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## **A Review on Advancement in MOF and Magnetic MOF-based Materials for Food Packaging and Safety: A Decade Overview of Synthesis Strategies, Application and Future Directions.**

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### **Abstract**

Food safety is a major worldwide problem because of the prevalent threats caused by environmental pollutants. These problems have been significantly improved by metal-organic frameworks (MOFs), whose vast functional surfaces, high porosity, and variable compositions offer tremendous potential. In terms of food safety and packaging, this review addresses the production and use of MOF and magnetic MOF-based materials. This review mainly focuses on the various MOF and magnetic MOF-based sensors developed over the last five years, focusing on their design, functioning mechanisms, and practical uses. The use of MOFs in food packaging materials enhances quality, shelf life, and effectiveness in removing hazardous compounds. Despite these tremendous advances, there are still issues with biocompatibility, non-reactivity, and effective pollutant detection. A comprehensive discussion is held regarding the materials synthesis techniques and fabrication methods, which outline potential directions for future research and development in utilizing novel metal-organic frameworks and magnetic MOF-based solutions for food safety applications.



**Keywords:** Food safety, Metal-organic frameworks, Magnetic metal-organic frameworks, Food packaging, Food contamination

## Introduction

Food packaging is frequently used to package, preserve, and market food products. In the past, people hunted food and consumed it for the sake of their lives. Food packaging was not an idea. They employed leaves and other naturally occurring materials to store or cover their food. Over time, the desire to preserve food emerged (Ullah, Hashmi, Lee, Youk, & Kim, 2022). Globally, there is increasing concern about food security and its assurance, especially in public health. Meat, poultry, milk, eggs, fish, game, and their byproducts are examples of foods with an animal origin. Food safety involves ensuring that food doesn't cause any risks to consumers during preparation and/or consumption under its intended usage (Djekić et al., 2023; Okpala & Korzeniowska, 2023). Food safety has grown in importance over the years due to its significant impact on human health, making it one of humanity's most pressing challenges. According to records from 8000 years ago, food pollutants have a major negative impact on human health (Rather, Koh, Paek, & Lim, 2017; Sadiku, Ashaolu, & Musa, 2020). The World Health Organization (WHO) and the Food and Agriculture Organization of the United Nations (FAO) reported an empirical study in 2019 that estimates 600 million people contract foodborne illnesses annually due to consuming contaminated food (W. Wang, 2020). Food safety problems can arise throughout several food processing,

production, transit, sale, and storage phases as a global concern. Contaminants from the environment, illegal adulterants, additives, and emigrants from packaging elements contaminate food (Gizaw, 2019).

Apart from food contamination, food deterioration is another important factor that renders food unfit for human consumption. To ensure food safety and quality, an analysis is necessary at every stage of the agricultural food supply chain, from the farm to the table (Ahumada & Villalobos, 2009; J. Luo, Leng, & Bai, 2022). To produce food that requires little to no preparation on the part of the consumer, different food processing techniques, such as pasteurization, heating, and sterilization, must be developed. This will increase the supply of various foods with an extended shelf life. People with hectic lifestyles increasingly choose processed foods that are ready to cook or eat, such as fast food, baby food, canned food, frozen food, snack packages, drinks and other varieties. The development or spread of pathogens, exposure to heat, moisture, contamination, and other factors can all be used to verify the quality and safety of processed foods. Furthermore, food freshness testing is crucial for many preservation methods, such as pressure packaging, fermentation, refrigeration, dehydration, and chemical preservatives. It is necessary to provide quick, accurate, dependable, efficient, and simple techniques for detecting food spoilage, impurities, and dangerous foods before they are consumed by people (Martindale & Schiebel, 2017; Rebezov et al., 2021).

The advancements in food packaging could be explained by the special qualities of nanomaterials, which influence their physical, chemical, and biological potentiality compared to their bulk counterparts. These characteristics include superior optical, barrier, thermal stability, antimicrobial activity, and advanced sensing capabilities (Y. Chen et al., 2018). By optimizing nanoparticles in the food sector, food products can have higher nutritional values and longer shelf life, which ensures their safety and integrity (Y. Zhang et al., 2019). Nanotechnologies, which offer enormous promise, might meet numerous demands in the agri-food industry. When nanomaterials, which can be modified by the invention at the 100 nm size range, are used, the sensing field benefits from a better and greater surface area (Bajpai et al., 2018; Biswas et al., 2022; Erdem, Eş, Akceoglu, Saylan, & Inci, 2021). The development of new, miniature nanotechnology devices that are integrated with analytical procedures is one of the main solutions. As a result, nanosensors are frequently employed to find any impurities in food that is sold for human consumption (Javaid, Haleem, Singh, Rab, & Suman, 2021). Recent years have seen the widespread adoption of metal-based sensors as a

superior analytical technique for food safety in terms of simplicity, high efficiency, and sensitivity. The sensor is an assembly made up of sensitive components that, when combined, can measure particular targets by transforming them into signals that may be used by a device in compliance with regulatory standards (Kaur et al., 2022; J. Li & Bo, 2022; Mohindroo, Sarvaiya, Dange, & Varma, 2023; Wu et al., 2024; Yin et al., 2024). Recently, novel materials such as metal oxides, carbon nanotubes, graphene, metal nanoparticles, quantum dots, and MOFs have been tested to develop new sensors with suitable sensing capabilities. metal-organic frameworks (MOFs) are crystalline solids of coordinated metal ions or clusters with organic ligands. Because of their enormous surface areas, porosity, and customizable topologies, MOFs have attracted much interest for various applications, including sensors. Researchers are very interested in the invention of MOF-based sensors. Due to their distinct qualities, MOFs have shown enormous promise in detection (Q. Chen, Qin, Shi, Kang, & Li, 2021; W. Liu et al., 2020; Osman et al., 2019; Rezki et al., 2021).

Conventional techniques are typically used in laboratories, which makes them unsuitable for on-site detection, which requires portable sensors in order to obtain results quickly. Thus, there is an increasing need for the creation of straightforward, quick, affordable, and portable analytical techniques for use in the detection of food safety (Ehiri & Morris, 1994; Linares-Morales, Gutiérrez-Méndez, Rivera-Chavira, Pérez-Vega, & Nevárez-Moorillón, 2018). Conventional detection techniques provide for very high sensitivity and strong selectivity. For instance, high-performance liquid chromatography-mass spectrometry may precisely measure a chemical analytes concentration. However, high-performance liquid chromatography-mass spectrometry is costly and time-consuming since it needs expensive equipment and trained personnel to handle samples (Nie & Nie, 2019; Núñez & Lucci, 2020; Tang, Vasas, Hatzakis, & Spyros, 2019). This increases their usefulness considerably for sensing and imaging. Because of their easy operation, portability, great visibility, and high sensitivity, nano-based and MOF-based sensor approaches are seen as feasible alternatives.

Many comprehensive reviews have been recently published on food safety and quality assessment. This review highlights on strategies for the synthesis and application metal organic framework and magnetic metal organic framework materials for the food safety and food packaging. This review mainly focused on the roadmap from the recent five years for the advancement in materials for the detection of toxic metals, pesticides, antibiotics and mycotoxins in food to assure the food safety and quality.

## **Recent Synthesis Strategies of MOF and Magnetic MOF materials**

### **Synthesis of MOF materials**

To create materials based on the Metal-Organic Framework (MOF) for food safety, porous structures must be created that can absorb and eliminate pollutants from food and packaging. Due to their large surface areas and adjustable pore sizes, these materials are excellent at absorbing chemicals, diseases, and poisons, improving the safety and quality of food. The various synthesis strategies for magnetic MOF-based materials are shown in **Fig.1**.

### **Hydrothermal synthesis**

The hydrothermal (HT) method is a well-known process that directly combines organic ligands with metal clusters at high temperatures to create crystalline metal-organic frameworks (MOFs). In a sealed system, this method entails heating an aqueous solution of the starting components in a high-pressure reactor to temperatures above the boiling point of water. Carefully regulating synthesis factors, including temperature, pressure, reactant concentration, and reaction time, allows for the creation of specific MOF structures (Krongkrachang, Thungngern, Asawaworarit, Houngkamhang, & Eiad-Ua, 2019). The hydrothermal method's adaptability makes synthesizing MOFs with specific pore sizes, shapes, and functions possible, making it perfect for sensing, gas storage, separation, and catalysis. Furthermore, closed systems guarantee high reproducibility and little contamination. The variety of MOF structures is growing due to continuous improvements in hydrothermal synthesis methods, which leads to new materials with enhanced performance (Abdullahi, Harun, & Othman, 2017). Min et al. developed biodegradable pullulan/polyvinyl alcohol (PUL/PVA) nanofibers combined with thymol-loaded porphyrin metal-organic framework nanoparticles (THY@PCN-224 NPs). By producing singlet oxygen ( $^1O_2$ ) in the presence of light, the hydrothermally synthesized PCN-224 demonstrated photodynamic bactericidal activity. A synergistic antibacterial activity against *E. coli*, *S. aureus*, and *Botrytis cinerea* was obtained by integrating thymol into the porous structure of PCN-224. The properties of the nanofibers, their controlled release behaviours, and their biocompatibility were also investigated. This work generally presents THY@PCN/PUL/PVA nanofibers as a novel active packaging material strategy (Min et al., 2021). Riahi et al. developed novel composite film by using red cabbage anthocyanin, copper-based metal-organic frameworks (Cu-MOFs), and gelatin/poly (vinyl alcohol) via hydrothermal method. These films displayed improved tensile strength, water resistance, UV protection antioxidant,

and antibacterial qualities. These films present novel choices for active and intelligent packaging that track the freshness of the food (Riahi, Khan, Rhim, Shin, & Kim, 2023).

