaces for post-synthetic modification. Coupling MOFs with magnetic particles enables rapid solid-liquid separation while retaining a porous adsorption interface. Early food-analysis studies demonstrated that Fe_3O_4@MOF-808, Fe_3O_4-NH_2@MOF-235, and IRMOF-3/SiO_2/Fe_3O_4 could enrich pesticides or antibiotics from beverages, honey, and fish samples [1–3].

A magnetic component combined with a MOF does not inherently confer high selectivity or matrix tolerance. Unprotected magnetic cores can aggregate and oxidize; MOF shells may detach or attenuate magnetic response; and nonspecific pore adsorption remains susceptible to competition from structural analogues and coexisting food constituents. Different analytes therefore require different material-design strategies. Aromatic pesticides, antibiotics, and plasticizers tend to benefit from π–π and hydrophobic interactions; PFAS require simultaneous consideration of the fluorinated chain and hydrophilic headgroup; inorganic contaminants such as Hg, Pb, Cd, and As depend more strongly on coordination with S-, N-, or O-containing sites; and mycotoxins or structurally similar small molecules may benefit from aptamers or molecular imprinting to improve recognition. Accordingly, this review follows a framework of construction strategy, mechanistic evidence, method validation, and application boundaries rather than cataloguing studies solely by contaminant class.

2. Construction Fundamentals of Magnetic MOF-Based Sorbents

2.1 Structural Components and Construction Modes

Functionalized magnetic MOF-based sorbents generally comprise a magnetic unit, an interfacial coupling layer, and a porous adsorption layer. Fe_3O_4 is the most common magnetic component, although Co- or Ni-containing frameworks and their derivatives have also been used to generate magnetic response. The MOF layer supplies pores, aromatic structures, metal sites, and post-modification interfaces, whereas polydopamine (PDA), SiO_2, chitosan, and amino- or carboxyl-functionalized layers can improve interfacial bonding, dispersibility, and stability. According to the spatial relationship between the magnetic component and the MOF, these materials can be classified as magnetic particles supported on MOF surfaces, MOF-coated magnetic cores, magnetic particles embedded in MOF crystals, or multilayer core-shell structures. Fe_3O_4@MOF-808 exemplifies a composite in which magnetic particles are attached to MOF crystal surfaces; the material retained a saturation magnetization of 40.35 emu/g and could be magnetically separated in approximately 0.5 min [1]. In Fe_3O_4-NH_2@MOF-235, coordination between surface amino groups and Fe^{3+} induces MOF growth, illustrating an interfacial anchoring–in situ nucleation strategy [2].

Direct composite formation is comparatively straightforward, but magnetic particles may be distributed nonuniformly or block pore entrances. Core-shell architectures provide clearer spatial division of functions and improved resistance to layer detachment, but thicker nonmagnetic shells generally reduce magnetization per unit mass and increase mass-transfer distance. In IRMOF-3/SiO_2/Fe_3O_4, the SiO_2 layer isolates the magnetic core and provides a modifiable surface; the material was applied to quinolone analysis in fish, where enrofloxacin and ciprofloxacin were detected [3]. MOFs can also be integrated with chitosan into millimeter-scale hydrogel beads, converting powders into macroscopic sorbents with easier handling and lower material loss [4]. Porous carbons obtained by pyrolyzing magnetic MOF precursors no longer retain the original coordination framework and should therefore be evaluated separately as MOF-derived materials.

2.2 Performance Requirements for Food Sample Preparation

Materials for food sample preparation cannot be evaluated solely on specific surface area and adsorption capacity in standard solutions. They must retain acceptable magnetic recovery kinetics in highly viscous

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extracts or extracts with high solid content, and they must remain structurally intact in aqueous media, across the relevant pH range, and under organic-solvent desorption conditions, while minimizing background release of metal ions, linkers, or polymers. Analytes must also reach effective binding sites within a practical time. When multilayer coatings block pore entrances or become excessively thick, a high theoretical surface area may not translate into improved practical recovery. After incorporation of a COF layer, Fe_3O_4@NH_2-MIL-88(Fe)@TpBD showed a marked increase in specific surface area but a decrease in saturation magnetization to approximately 10.14 emu/g, illustrating a trade-off between increasing porosity and preserving magnetic response [5].

