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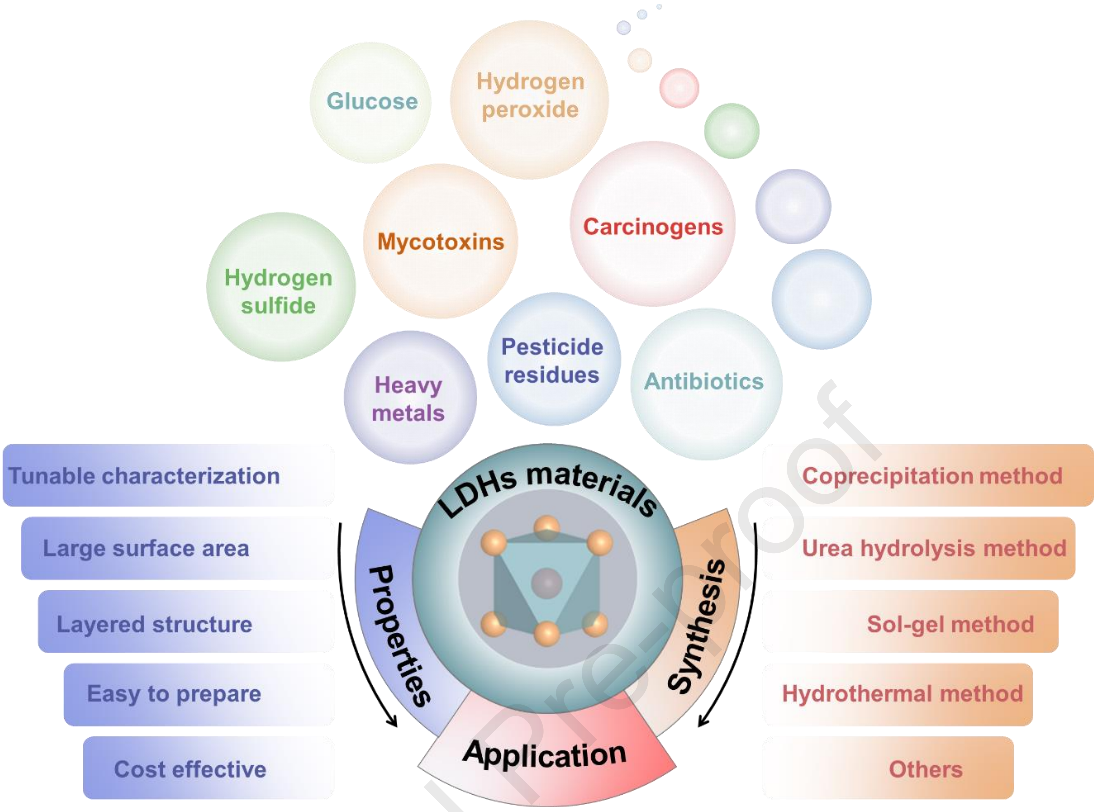
Graphics



Scheme 1. The properties, synthesis methods, and applications of LDHs.

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Abstract

Background**:** Inadequate or untimely food safety detection and monitoring technologies often lead to substantial resource and cost wastage. Consequently, the development and implementation of portable, rapid, and intelligent technologies have become critical to ensure food safety and the preservation of product quality. Layered double hydroxides (LDHs), due to their large specific surface area, excellent stability, simple synthesis methods, and low cost, have garnered significant attention from researchers in recent years. LDHs have emerged as an ideal choice for achieving high sensitivity and strong specificity in food safety sensing and detection.

Scope and approach**:** A detailed overview of the fundamental concepts of LDHs is provided, accompanied by a comprehensive analysis of their synthesis methods. Next, the sources of catalytic activity in LDHs are explored, and strategies for regulating their performance are proposed to enhance their effectiveness in food contaminant detection. Finally, the progress in practical applications of LDHs for detecting various food contaminants is reviewed, with an emphasis on their effectiveness in improving detection sensitivity, increasing detection efficiency, and facilitating the development of portable and intelligent detection systems.

Key findings and conclusion**:** LDHs can be applied in the detection of heavy metals, pesticide residues, mycotoxins, and antibiotics. However, LDHs still face certain limitations in achieving high sensitivity and strong specificity in detecting target substances. One promising strategy to overcome these challenges is the synergistic incorporation of high-performance catalytic materials, such as single- or dual-atom catalysts and other nanomaterials. This development not only offers new perspectives for advancing food safety detection technologies but also expands the possibilities for broader applications of LDHs.

Keywords：Layered double metal hydroxides; Food contaminant detection; Food safety; Intelligent detection.

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1. Introduction

Food safety remains a critical global concern, as it directly impacts human health and quality of life. With the growing complexity of globalization and food supply chains, the precise and efficient detection of contaminants such as heavy metal ions, pesticide residues, carcinogens, and antibiotics has become critically important. In recent years, food safety detection technologies have advanced rapidly, incorporating methods from traditional laboratory analyses to novel sensor-based detection approaches. These advancements have not only optimized the ability to meet practical detection needs and minimized resource and cost wastage, but have also significantly improved detection efficiency, enabling rapid, convenient, and intelligent monitoring of food contaminants. ~~These advancements provide strong support to ensure food safety and quality.~~ However, developing detection technologies capable of achieving high sensitivity and specificity in detecting target substances remains a significant challenge.