### **Solvothermal Approach**

A standard method for creating metal-organic frameworks (MOFs) is the solvothermal (ST) approach, which is similar to the hydrothermal (HT) method. The combination of these two techniques is frequently termed as solvothermal synthesis. The use of organic solvents in solvothermal reactions is the primary way solvothermal and hydrothermal procedures differ. These reactions occur in a sealed container at room temperature and pressure, above the solvent's boiling point. This method improves the crystallinity and structural characteristics of the resultant MOFs by providing exact control over the reaction conditions (Absalan et al., 2024; González, Kharisov, Kharissova, & Quezada, 2021). Furthermore, solvothermal synthesis provides access to a greater variety of chemicals and solvents, increasing the spectrum of MOF structures that can be produced. Khan et al. produced carbon dots (CDs)-doped Ti-metal organic framework (Ti-MOF) by using solvothermal-assisted mechanical stirring and integrated into CMC/Agar-based active packaging sheets. Along with significant antioxidant and antibacterial activity, this CD@Ti-MOF addition increased the film's tensile strength by 17.4% and markedly improved its oxygen permeability, water vapour, and UV-blocking qualities. Cherry tomatoes kept at 4°C had a longer shelf life thanks to the film's efficient inhibition of *L. monocytogenes* and *E. coli* (Khan, Riahi, Kim, & Rhim, 2024). Li et al. synthesized a novel cobalt-based metal-organic framework (Co-ATMP) by using aminotrimethylene phosphonic acid (ATMP) through a simple thermal method and integrated into a polyvinyl alcohol (PVA) matrix to create smart active packaging materials. The Co-ATMP nanocrystal improves the PVA film's mechanical strength, UV-blocking ability, ammonia sensitivity, and antibacterial qualities. By monitoring shrimp spoilage, the films provide an economical method of creating packaging materials that guarantee food safety by measuring freshness in real-time (S. Li et al., 2023). Jafarzadeh et al. employed a solvothermal technique to develop the hydrophobic cerium-based metal-organic framework (Ce-MOF), which was then integrated into a cassava starch matrix to produce active packaging films through solution casting. The film reduced water solubility, hydrophobicity, thermal stability, and crystallinity were all markedly enhanced by the uniform dispersion of Ce-MOF inside the cassava matrix. Cassava/Ce-MOF films offer great potential for application in active food packaging based on these improvements (Jafarzadeh et al., 2024).

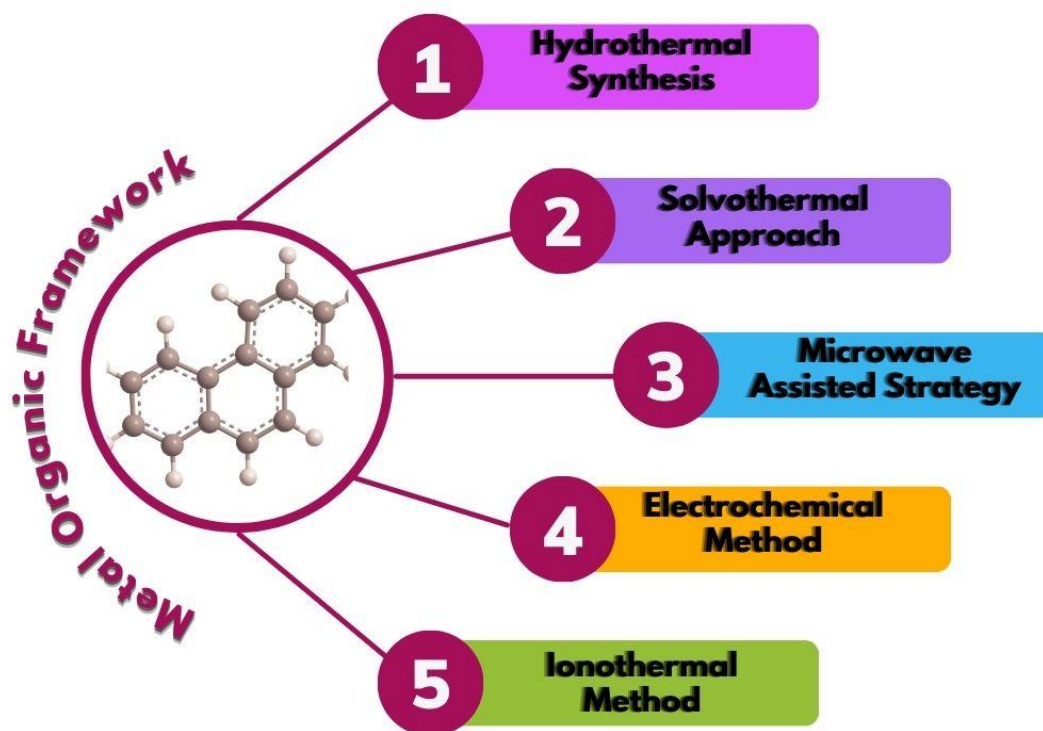
### **Microwave Assisted Strategy**

The microwave (MW)- assisted method, which mixes electromagnetic radiation with mobile electronic carriers, accelerates MOF crystal nucleation and development. In polar liquids, these waves interact with molecules or ions; in solids, they interact with electrons or ions. In both cases, heat is produced, and reaction rates are elevated (M. Li, 2024). Crystal development can be regulated by varying critical parameters such as reactant concentration, reaction time, solvent properties, and energy power. Even little adjustments to these parameters can affect the morphological features of metal-organic frameworks. Compared to conventional heating procedures, this technology offers a distinct reaction environment and dramatically accelerates the synthesis of smaller crystals (Gerbec, Magana, Washington, & Strouse, 2005; Kumar, Kuang, Liang, & Sun, 2020). During MOF synthesis, fast boiling and nucleation rates usually lead to better crystallization, faster manufacturing, and higher purity MOF products. Wang et al. synthesized UiO-66-NH<sub>2</sub> and aptamers, a novel label-free and enzyme-free electrochemical biosensor through microwave-assisted solvothermal method for the isocarbophos (ICP) detection. The biosensor detection limits were 6 ng/mL and 0.9 ng/mL, indicating its great sensitivity. Its efficacy in identifying ICP in fruit and vegetable samples suggests that it may be helpful in applications related to food safety (N. Wang et al., 2022). F.Ayyildiz et al. synthesized cobalt ferrite magnetic nanoparticles via microwave-assisted digestion method for the detection of lead ions from rooibos tea samples. The method demonstrated an 80-fold improvement in detection limits, along with great sensitivity and precision. The outcomes demonstrated that the CoFe<sub>2</sub>O<sub>4</sub> MNPs-based extraction method is an easy, quick, economical, secure, and environmentally friendly way to detect lead in beverage samples accurately (Ayyıldız et al., 2023).

### **Electrochemical Method**

Electrochemical (EC) methods can create metal-organic frameworks (MOFs) by combining an organic linker and a metal salt with an anode and cathode electrode. Compared to conventional limited synthesis methods, this method improves the solid content of the resultant MOFs by continually supplying metal ions from the metal salt (W.-J. Li, Tu, Cao, & Fischer, 2016). The dissolution of metal anodes provides a consistent supply of metal ions that react with the organic linker in the EC medium. Higher porosity and crystalline MOFs are formed due to this constant supply (Ghoorchian, Afkhami, Madrakian, & Ahmadi, 2020). Moreover, the electrochemical technique may create high-pore MOFs even without extra anions. Xiong et al. synthesized a novel molecularly imprinted electrochemical sensor

(MIES) to identify the presence of ciprofloxacin hydrochloride (CPFX) in infant formula. S-CoFe-MOFs (metal-organic frameworks), poly-pyrrole, and gold nanoparticles (AuNPs) are combined to create molecularly imprinted polymers (MIPs). It showed excellent stability and selectivity and relative standard deviations (RSD) below 5.5% and milk sample recovery rates ranging from 89.04% to 110.26% (Xiong et al., 2023).



**Fig.1-Variou synthesis strategies of MOF-based materials.**

### **Ionothermal Method Synthesis**

Ionic liquids (ILs) substitute water or conventional organic solvents in synthesizing metal-organic frameworks (MOFs). Ionic liquids are perfect for chemical synthesis because of their unique qualities, which include great thermal stability, excellent solvating capabilities, zero vapour pressure, and recyclable nature. To neutralize the anionic charge of the framework and serve as a structural template, the IL cation is frequently incorporated into the open cavities of MOFs, facilitating the development of new MOF structures due to the high concentration of both cations and anions in ILs. Furthermore, even if the IL anion is not included in the final MOF structure, ILs can direct MOF formation. For instance, chiral ILs can be used to create homochiral MOFs (Vaid, Kelley, & Rogers, 2017). The ILs has a variety of roles in the synthesis of ionothermal MOFs and in influencing their



structure. Lin Shen and coworkers created 3D fluorescent coordination polymer [PMI]<sub>2</sub>[Eu<sub>2</sub>(BPDC)<sub>3</sub>Cl<sub>2</sub>] by the ionothermal reaction of EuCl<sub>3</sub> with 4,4'-biphenyldicarboxylic acid (H<sub>2</sub>BPDC). This polymer was spread out on a bandage and mixed with PVDF (polyvinylidene fluoride) to create a red-fluorescent composite film. The film exhibits a quick and precise reaction to aniline vapour, suggesting it could be used as an aniline fluorescence turn-off sensor (Shen, Wei, Xu, Liu, & Jiao, 2019).