Selectivity and matrix tolerance are central to food applications. More persuasive evidence includes spike-recovery experiments across multiple food matrices, incurred samples, quality control materials (QCMs) or certified reference materials (CRMs), and consistency across sorbent batches. These evidence types serve different purposes: incurred samples primarily demonstrate method utility under authentic contamination conditions, whereas CRMs and QCMs are more directly used to assess accuracy and agreement with assigned or reference values; they are not interchangeable. Fe_3O_4@UiO-66-SH has been used for mercury speciation in fish tissue with CRM validation [6], and a related material was used to separate Pb and Cd from high-salt foods such as soy sauce [7]. An aptamer-functionalized magnetic MOF study of aflatoxin B_1 (AFB_1) in cereals used contaminated-grain QCMs [8]. Together, these studies represent relatively complete methodological chains spanning adsorption, magnetic separation, desorption, instrumental analysis, and matrix validation.

3. Functionalization Strategies and Structure–Performance-Oriented Design

3.1 Interfacial Engineering and Core-Shell Structures

Interfacial engineering primarily addresses magnetic-core aggregation, nonuniform MOF nucleation, and inadequate interlayer adhesion; it does not inherently provide molecular-level specificity. Polydopamine can self-polymerize under mildly alkaline conditions and adhere to various inorganic surfaces through catechol, amine, and aromatic functionalities, thereby providing anchoring sites for metal-ion coordination and MOF growth. Tang et al. coated Fe_3O_4 with PDA and then grew MIL-101(Cr), producing Fe_3O_4@PDA/MIL-101(Cr) for cleanup of five mycotoxins in licorice. The method achieved limits of detection (LODs) of 0.01–0.09 μg/kg and recoveries of 78.53%-116.28% [9]. PDA has also been used in constructing Fe_3O_4@UiO-66-NH_2 and provided a composite platform for coupling an AFB_1 aptamer [8]. These findings support the roles of PDA in interfacial adhesion, MOF loading, and secondary functionalization, but they do not by themselves demonstrate improved analyte specificity.

SiO_2 can reduce the risks of Fe_3O_4 dissolution and oxidation under acidic conditions and can be further functionalized through silanol groups with amino, thiol, or other ligands. IRMOF-3/SiO_2/Fe_3O_4 retained its main crystalline and porous features over a relatively broad pH range [3], and Fe_3O_4-SiO_2-MIL-53(Fe) was used to enrich Cd from spices, chocolate, and tea [10]. Because SiO_2 itself contributes neither magnetism nor high adsorption capacity, an excessively thick coating dilutes the effective MOF content. It is therefore better used as a protective and anchoring layer than as a feature to be maximized in thickness.

Natural-polymer interfaces can support both dispersion stability and macroscopic shaping. In Fe_3O_4@CS@ZIF-67, amino and hydroxyl groups in chitosan both coat Fe_3O_4 and provide nucleation sites for in situ ZIF-67 growth, enabling preparation under ambient temperature and pressure and relatively rapid adsorption of benzoylurea insecticides from tea infusions [11]. A magnetic NH_2-UiO-66-chitosan hydrogel further integrated particles into millimeter-scale beads, reducing powder loss and handling difficulty [4]. Because macroscopic shaping can reduce site accessibility, interfacial design should achieve a measurable balance among stability, shell thickness, pore accessibility, and magnetic recovery.

3.2 Multicomponent Hybridization and MOF-Derived Materials

The purpose of multicomponent hybridization is to create complementary interfaces rather than merely increase the number of material components. Graphene oxide (GO) provides two-dimensional sheets, aromatic interfaces, and oxygen-containing groups that can support MOF growth and surface imprinting. Alilou et al. constructed a molecularly imprinted polymer (MIP) layer on magnetic GO/MOF-808 to obtain MGO/MOF-808@MIP. Non-imprinted controls, component-ratio experiments, and adsorption tests reported in the full

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article indicated that GO helped provide accessible growth interfaces, whereas MIP sites were primarily responsible for aflatoxin recognition [12]. The same study showed that excessive GO content reduced accessible porosity, indicating that the ability of a two-dimensional support to expand the accessible interface must be demonstrated through compositional controls rather than inferred from the material name alone.