Materials are frequently regarded as the ‘cornerstone of human civilization’. The research, development, and application of novel high-performance chemical materials have long been central to interdisciplinary scientific inquiry (Tan, Yi, & Zhao, 2024). In recent years, materials such as organic and inorganic nanomaterials, two-dimensional (2D) materials (e.g., graphene, transition metal dichalcogenides) (Ashiq, Saeed, Li, & Nawaz, 2024; Z. L. Lei & Guo, 2021) and metal-organic frameworks (Jian-Hua, et al., 2020) have garnered significant attention.

Layered double hydroxides (LDHs), are novel functionalized 2D materials that stand out due to their unique layered structure (single or multi-layer), simple preparation process, high surface area (Taviot-Gueho, et al., 2018), good biocompatibility (Jingfang Yu, Wang, O'Hare, & Sun, 2017), and excellent catalytic activity. These advantages have led to their widespread application in fields such as environmental monitoring, food safety detection, and biomedicine (R. Li, et al., 2023; Q. Liu, et al., 2017). Keyword clustering analysis of papers published over the past decade revealed that numerous countries have conducted extensive and systematic research in areas such as food contaminant detection, nano-sensors, and LDHs (Kumari,

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Soni, Sharma, Kumar, Sharma, Jaswal, Bhatia & Sharma, 2024; Sohrabi, Arbabzadeh, Falaki, Majidi, Han, Yoon & Khataee, 2022) (Figure 1A-C). Countries such as China, the United States, and Japan are at the forefront of these research efforts (Figure 1D-F). Additionally, over the last 10 years, the research directions for LDHs have become increasingly diversified and decentralized. Initially concentrated in the field of chemistry, the scope has gradually expanded to intersect with biology, physics, environmental science, and food science, showing an increasingly strong development trend (Figure 1G). This indicates the significant research importance of LDH-based detection technologies in the field of food contaminant detection (Stanley, Sriram, Wang, Sherlin V, Kogularasu & George, 2025; Muthukumaran, Govindaraj, Kogularasu, Siriram, Raja, Wang, Chien & Selvi J, 2024; Sakthivel, Balasubramanian, Chien, Wang, Ahammad, Billey & Platero, 2024; Stanley, Sriram, Wang, Sherlin V, Kogularasu, Lee & Geotge, 2024; Kogularasu, Sriram, Wang & Sheu, 2022).

This study focuses on the research applications of LDHs in food contaminant detection. It systematically reviews the advantages and disadvantages of various LDHs synthesis strategies, the sources of their catalytic activity, the methods for regulating their activity, and their applications in detecting various food contaminants such as heavy metals, pesticide residues, mycotoxins, and antibiotics. Additionally, the study evaluates the opportunities and challenges associated with using LDHs in food contaminant detection. The overarching aim is to provide a solid research foundation for the development of rapid, portable, and intelligent detection technologies that can meet the increasingly complex food safety monitoring needs.

2. Definition, synthesis, and functional properties of LDHs

The synthesis process is a crucial step in determining the physical and chemical properties of materials. A clear understanding of the definition and basic synthesis procedures of LDHs is essential for the subsequent development of cost-effective, green, and sustainable eco-friendly synthesis methods. Furthermore, a deeper insight into their structural characteristics provides an important theoretical foundation for elucidating the functional properties of LDHs.