### **Synthesis of Magnetic MOF-based materials**

Materials based on magnetic MOFs are produced by incorporating magnetic elements into metal-organic frameworks. This synthesis gives the MOFs magnetic characteristics, which increases their suitability for use in food preservation and packaging. The various synthesis strategies for magnetic MOF-based materials are shown in **Fig.2**.

### **Hybrid preparation method**

In the hybrid preparation, metal-organic frameworks (MOFs) and pre-prepared magnetic nanoparticles (MNPs) are directly integrated to generate MMOF composites using high-temperature polymerization or ultrasonic treatment. One may precisely regulate the structure and performance of the resulting MMOFs by modifying the MNPs on the MOFs surface through chemical bonding or electrostatic interactions. The MMOFs characteristics can be precisely controlled by modifying these interactions (Jingying Yang et al., 2020). The surface of the MOFs containing MNPs usually improves the composites' magnetic responsiveness, making them appropriate for various cutting-edge uses, such as electrochemical synthesis. With the addition of magnetic properties, which can be essential for catalyst recovery and reuse in electrochemical processes, this technique preserves the natural porosity and surface area of MOFs. Customized MMOFs are also helpful in various electrochemical reactions since they can show increased catalytic activity and selectivity (Mai, Rafiq, & Yoo, 2017). Due to its adaptability, novel applications in environmental remediation, energy storage, and sensing technologies can be created with materials with synergistic qualities and many functions. Keshavarzi et al. developed a novel and efficient dispersive micro solid-phase extraction method to extract phthalate esters from bottled water and fruit juice. The sorbent, made from ZIF-8 @ ZIF-67, Fe<sub>3</sub>O<sub>4</sub> nanoparticles, and SiO<sub>2</sub>, was optimized and characterized using various techniques. The method showed excellent linearity, low detection limits, and high precision. It successfully measured phthalate esters in real samples with satisfactory recoveries and relative standard deviations (Keshavarzi et al., 2022).

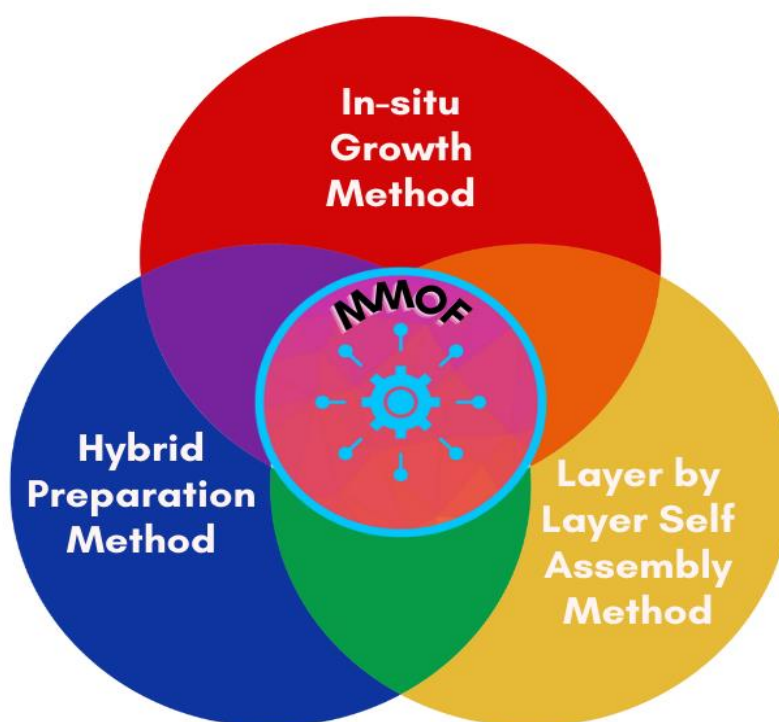
### **In Situ Growth Method**

On magnetic nanoparticles (MNPs), metal-organic frameworks (MOFs) are grown in situ under ultrasonic or hydrothermal conditions directly on the MNP surface within the MOF precursor solution. The magnetic MOFs (MMOFs) produced by this process have good adsorption capability because their structural integrity is guaranteed. MNPs can adhere to the MOF surfaces by dispersing MOFs in a solution containing reagents for MNP production. Although this method has benefits, including improved magnetic responsiveness and adsorption qualities, there are still technical issues. Some free MNPs make the separation process more complex, and maintaining MOF stability is necessary to avoid structural collapse (S.-H. Huo et al., 2017). Future studies must address these concerns to maximize this approach. Shi et al. manufactured a magnetic metal organic frameworks (MOFs) composite called Cu-MOFs/Fe<sub>3</sub>O<sub>4</sub> in order to remove lead (Pb(II)) and malachite green (MG) from wastewater. It is simple to manufacture Cu-MOFs/Fe<sub>3</sub>O<sub>4</sub> by growing Cu-MOFs in-situ and doping them with Fe<sub>3</sub>O<sub>4</sub> nanoparticles. Cu-MOFs/Fe<sub>3</sub>O<sub>4</sub> was found to be an effective adsorbent in the simultaneous removal of MG and Pb(II) in the adsorption studies. Fe<sub>3</sub>O<sub>4</sub>/Cu-MOFs have the potential to be used as an adsorbent for the treatment of waste water because it was discovered that they are reusable for the removal of Pb(II) and malachite green (Shi et al., 2018). Jiang et al. produced CoFe<sub>2</sub>O<sub>4</sub>/porous carbon from waste paper and then employing an in-situ hydrothermal growth technique and created a magnetically bimetallic FeCo-MOF@CoFe<sub>2</sub>O<sub>4</sub>/porous carbon material. This composite combines bimetallic MOFs and magnetic porous carbon. It is useful for removing tetracycline from wastewater and can also be used as a preserver (Jiang et al., 2023).

### **Layer by Layer Self Assembly Method**

The manufacture of nanomaterials has drawn much attention to layer-by-layer (LbL) self-assembly technology, known for its ease of use and effectiveness in surface modification. This has advanced research in material preparation. Using this technique, magnetic nanomaterials' surfaces can be functionalized with the correct chemical groups to prepare metal-organic frameworks (MOFs), which can then be mixed with MOF ligand solutions. This functionalization encourages the formation of stable core-shell structures and the growth of MOF crystal nuclei by facilitating the slow extension and spontaneous assembly of MOF ligands on magnetic nanomaterials (Xiao, Pan, Li, Zhang, & Zhang, 2019). By varying the number of self-assembly cycles under comparatively mild reaction conditions, the LbL self-assembly approach provides fine control over the thickness and characteristics of the

resulting MOFs. Although the process has benefits, it takes a long time to reach the required thickness and is restricted by the types of MOF ligands that are now available, limiting the creation of novel MOFs with different crystal structures. Integrating electrochemical synthesis techniques, which may speed up the assembly process and increase the number of functional MOFs available for various uses, further improves the LbL methods efficiency and adaptability (D.H. Chen, Gliemann, & Wöll, 2023). Liu et al. developed five different structures with different MOF layers in hierarchical  $\text{Fe}_3\text{O}_4@\text{HKUST-1}/\text{MIL-100}(\text{Fe})$  microparticles by layer-by-layer deposition method. Although, displaying distinct surface areas and pore volumes, these hybrids maintain the MIL-100(Fe) catalytic characteristics. Even though these hybrids have fewer layers, their methylene blue (MB) removal effectiveness is equivalent to pure  $\text{Fe}_3\text{O}_4@\text{MIL-100}(\text{Fe})$  material (J. Liu et al., 2019).