MOF/COF composites can exploit the conjugated aromatic skeletons and solvent stability of COFs. In Fe_3O_4@NH_2-MIL-88(Fe)@TpBD, the TpBD-COF layer expanded the π-conjugated interface; when the material was applied to three fluoroquinolones in chicken, recoveries of 85.8%-112.2% were obtained [5]. At the same time, COF incorporation reduced magnetization, indicating that the mass fraction and thickness of the functional layer require control. Fe_3O_4-MWCNT/MOF-199 used a one-dimensional carbon-nanotube interface for extracting triazole pesticides from fruits, vegetables, and fruit juices [13].

MOF-derived carbons generated by pyrolysis can improve aqueous, thermal, and chemical stability, but the original MOF coordination framework is lost. Davari et al. pyrolyzed magnetic MIL-101(Fe) to obtain magnetic porous carbon and then grafted a polypyrrole-polyaniline copolymer layer, producing MPC@PPy-PANI for the extraction of furfural and 5-hydroxymethylfurfural (HMF) from infant foods and milk powder. The method achieved recoveries of 86%-111%, and the material was reused for six cycles [14]. In this system, carbonization provides structural stabilization, whereas secondary polymer functionalization adds hydrogen-bonding and aromatic-interaction sites for relatively polar analytes. Intact MOF composites and MOF-derived carbons have fundamentally different structural bases and should not be ranked directly using LOD or adsorption capacity alone.

3.3 Surface Chemical Tailoring and Specific Recognition

Surface chemical tailoring directly alters interfacial affinity for target analytes. Han et al. immobilized a deep eutectic solvent (DES) composed of levulinic acid and trifluoromethoxybenzene on magnetic UiO-66-NH_2 for the analysis of fluorotelomer alcohols (FTOHs) in edible oils [15]. By combining $^1H/^{19}F$ nuclear magnetic resonance (NMR), noncovalent-interaction analysis, and density functional theory (DFT) calculations, the study proposed that dual hydrogen bonding and fluorinated-group interactions jointly contributed to extraction. Its mechanistic evidence is therefore stronger than an inference based only on functional-group structure. A study of Fe_3O_4@ZIF-8&F13 for five perfluoroalkyl carboxylic acids (PFCAs) combined contact-angle measurements, pH effects, homologous-series adsorption trends, and DFT calculations, supporting joint contributions from electrostatic, hydrophobic, and fluorinated-chain interactions [16]. For PFAS, sorbent design should therefore match both the hydrophilic headgroup and the fluorinated chain rather than simply maximize surface hydrophobicity.

Aptamers and molecular imprinting provide more explicitly defined recognition sites. A chloramphenicol aptamer-functionalized magnetic MIL-53(Al)-NH_2 was used for selective extraction from milk and honey [17]. An AFB_1 aptamer-functionalized Fe_3O_4@MOF achieved recoveries of 89.7%-99.5% across four grain matrices, with accuracy assessed using QCMs and reusability evaluated through repeated adsorption-desorption cycles [8]. Its reported operational lifetime should nevertheless be regarded as study-specific.

Molecularly imprinted polymers (MIPs) form cavities complementary to target shape, size, and functional groups through template-monomer preorganization and crosslinking. Fe_3O_4@ZIF-8@MIP achieved a method LOD of 0.1 μg/L for bisphenol A and was validated in three sample matrices [18]. Fe_3O_4@MOF@MIP-160 showed high adsorption capacities for DBP and DEHP, rapid equilibration, and reuse over six cycles [19]. These findings support enhanced analyte-site complementarity, although imprint-layer thickness, site heterogeneity, template leakage, and nonspecific polymer adsorption remain important.