**Fig.2-Variou synthetic strategies for magnetic MOF-based materials.**

### **Application MOF-based materials and biosensors for Food Monitoring**

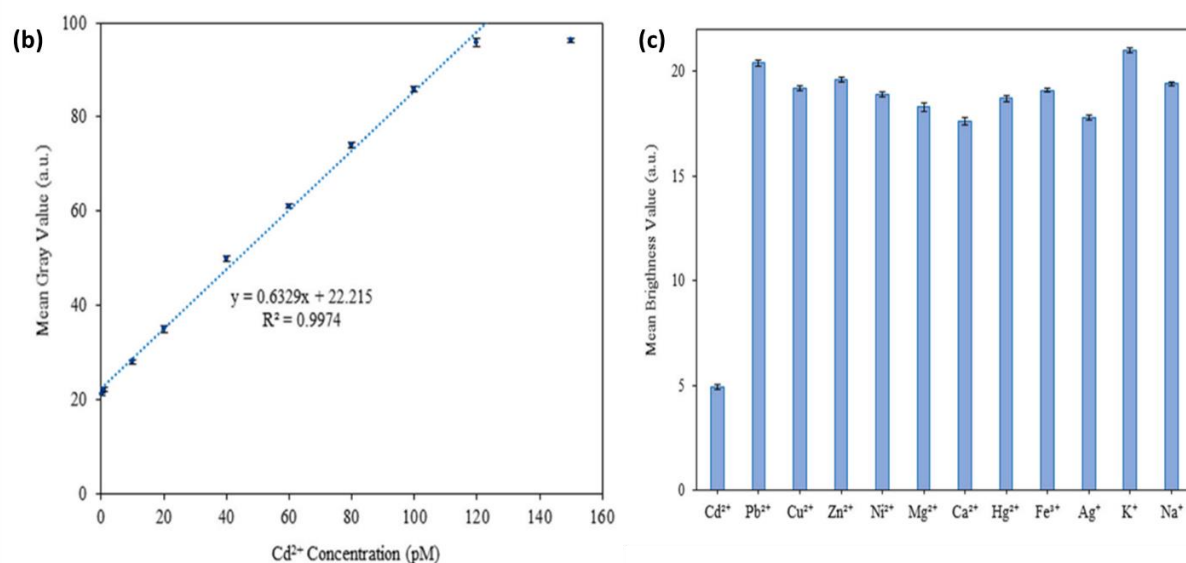
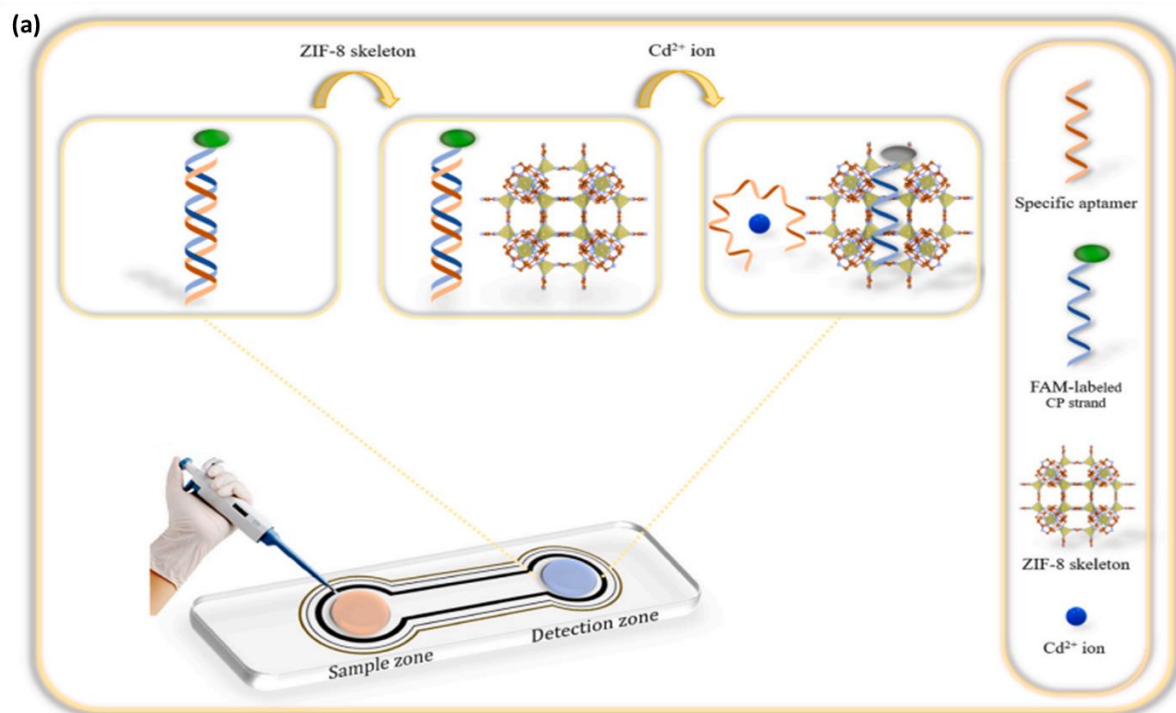
New developments have developed innovative analytical methods for ensuring food safety and quality. Metal-organic frameworks (MOFs) have demonstrated great promise as materials for the food sector because of their special qualities. Better packaging, quality

control, and the elimination of contaminants can all benefit from them (Kalaj et al., 2020; Mehta, Bhardwaj, Bhardwaj, Kim, & Deep, 2016; J. Wang et al., 2021; Yu et al., 2017). Designing MOF-based sensors to improve the assessment of food quality is of particular interest to scholars. Despite continuous research aimed to enhance their effectiveness within that domain, MOF materials have great potential for advancement in food analysis in **Table 1**.

### **Toxic Heavy Metals**

The environment contains trace levels of naturally occurring elements recognized as heavy metals. The wellness of an individual depends on certain heavy metals in small amounts, but larger concentrations can be hazardous and cause serious health concerns. The primary sources of these dangerous metals that are exposed to endangering living things are food and water (Rai, Lee, Zhang, Tsang, & Kim, 2019; L. Wang et al., 2020). It's critical to quickly and often check the levels of heavy metals in the food system to protect consumers from potentially hazardous situations. Pei et al. synthesized UiO-66-NH<sub>2</sub> MOFs by combining it with 3D macroporous carbon (3D-KSC) produced from kenaf stems and further functionalized with an isothiocyanate group to form UiO-66-NHC(S)NHMe/3D-KSC nanocomposites. Their highest Cd<sup>2+</sup>, Pb<sup>2+</sup>, Cu<sup>2+</sup>, and Hg<sup>2+</sup> adsorption capacities were 88, 391, 223, and 935 mg·g<sup>-1</sup>, respectively. They also showed good sensitivity, repeatability, stability, and anti-interference ability. These findings show that UiO-66-NHC(S)NHMe/3D-KSC nanocomposites have good HMI adsorption and detection potential (Pei, Yang, Chen, Wang, & Research, 2022). Zhang et al. developed a Pb<sup>2+</sup>-dependent DNAzyme and a porphyrin-functionalized metal-organic framework (porph@MOF), a novel electrochemical sensor for detecting levels of lead in leafy vegetables. The sensor can be used for periodic lead pollution monitoring because its Pb<sup>2+</sup> detection limit is 5 pM (1 ppt by weight). The procedure is made simpler by its one-step assembly, and precise detection is guaranteed even in the presence of interfering metal ions thanks to the DNAzyme's particular identification of Pb<sup>2+</sup>. This particular sensor provides a quick, accurate, and targeted way to assess if leafy vegetables are contaminated with lead (X. Zhang et al., 2020). Khoshbin et al. used the fluorescence quenching capabilities of zeolitic imidazolate framework-8 (ZIF-8) as shown in **Fig.3 (a)** with a highly sensitive and selective aptamer probe to create a straightforward detection system for ultra-low levels of Cd<sup>2+</sup> ions as shown in **Fig.3 (c)**. Despite a detection limit of 0.076 pM, the paper-supported, portable aptasensor demonstrated a linear response within the range of 0.1–120 pM having calibration curve shown in **Fig.3 (b)**. It identified Cd<sup>2+</sup> in

various samples, such as human blood serum, milk, coffee, and tap water (Khoshbin, Moeenfar, Zahraee, & Davoodian, 2022).



**Fig.3-(a)** Graphical illustration of designed aptasensor to sensitively monitor Cd<sup>2+</sup> ion; **(b)** the calibration curve for Cd<sup>2+</sup> concentration and, **(c)** Selectivity of the aptasensor for the Cd<sup>2+</sup> detection. Reproduced with permission from reference (Khoshbin et al., 2022). Copyright 2022. Elsevier.

Hu et al. designed electrochemical sensor based metal-organic framework based composite for the detection of several metal ions. Two signal markers and a detecting probe were included in the design of the bifunctional MOFs. Target metal ions cause an ion-exchange reaction in which the framework metal-ion centre is replaced by the target ions.

This process produces ratiometric electrochemical signals at various applied potentials. To increase repeatability, stability, and sensitivity, the sensor uses the signal from target metal ions and the  $\text{Cu}^{2+}$  signal from electrically active MOFs as an internal reference point (Hu et al., 2020). Pournara et al. showed that two dimensional metal organic framework named as Ca-MOF can detect and effectively remove heavy metal ions from water. It can remove  $\text{Pb}^{2+}$  traces (about 100 ppb) from wastewater and exhibits strong sorption capacities for both  $\text{Cd}^{2+}$  ( $\sim 220 \text{ mg g}^{-1}$ ) and  $\text{Pb}^{2+}$  ( $\sim 522 \text{ mg g}^{-1}$ ). Furthermore, utilizing anodic stripping voltammetry (ASV), a Ca-MOF sensor detects  $\text{Pb}^{2+}$ ,  $\text{Cd}^{2+}$ ,  $\text{Cu}^{2+}$ , and  $\text{Zn}^{2+}$  at  $\mu\text{g L}^{-1}$  levels. Ca-MOFs dual role as a sensor and sorbent presents environmental monitoring and remediation opportunities (Pournara et al., 2019). Song et al. developed a copper hydroxy phosphate@MOF composite (DMP-Cu) decorated with 2,5-dimercapto-1,3,4-thiadiazole to identify traces of mercury in rice. The technique, which coupled atomic fluorescence spectroscopy with dispersive solid-phase extraction, produced a detection limit of  $0.0125 \text{ ng mL}^{-1}$  and a high adsorption capacity of  $249.5 \text{ mg g}^{-1}$ . Its relatively small standard deviation ( $<6\%$ ) and good recovery (98.8–109%) make it an extremely useful tool for mercury detection (C. Song et al., 2021). Huo et al. synthesized a copper hydroxy phosphate@MOF composite (DMP-Cu) decorated with 2,5-dimercapto-1,3,4-thiadiazole to identify traces of mercury in rice. The technique, which coupled atomic fluorescence spectroscopy with dispersive solid-phase extraction, produced a detection limit of  $0.0125 \text{ ng mL}^{-1}$  and a high adsorption capacity of  $249.5 \text{ mg g}^{-1}$ . Its relatively small standard deviation ( $<6\%$ ) and good recovery (98.8–109%) make it an extremely useful tool for mercury detection (D. Huo et al., 2022).

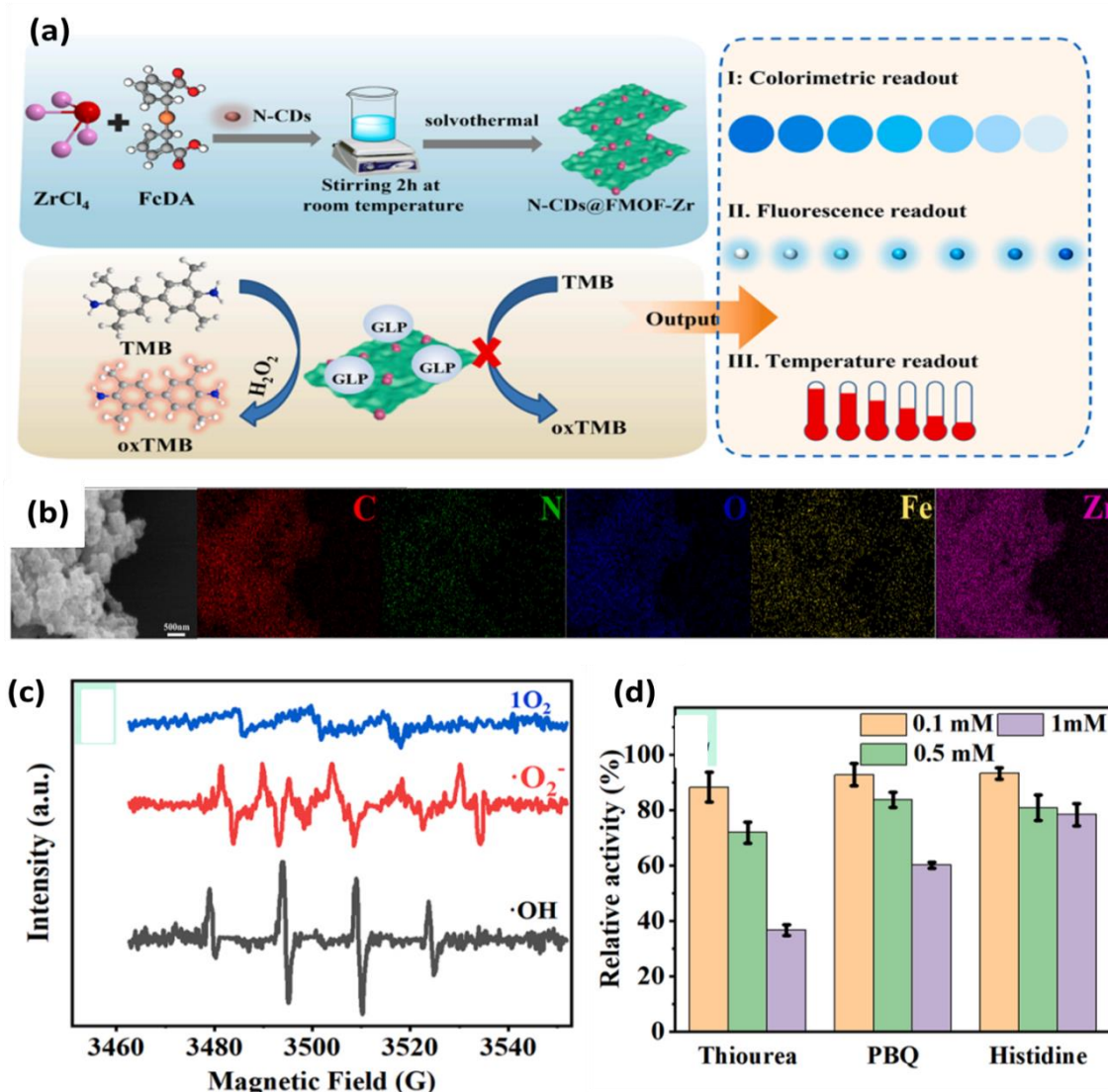
### **Detection of Pesticides and Mycotoxins**

Pesticides are often used to protect crops against weeds, fungi, and pests, but overuse pollutes the environment and raises social and environmental issues worldwide. As a result, the use of MOF-based sensors to find pesticide residues in food is becoming increasingly popular. Strict monitoring and management of residues, as dictated by laws like Maximum Residual Limits (MRLs) across the food supply chain, are necessary to guarantee food safety (Authority, Bellisai, Bernasconi, Brancato, Cabrera, Castellan, Del Aguila, et al., 2022; Authority, Bellisai, Bernasconi, Brancato, Cabrera, Castellan, Ferreira, et al., 2022; Pérez-Fernández, Costa-García, & Muñiz, 2020). Han et al. synthesized a unique three-dimensional Cd(II)-based MOF (WAU-1) by using 2,5-bis(pyrid-4-yl)pyridine and 1,3,5-benzenetricarboxylic acid. A 3D pillar-supported structure with a 6-node pcu-like topology is

formed by WAU-1, according to single crystal X-ray research. Fluorescence quenching is utilized to detect the herbicide pymetrozine, and it functions as an extremely sensitive fluorescent sensor for this purpose. With the use of theoretical computations, the fluorescence quenching process has been suggested (Han et al., 2021). Qing Wang and coworkers introduced a novel composite  $[Zn_3(DDB)(DPE)] \cdot H_2O$  exhibiting a 3D structure and good stability in water at different pH levels. Despite low detection limits, this MOF can identify Fe(III), Cr(III), Cr(VI), Mn(VII), and the herbicide 2,6-Dich-4-NA in aqueous solutions. Even in the presence of surfactants, it retains its detecting powers in actual samples. This was the very first MOF-based probe to find these analytes simultaneously (X.-Q. Wang et al., 2019). Li et al. developed zeolitic imidazolate framework-8 (ZIF-8) and acetylcholinesterase (AChE) by covering methylene blue (MB) by using a unique homogenous electrochemical sensing technique to identify pesticides. In acidic environments, the pH-responsive ZIF-8/MB composites disintegrate, releasing MB and producing a potent diffusion current. Using a 1.7 ng/mL detection limit for paraoxon, this approach demonstrated good sensitivity for detecting organophosphates and carbamates. It also demonstrated efficacy in real samples for environmental and food safety (X. Li et al., 2020). Wang et al. constructed a highly selective immunochromatographic test strip (ITS) for the detection of imidacloprid (IDP) by using Scandium-Tetrakis (4-carboxyphenyl) porphyrin (TCPP) metal-organic framework nanocubes (ScTMNs). ScTMNs were selected due to their great antibody affinity, biocompatibility, and excellent optical features. ScTMNs are produced by combining TCPP as the ligand and  $Sc^{3+}$  as the metal centre. With a detection limit of 0.04 ng/mL, the ITS showed a linear response in the range of 0.04–3 ng/mL and successfully detected IDP in several food samples with excellent recoveries. ScTMNs have never been used in immunochromatography, suggesting they may help detect other materials (Y. Wang et al., 2023). Luo et al. synthesized nanozyme-mediated platform with multiplex signal responses (temperature, fluorescence, and colorimetric) to detect glyphosate (GLP) as shown in **Fig.4 (a)**. An N-CDs/FMOF-Zr nanosensor, which has outstanding peroxidase-like activity, is used in this platform. This activity is suppressed when GLP interacts with the nanosensor, changing the signal responses having EDX elemental mapping, scavenger activity and EPR spectra shown in **Fig.4 (b-d)**. This multimodal sensor is very versatile for real-world applications with sensitive fluorescence detection (X. Luo et al., 2023).