Studies of inorganic contaminants should distinguish among speciation analysis, selective extraction, and matrix cleanup. Fe_3O_4@UiO-66-SH exploits thiol affinity for Hg species to enrich Hg^{2+} and methylmercury and was validated with a fish-tissue CRM [6]; the objective is speciation analysis before chromatographic separation. A magnetic MOF-199/dithiocarbamate composite selectively retained As(III) at pH 3, followed by a reduction step to determine total inorganic arsenic, representing oxidation-state-selective extraction [20]. A related sulfur-containing material separated Pb and Cd from high-salt foods such as soy sauce, where its principal function was salt-matrix cleanup [7]. A study of dithizone-functionalized magnetic MIL-101(Fe) prepared by post-synthetic modification further showed that post-synthetic chelating groups can increase metal-binding sites [21]. Full-text evidence for ionic-liquid-functionalized magnetic MOFs in food-

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contaminant sample preparation remains insufficient, and this review therefore does not classify that approach as a mature route.

4. Adsorption Mechanisms and Structure-Performance Relationships

4.1 Multiple Interactions and Strength of Mechanistic Evidence

Enrichment by functionalized magnetic MOFs may involve pore filling, π–π interactions, hydrophobic interactions, hydrogen bonding, electrostatic interactions, metal coordination, and specific recognition. However, the extent to which these mechanisms are demonstrated differs substantially among studies. Mechanistic interpretations are more persuasive when supported by convergent evidence types, such as NMR, X-ray photoelectron spectroscopy (XPS) changes before and after adsorption, competition experiments, and theoretical calculations. Comparisons before and after functionalization or with non-imprinted controls, together with pH, ionic-strength, homologous-series, or selectivity experiments, provide indirect support. Hypotheses based mainly on functional-group structures and adsorption outcomes remain less conclusive. These distinctions reflect the sufficiency of mechanistic evidence, not the intrinsic importance of a mechanism.

For Fe_3O_4@MOF-808, enrichment factors for benzoylurea insecticides generally increased with hydrophobicity, while hydrogen bonding and π–π interactions were also proposed on the basis of amide and aromatic-ring structures [1]. This interpretation is supported by homologous-series trends but lacks direct characterization of binding. Studies of DES-functionalized and fluorinated magnetic MOFs combined NMR or DFT analysis, providing stronger support for their mechanistic interpretations of FTOHs and PFCAs [15, 16]. For aptamers and MIPs, structural analogues, non-imprinted materials, and competition experiments can support specificity, although nonspecific hydrophobic and π interactions may still contribute. A good fit to a pseudo-second-order kinetic model is an empirical modeling result and, by itself, does not constitute evidence of chemisorption.

4.2 Contaminant Structure, Food Matrix, and Material Selection

Target structure should be the starting point for sorbent selection. Pesticides, veterinary drugs, and plasticizers with multiple aromatic rings and strong hydrophobicity are well suited to materials containing aromatic linkers, COFs, GO, or carbon interfaces, with amino, carboxyl, or metal sites providing additional hydrogen-bonding interactions. Sorbent design for PFAS should account simultaneously for fluorinated-chain affinity and headgroup interactions: neutral FTOHs may be targeted using hydrogen bonding and fluorinated interfaces, whereas ionic PFCAs additionally require consideration of surface charge and working pH. For structurally similar analytes such as mycotoxins that are regulated at low levels, aptamers or surface imprinting are more likely to reduce interference from analogues. For heavy metals and metalloids, thiols, dithiocarbamates, dithizone, or nitrogen-containing ligands should be selected according to the hard–soft acid–base (HSAB) principle and oxidation state, rather than simply maximizing porosity.

The food matrix determines whether functional sites remain accessible. Fatty acids in edible oils may compete for hydrophobic interfaces or foul surfaces; pigments, polysaccharides, and natural phenolics in cereals, licorice, and tea can increase co-extraction; and fish and dairy products generally still require prior solvent extraction, defatting, or protein precipitation because magnetic MOFs cannot fully replace the analyte-release step. In high-salt foods, the practical value of sulfur-functionalized magnetic UiO-66 lies not only in enriching Pb and Cd but also in transferring the metals from the salt matrix to a desorbable solid phase, thereby reducing the salt load introduced into the ICP-MS system [7]. Material performance must therefore be interpreted within the specific combination of target analyte, extraction medium, coexisting matrix components, and detection platform.