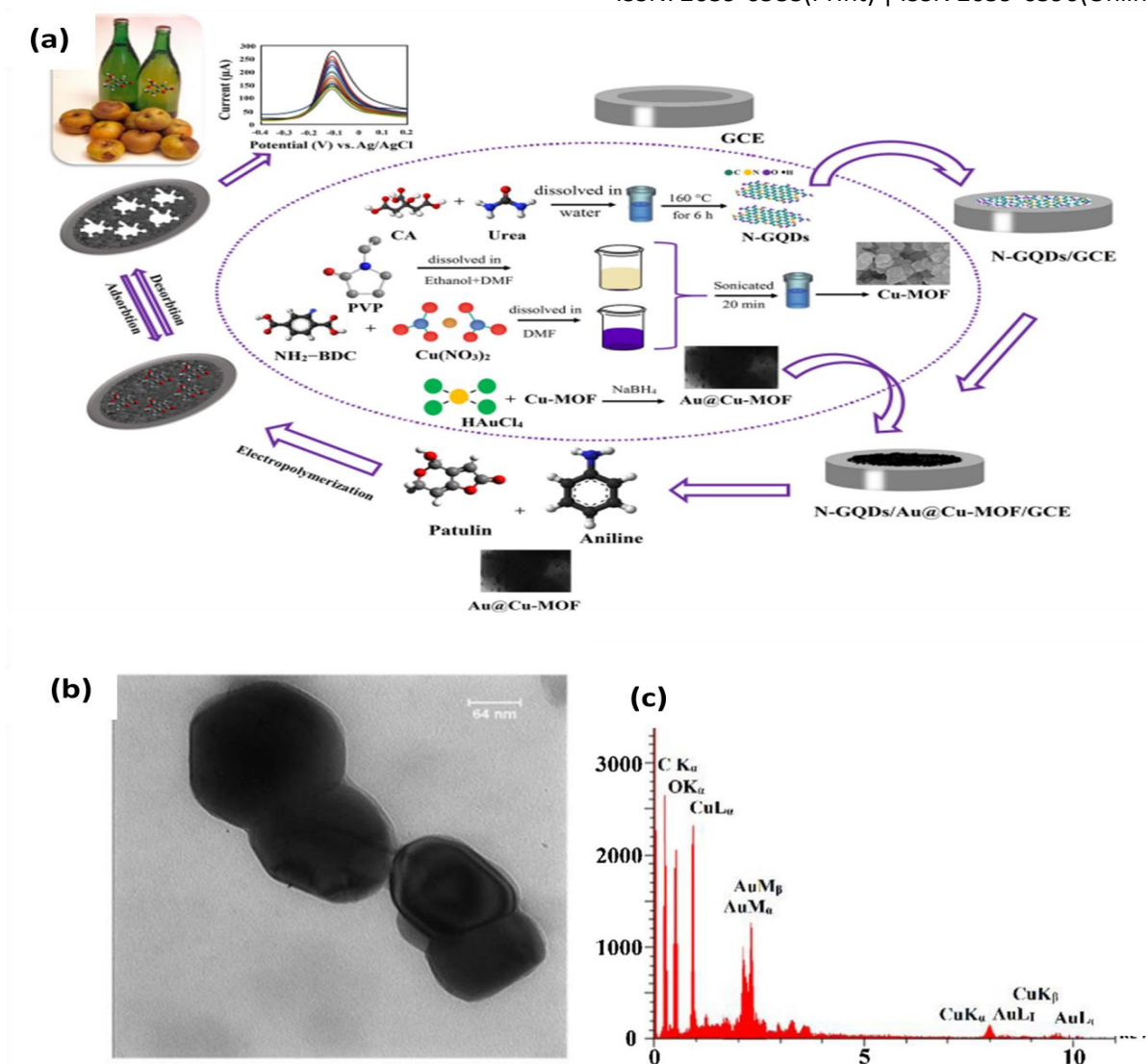
It is crucial to decorate extremely sensitive and selective methods for quick identification of mycotoxins in order to guarantee food safety and quality. Hatamluyi et al.

created electrochemical sensor that is highly selective and sensitive for patulin detection as shown in **Fig.5 (a)**, having transmission electron microscope (TEM) and EDX mapping images depicted in **Fig.5 (b,c)**. The sensor showed a low detection limit and an extensive detection range after being enhanced using nitrogen-doped graphene quantum dot particles as well as gold nanoparticle-functionalized copper organic frameworks. This approach proved to be a cost-effective substitute for chromatographic methods in the analysis of patulin in apple juices, exhibiting good accuracy and precision and great selectivity, sensitivity, stability, and reproducibility (Hatamluyi, Rezayi, Beheshti, Boroushaki, & Chemical, 2020).



**Fig.4-(a)** Schematic illustration for preparation of N-CDs/FMOF-Zr nanozyme; **(b)** EDS element mappings among associated elements of C, N, O, Fe, and Zr in N-CDs/FMOF-Zr composite; **(c)** Comparative scavenger activity on N-CDs/FMOF-Zr + TMB +  $H_2O_2$  (n = 3) and, **(d)** EPR spectra of N-CDs/FMOF-Zr +  $H_2O_2$  system sample. Reproduced with permission from reference (X. Luo et al., 2023). Copyright 2023. Elsevier.





**Fig.5-(a) Schematic illustration for preparation of MIP/Au@Cu-MOF/N-DGQs/GCE and electrochemical detection of Patulin; (b) TEM images of Cu-MOF and Au@Cu-MOF and, (c) EDX image of Au@Cu-MOF. Reproduced with permission from reference (Hatamluyi et al., 2020). Copyright 2020. Elsevier.**

Song et al. produced a range of  $\text{NH}_2\text{-MIL-101(Fe)}@\text{CoPc}$  composites by loading  $\text{NH}_2\text{-MIL-101(Fe)}$  with different concentrations of CoPc (cobalt phthalocyanine nanoparticles). These nanocomposites, which combine the stability and affinity of CoPc with the significant porosity and electrochemical capacity of  $\text{NH}_2\text{-MIL-101(Fe)}$ , were utilized to develop impedimetric aptasensors for the identification of ochratoxin A (OTA). The aptasensor with a 6:1 mass ratio of  $\text{NH}_2\text{-MIL-101(Fe)}$  to CoPc demonstrated the best performance, with a detection limit of  $0.063 \text{ fg}\cdot\text{mL}^{-1}$  and superior selectivity, sensitivity, and repeatability. The potential for OTA detection to enhance food safety is demonstrated by this technology (Y. Song et al., 2021).

**Table 1. Comparison of recent MOF-based materials for detection of heavy metals, pesticides and mycotoxins.**

Category	MOF	Contaminant agent	Detection Method	Application	Limit of Detection	References
Heavy Metals	UiO-66-NHC(S)NHMe/3D-KSC	Cd <sup>2+</sup> Pb <sup>2+</sup> Cu <sup>2+</sup> Hg <sup>2+</sup>	Electrochemical	Water	0.0125 μM 0.0124 μM 0.0111 μM 0.0094 μM	(Pei et al., 2022)
	porph@MOF	Pb <sup>2+</sup>	Electrochemical	Leafy vegetables	5 pM	(X. Zhang et al., 2020)
	ZIF-8	Cd <sup>2+</sup>	Fluorescence	Human blood serum, milk, coffee, tap water.	0.076 pM	(Khoshbin et al., 2022)
	MOFs HKUST-1	Cu <sup>2+</sup> Pb <sup>2+</sup>	Electrochemical	Water	0.002 μM 0.005 μM	(Hu et al., 2020)
	Ca-MOF/GPA	Cd <sup>2+</sup> Pb <sup>2+</sup> Cu <sup>2+</sup> Hg <sup>2+</sup>	Electrochemical	Waste water	1.3 μg L <sup>-1</sup> 0.64 μg L <sup>-1</sup> 1.4 μg L <sup>-1</sup> –	(Pournara et al., 2019)
	DMP-Cu	Hg <sup>2+</sup>	Fluorescence	Rice	0.0125 ng	(C. Song et al., 2021)
	3DGO/UiO-66-NH <sub>2</sub>	Cd <sup>2+</sup> Pb <sup>2+</sup> Cu <sup>2+</sup> Hg <sup>2+</sup>	Fluorescence	Rice and honey samples	1.09e <sup>-8</sup> μM 5.98e <sup>-9</sup> μM 2.89e <sup>-9</sup> μM 3.1e <sup>-9</sup> μM	(D. Huo et al., 2022)
Pesticides	WAU-1	Pymetrozine	Fluorescence	Water	86 mg/L	(Han et al., 2021)
	Zn <sub>3</sub> (DDB)(DPE)·H <sub>2</sub> O	2,6-Dichloro-4-nitroaniline	Fluorescence	Carrot and grapes	0.166 mg/L	(X.-Q. Wang et al.,

						2019)
	ZIF-8/MB	Paraoxon	Electrochemical	Apple and eggplant	1.7 ng/L	(X. Li et al., 2020)
	Sc-TCPP 3D MOF	Imidacloprid	Immunochromatographic strip	Tomatoes, millet, corn and carrot samples	0.04 ng/mL	(Y. Wang et al., 2023)
	FMOF-Zr	Glyphosate	Colorimetric and photothermal	Rice, millet and soybeans	0.0131, 0.0015, and 0.0115 µg/ mL	(X. Luo et al., 2023)
<b>Mycotoxins</b>	MIP/Au@Cu-MOF/N-GQDs/GCE	Patulin	Electrochemical	Fruit juices	0.0007 ng/mL	(Hatamluyi et al., 2020)
	NH <sub>2</sub> -MIL-101(Fe)@CoPc	Ochratoxin A	Electrochemical	Water melon	0.063 fg/mL	(Y. Song et al., 2021)
	CoCoPBA@PCN-221	Deoxynivalenol	Electrochemical	Corn and peanut	0.14 fg/mL	(Cui et al., 2023)