Accordingly, functionalized magnetic MOFs should be viewed as complementary rather than universal substitutes for established workflows. Their strongest application boundary lies in cases requiring simultaneous trace enrichment and matrix cleanup, tailored recognition of structural analogues, or simplified magnetic separation. QuEChERS remains preferable for high-throughput multiresidue analysis, immunoaffinity columns for standardized mycotoxin cleanup, and commercial SPE for routine transferability. For inorganic contaminants, performance criteria should match the analytical task: species preservation for Hg, oxidation-state selectivity for As, and salt-matrix removal for Pb and Cd.

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5. Discussion

5.1 Current Limitations

Material stability and safety remain prerequisites for application. Some MOFs may undergo coordination-bond cleavage in water, acid, base, or high-salt media, while magnetic cores may oxidize, aggregate, or release metal ions. Multilayer structures add washing, activation, and solvent-exchange steps, thereby amplifying batch-to-batch variation. Existing studies frequently report initial magnetization and crystal structure but less often evaluate metal leaching, linker release, pore structure, and magnetic recovery time after repeated acidic or alkaline desorption cycles. Sorbents intended for food analysis should also report procedural blanks and the potential for secondary contamination.

The scope and definitions of method validation remain inconsistent. Spiking commercially purchased samples confirmed to be negative is not equivalent to analyzing naturally contaminated or otherwise genuinely positive samples. CRMs and QCMs demonstrate accuracy but do not replace evaluation of applicability to incurred matrices. Reuse is sometimes judged only from recovery values without confirming whether structure, magnetic response, and functional layers are retained.

Mechanistic claims should remain proportional to the available evidence. Reliable structure-performance relationships require functionalization controls, competitor experiments, and, where possible, complementary spectroscopic or computational evidence. Inconsistent reporting of sample volume, sorbent dose, enrichment factor, LOD, and reuse cycles further limits quantitative comparisons across studies.

5.2 Development Priorities

The immediate priority should be methodological verifiability rather than adding further functional layers. A minimum reporting framework should include sorbent batch information, magnetic separation time, absolute and relative recoveries, matrix effects, desorption efficiency, the definition of a reuse cycle, post-cycle structure, and metal and linker leaching. Validation should also be expanded to include CRMs/QCMs, naturally contaminated samples, and high-protein, high-lipid, high-salt, and highly pigmented matrices. A study using a food-grade cyclodextrin MOF modified with edible organic acids for the extraction of polycyclic aromatic hydrocarbons (PAHs) from foods points to a potentially useful direction for lower-toxicity functionalization [22].

A medium-term objective is to convert manual dispersion, washing, and desorption into reproducible workflows. Magnetic rods, 96-well plates, flow-injection formats, and online valve switching may reduce operator-dependent variability, but automated devices should first demonstrate sorbent batch consistency, quantitative magnetic recovery, and control of cross-contamination. Only after these basic metrics are stable can magnetic MOFs be compared fairly with established sample-preparation workflows in terms of cost, throughput, and robustness.

Machine learning, microfluidics, and integrated enrichment-readout systems remain exploratory. Their development requires standardized datasets, external validation in authentic matrices, and clear separation of the contributions of enrichment, recognition, and signal amplification.

6. Conclusions

Representative full-text studies indicate that functionalized magnetic MOFs provide a tailorable platform for the extraction, cleanup, and enrichment of food contaminants, but the maturity of evidence differs across functionalization routes. Interfacial engineering mainly improves magnetic-core protection, MOF nucleation, and material handling; GO, COFs, carbon nanotubes, and MOF-derived carbons can expand interfaces or improve stability; and fluorination, DESs, sulfur-containing ligands, aptamers, and molecular imprinting more directly alter analyte–site complementarity. Specific surface area alone cannot predict method performance; site accessibility, magnetic separation, matrix competition, and the detection platform collectively determine the final analytical outcome. The next stage should focus less on pursuing isolated record values and more on demonstrating reproducibility and practical value through reference materials, incurred samples, harmonized reporting, and direct technology comparisons.

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Conflicts of Interest

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