### **Application Magnetic MOF-based materials and biosensors for Food Monitoring**

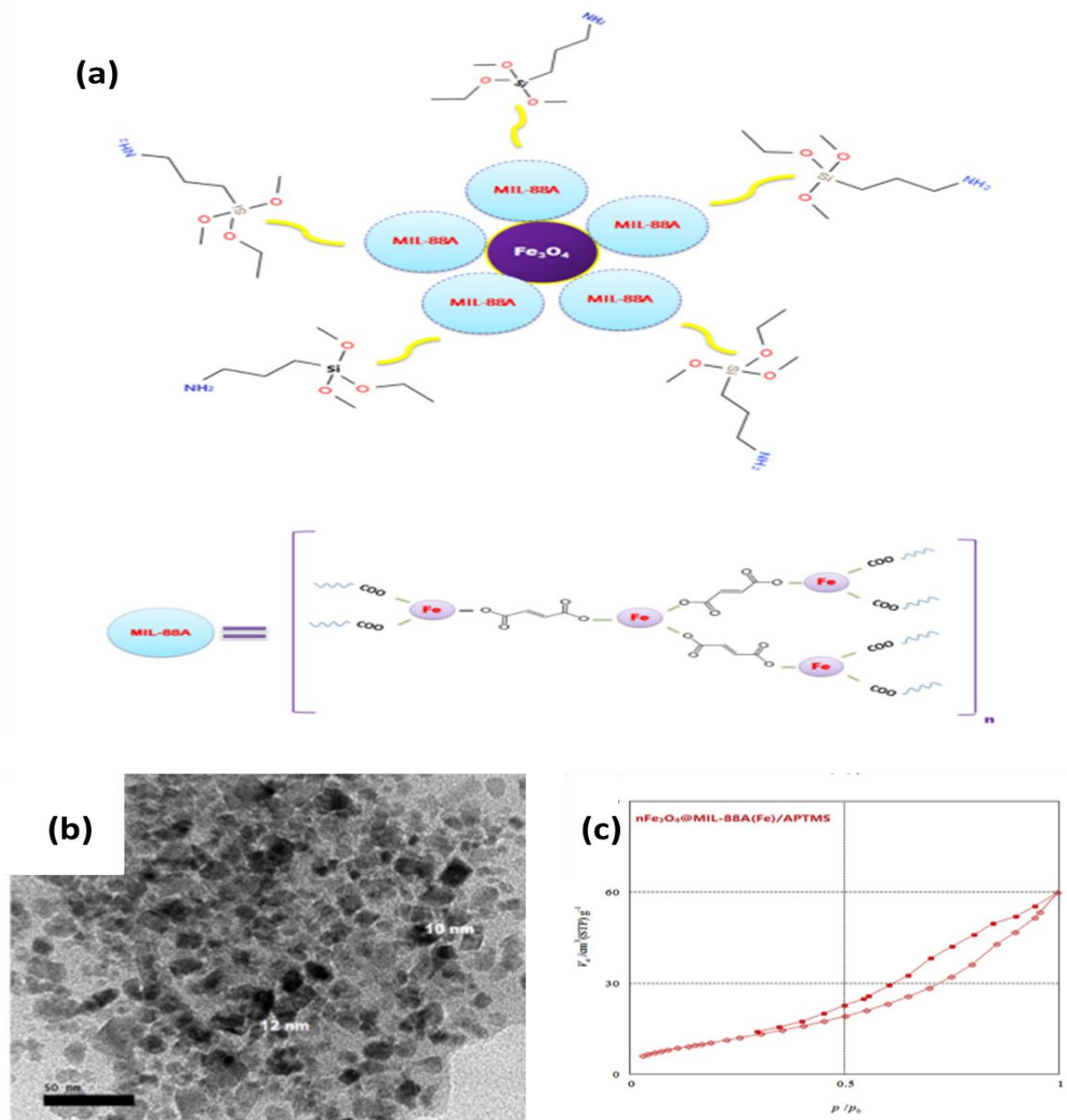
The use of an external magnetic field results magnetic MOFs effectively extracts dispersed adsorbents from large samples while preventing the usage of organic solvents and problems with excessive back pressure and packed bed obstructions. This novel approach shows great potential in the pretreatment of various complicated sample matrices. It is commended for its ease of use, efficacy, and broad applicability in enriching trace analytes. Moreover, MSPEs capacity to optimize sample preparation procedures greatly decreases the environmental effect and operational expenses (Far et al., 2020; Musarurwa, Chimuka, & Tavengwa, 2021). It is a very useful tool for industrial and research applications due to its versatility. Multifunctionalized metal-organic frameworks (MMOFs) integrate the advantageous characteristics of metal-organic frameworks (MOFs), including their huge specific surface areas, exceptional adsorption capabilities, rapid enrichment and separation rates, environmental friendliness, and durability (Xie et al., 2014). By leveraging these qualities, MSPE that employs MMOFs exhibits simple operation and quick, effective enrichment, establishing them as indispensable instruments in the quick pretreatment of environmental and food samples for quality and safety evaluations in **Table 2**.

### **Toxic Heavy Metals and Agrochemicals**

The food business has been impacted by the rise in heavy metal contamination in water and soil caused by increasing production, which poses health problems. The restricted surface areas and stability of conventional adsorption materials, such as zeolite and activated carbon, render them inefficient (Ma et al., 2020; Zare, Motahari, & Sillanpää, 2018). Traditional adsorbents are being replaced by stable and efficient alternatives like metal-organic frameworks (MOFs), which are improving food safety procedures and greatly improving public health protection. Due to their large surface areas, MOFs are a viable solution because they can effectively hold different heavy metal ions (Abolhasani et al., 2015; Esmailzadeh, 2019; Juan Yang et al., 2020). Magnetic metal-organic frameworks can be used to quickly remove heavy metals from samples of food and the environment. Their effectiveness and precision make them indispensable for guaranteeing safety and adherence to established guidelines. Mehraban et al. synthesized a novel nanocomposite by combining MIL-101(Cr) and phenylthiosemicarbazide magnetite nanoparticles ( $\text{Fe}_3\text{O}_4@PTSC$ ) to magnetically extract heavy metals such as  $\text{Cd}^{2+}$ ,  $\text{Pb}^{2+}$ , and  $\text{Ni}^{2+}$  from the samples of seafood and agriculture and analyze the results. The method was verified using a certified reference material and showed outstanding results with detection limits of 0.07–0.5  $\mu\text{g}/\text{kg}$  and high

extraction recoveries (Mehraban, Manoochehri, & Taromi, 2018). E.Mahmoud et al. used a microwave green chemical technique to synthesize a nanocomposite with a  $-\text{NH}_2$  functional group attached by 3-aminopropyltrimethoxysilane (APTMS) as shown in **Fig.6 (a)**. For Cd(II), Pb(II), and Cr(VI) ions, it showed strong adsorption capabilities that suited the Langmuir model and had pseudo-second-order kinetics with TEM images as depicted in **Fig.6 (b)**. With low-efficiency loss after five regeneration cycles and high recovery rates from different water sources, the BET adsorption process as shown in **Fig.6 (c)** was endothermic and spontaneous (Mahmoud, Amira, Seleim, & Mohamed, 2020). Li et al. developed a new magnetic composite  $\text{Fe}_3\text{O}_4@\text{MOF}@\text{COF}$  for preconcentration and selective separation of  $\text{Cu}^{2+}$  ions. Despite a maximum  $\text{Cu}^{2+}$  adsorption capacity of  $37.29 \text{ mg g}^{-1}$ , it exhibits good efficiency and is readily separated using a magnet. The efficiency of the composite is shown by catalysis and  $\text{Cu}^{2+}$  detection using UV-vis spectrophotometry, which yields a 37.6 nM detection limit (W.-T. Li et al., 2020).

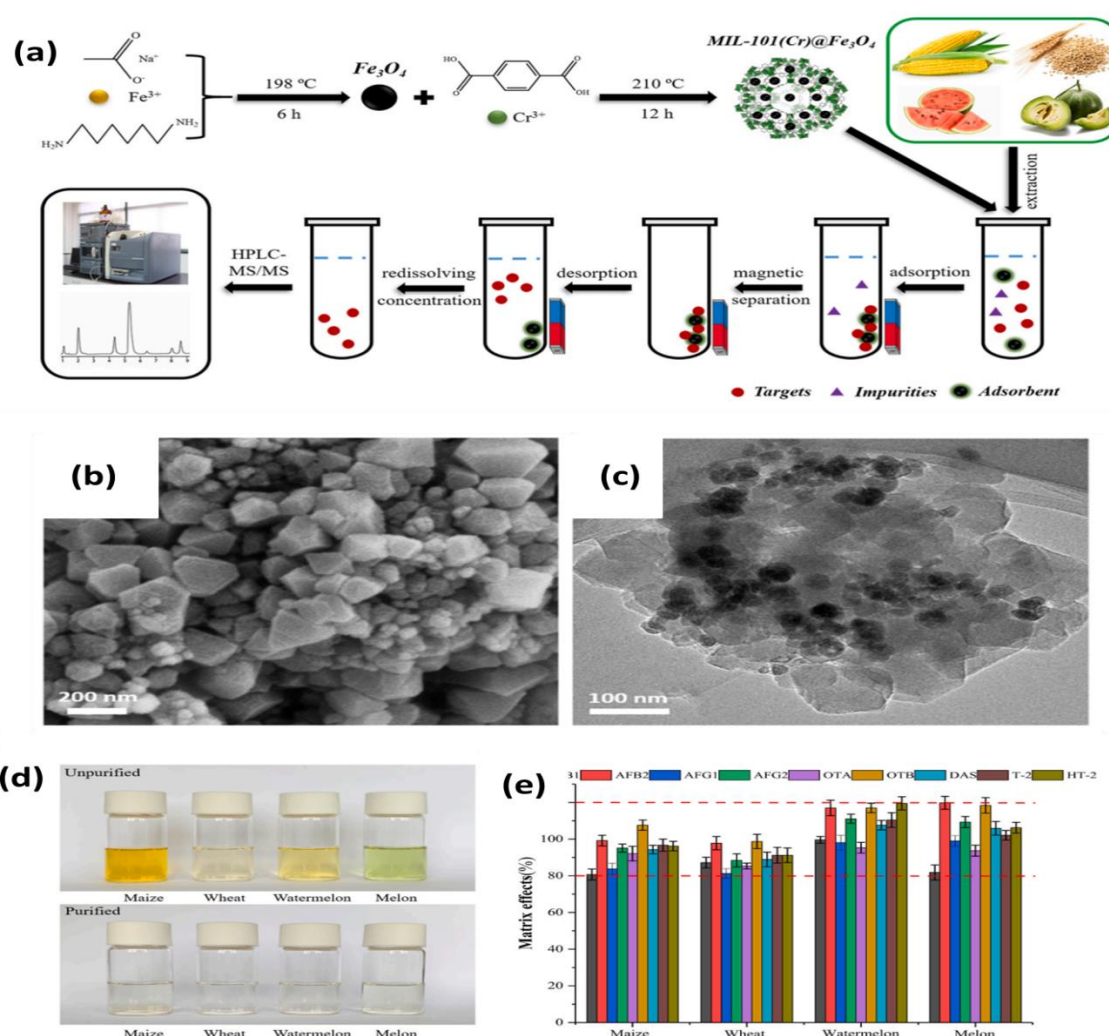
The widespread nature of pesticide residues and their possible health hazards make them a serious concern in food. When they enter the body through food consumption, these toxins can induce cumulative toxicity, cause different allergic reactions, or cause diseases (Jadhav et al., 2019). Pesticides are necessary for controlling diseases and pests in agriculture, but their misuse and overuse have led to severe contamination. Several tons are utilized annually in China alone, but very little of it manages pests efficiently. Most pesticides become residues in soil, plants, or groundwater, contaminating food and surrounding areas (Monteiro et al., 2020; Zhou, Zou, Song, Chen, & Chemistry, 2018). Liu et al. developed a novel magnetic copper-based metal-organic framework (M-MOF-199) nanocomposite to recover triazole insecticides from water samples. This research demonstrated recoveries of 72.3%–91.53% and detection limits of 0.05–0.1, respectively. Triazole insecticides have the potential to be effectively removed from environmental water using M-MOF-199 as an adsorbent (G. Liu, Li, Huang, Zheng, Xu, Xu, et al., 2018). Liu et al. synthesized the magnetic multi-walled nanotubes (M-M-ZIF-8) and adsorbent ZIF-8 to remove organophosphorus pesticides from soil and water samples. Despite a porous structure and large specific surface area, M-M-ZIF-8 demonstrated strong adsorption capacities and fit the Freundlich model. M-M-ZIF-8 is viable for removing environmental pollutants since the adsorption mechanism incorporates valence-electron-driven interactions (G. Liu, Li, Huang, Zheng, Xu, Liu, et al., 2018).



**Fig.6-(a) Proposed structure of  $n\text{Fe}_3\text{O}_4@MIL-88A(Fe)/APTMS$ ; (b) TEM images of  $n\text{Fe}_3\text{O}_4@MIL-88A(Fe)/APTMS$  and, (c) BET  $\text{N}_2$  adsorption–desorption onto different MOFs. Reproduced with permission from reference (Mahmoud et al., 2020). Copyright 2020. Elsevier.**

Mycotoxins are dangerous byproducts of fungal metabolism. They are frequently present in oily substances, grains, and other food items. Mycotoxins can appear at any point during food manufacture, processing, storage, or transportation. Consuming meals high in mycotoxins can be extremely harmful to a person's health (El-Sayed, Jebur, Kang, & El-Demerdash, 2022). The complex nature of food matrices and the low quantities of these toxins make detecting mycotoxin contamination in food difficult. However, the Magnetic

MOFs offers a very effective way to purify using metal-organic frameworks. Guo et al. synthesized MIL-101(Cr) $@$ Fe<sub>3</sub>O<sub>4</sub> nanocomposites, a fast magnetic solid phase extraction (MSPE) technique to concentrate and purify nine mycotoxins found in agricultural products as shown in **Fig.7 (a)**. The technique demonstrated outstanding performance in terms of precision, good recovery, and high sensitivity. The synthesized nanocomposites morphological properties can be visualized through their SEM and TEM images as given in **Fig.7 (b,c)** along with appearance features in **Fig.7 (d)**. It eliminated matrix effects, enabling precise quantification using tandem mass spectrometry and ultra-high-performance liquid chromatography as depicted in **Fig.7 (e)**. Natural watermelon, melon, wheat, and maize samples were used to validate the procedure (Guo et al., 2023).



**Fig.7-(a)** Schematic illustration of magnetic solid phase extraction process based on MIL-101(Cr) $@$ Fe<sub>3</sub>O<sub>4</sub> nanocomposites; (b,c) SEM, TEM images of MIL-101(Cr) $@$ Fe<sub>3</sub>O<sub>4</sub> and, (d,e) Appearance features and matrix effect of mycotoxins. Reproduced with permission from reference (Guo et al., 2023). Copyright 2023. Elsevier.

**Table 2. Comparison of recent magnetic MOF-based materials for detection of heavy metals, pesticides and mycotoxins.**

Category	Magnetic MOF	Contaminant agent	Detection Method	Application	Limit of Detection	References
<b>Heavy Metals</b>	Fe <sub>3</sub> O <sub>4</sub> @PTSC[MIL-101(Cr)]	Cd <sup>2+</sup> Pb <sup>2+</sup> Ni <sup>2+</sup>	Flame atomic absorption spectroscopy (FAAS)	Agriculture and seafood samples	0.07–0.5 µg kg <sup>-1</sup> 0.2–2.0 µg kg <sup>-1</sup> 0.25–250 µg kg <sup>-1</sup>	(Mehraban et al., 2018)
	nFe <sub>3</sub> O <sub>4</sub> @MIL-88A(Fe)/APTMS	Cd <sup>2+</sup> Pb <sup>2+</sup> Cr <sup>6+</sup>	Atomic absorption spectroscopy (AAS)	Tape water, sea water and waste water	– – –	(Mahmoud et al., 2020)
	Fe <sub>3</sub> O <sub>4</sub> @MOF@COF	Cu <sup>2+</sup>	UV-Visible spectroscopy	Water samples	37.6 nM	(W.-T. Li et al., 2020)
<b>Pesticides</b>	M-MOF-199	triazole	HPLC–MS/MS	Environment water samples	0.05–0.1 µg/L	(G. Liu, Li, Huang, Zheng, Xu, Xu, et al., 2018)
	M-M-ZIF-8	organophosphorus	AAS	Environment water and soil samples	–	(G. Liu, Li, Huang, Zheng, Xu, Liu, et al., 2018)
<b>Mycotoxins</b>	MIL-101(Cr)@Fe <sub>3</sub> O <sub>4</sub>	OTB, DAS, AFG-1, AFG-2, T-2, OTB, HT-2, AFB-1 and AFB-2	HPLC–MS/MS	Water melon, maize, melon and wheat samples	0.08–0.20 µg kg <sup>-1</sup>	(Guo et al., 2023)



## Conclusions and Future Perspectives

The distinctive properties of MOF and magnetic MOF-based materials, such as their high porosity, extensive surface area, controlled structure, and increased binding capacity, make them useful in various applications, particularly food protection and packaging. These materials have shown great potential as contaminant sensors due to their exceptional sensitivity, wide linear range, excellent efficiency, and extraordinary stability. Recent developments in MOF-based sensors demonstrate how well they can identify different food pollutants. Since conducting MOFs have many active metal sites and tunable porous architectures that allow for quick electron transfer, they are very good at electrochemical sensing when utilized as electrode platforms. Nevertheless, problems with poor recycling ratios, unstable modified electrodes, weak stability, and challenging contamination detection in intricate food matrices continue to arise worldwide. In addition, the synthesis techniques for MOF-based sensors require exact control over the size, shape, and dispersion of active metals to enhance surface area and homogeneity. It is necessary to improve MOF stability in aqueous and unfavorable conditions, perhaps by coordinating high-valence metal ions with hydrophobic ligands. Furthermore, pure MOFs often have low conductivity, which makes it difficult to use them directly in sensors, particularly as electrode material. Future studies should concentrate on developing more resilient MOF transducers with superior sensitivity, selectivity, and reusability to overcome these difficulties. It is crucial to create novel synthesis techniques to regulate the size, shape, and dispersion of active metals on MOF surfaces as well as to increase MOF stability. Despite the promised advances in food safety and packaging that MOF and magnetic MOF-based sensors show, resolving these issues will increase their usefulness and commercial viability.

## Conflict of Interest

There is not any form of conflict of interest

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