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2.1. Definition of LDHs

In 1842, the Swedish scientist Crica first discovered LDHs in natural fiber materials (Cui, Jiao, Zhao, & Zhou, 2012). In 1942, LDHs were synthesized successfully in the laboratory for the first time, leading to the proposal of the concept of a layered structure model. It was not until 1969 that this layered structure was formally confirmed through single-crystal diffraction techniques (P. Wang, et al., 2023). With the development of technology, researchers have gained a deeper understanding of the concept and structure of LDHs. LDHs, also known as hydrotalcite, hydrotalcite-like materials, or anionic clays, represent a class of layered compounds with a 2D nanoscale structure. They consist of positively charged metal hydroxide layers and intercalated anionic species that balance the overall charge. The main structural layers are formed by the coordination of different metal ions (Kumari, et al., 2022). Compared with other layered materials, the fundamental structure of LDHs is characterized by a layered framework formed by the alternating arrangement of divalent and trivalent metal ions. The specific structure is depicted in Figure 2, consisting primarily of positively charged metal hydroxide layers and intercalated anions. The general chemical formula is expressed as: $[M_{1-x}^{2+}M_x^{3+}(OH)_2]^{x+}A_{x/n}^{n-}\cdot zH_2O$, where M^{2+} typically represents divalent metal cations such as magnesium (Mg^{2+}), cobalt (Co^{2+}), nickel (Ni^{2+}), zinc (Zn^{2+}), and copper (Cu^{2+}) (R. Li, et al., 2023). M^{3+} represents trivalent metal cations such as aluminum (Al^{3+}), chromium (Cr^{3+}), iron (Fe^{3+}), and gallium (Ga^{3+}) (Q. Wang & O'Hare, 2012; H. Wu, et al., 2024) (Figure 3A). A^{n-} denotes anions, including inorganic and organic ions such as nitrate (NO_3^-), chloride (Cl^-), hydroxide (OH^-), sulfate (SO_4^{2-}), phosphate (PO_4^{3-}), and $C_6H_4(COO)_2^{2-}$, as well as complexed ions (M. Xu & Wei, 2018). Moreover, x refers to the molar ratio of $M^{3+}/(M^{2+}+M^{3+})$, typically ranging from 0.2 to 0.4 (Fan, Li, Evans, & Duan, 2015); and z indicates the number of crystallization water molecules bridging the interlayer, typically associated with the metal hydroxide and hydrogen bonding within the LDHs structure (Y. Li, et al., 2022). Among the various structures currently available, the most typical chemical composition of LDHs is $Mg_6Al_2(OH)_{16}CO_3\cdot 4H_2O$, commonly referred

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to as MgAl-LDHs. Furthermore, in LDHs constructed from different divalent and trivalent metal ions, the corresponding band potentials vary, with MgAl-LDHs exhibiting the highest band potential (Figure 3B). This variation in band potentials provides a theoretical foundation for their applications in diverse fields. Therefore, a clear understanding of the concept and structure of LDHs is crucial, as it lays the essential groundwork for further elucidating their synthesis strategies and material properties.

2.2. Strategies for the synthesis of LDHs

The main methods used to synthesis LDHs include coprecipitation, hydrothermal, urea hydrolysis, sol-gel, template, interlayer anion exchange, and electrodeposition (Ge, 2016; Inayat, Klumpp, & Schwieger, 2011; Sangtam, Saikia, Goswamee, Sinha, & Sinha, 2022; P. Wang, et al., 2023; J. Zhang, Yu, Chen, & Lou, 2020; Wei Zhang, Wu, Qi, Chen, & Cao, 2017; Zhong, et al., 2019) (Figure 4 and Table 1).

2.2.1. Coprecipitation method

The coprecipitation method is widely regarded as the most commonly employed and straightforward one-pot synthesis approach for LDHs (Chengqian, et al., 2022). It can be further divided into the variable pH method (Song, et al., 2023) and the constant pH method (Kou, Li, Zhao, Wang, & Ma, 2018). In this process, a metal ion-containing solution is mixed with a precipitating agent (such as sodium hydroxide or ammonia solution), and the reaction conditions are controlled to facilitate the precipitation of metal hydroxides. The formation of LDHs is then promoted by adjusting the pH (typically in the range of 10-11) and temperature. The pH value plays a crucial role throughout the coprecipitation process; an excessively high or low pH value can hinder the successful coprecipitation (Figueiredo, et al., 2022). Despite its advantages, this method exhibits certain limitations, including the localized over-concentration of the precipitant, non-uniform particle size, and poor crystallinity.

2.2.2. Urea hydrolysis method

The urea hydrolysis method is an improvement upon the conventional coprecipitation method and enables the synthesis of LDHs with a uniform particle size

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distribution and high crystallinity. This approach is also referred to as uniform coprecipitation (Staal, et al., 2017). In this method, this approach employs urea as a substitute for traditional alkaline solution. Urea is highly soluble in water, and at low temperatures, forms a neutral solution with metal ions, ensuring a homogeneous mixture. As the reaction temperature rises to 70-100°C, urea undergoes hydrolysis, releasing ammonia gas, which promotes the precipitation of metal hydroxides. By controlling the reaction temperature, the hydrolysis rate can be easily adjusted. Moreover, the uniform presence of urea in the solution ensures that the components precipitate homogeneously. However, this method also has certain limitations. During urea hydrolysis, carbon dioxide is generated, which dissolves in water to form carbonate (CO_3^{2-}) ions. These ions have a strong affinity for the LDHs layers, making it difficult to substitute them with other anions. As a result, LDHs synthesized via urea hydrolysis typically contain CO_3^{2-} within their layered structures (P. Wang, et al., 2023), which limits their applicability in subsequent detection and application processes.

2.2.3. Hydrothermal method

The hydrothermal method represents a highly innovative approach for synthesizing LDHs by facilitating the reaction of metal salts with a base. Although the yield of this method is lower than that of coprecipitation, it offers superior control over the crystal structure, morphology, and crystallinity (Tang, Mu, Zong, & Wang, 2018). LDHs synthesized through the hydrothermal method typically exhibit high crystallinity and a uniform morphology and size, making it the preferred technique for obtaining high-quality LDHs. This method is suitable for the synthesis of LDHs with various structures and compositions (Nguyen, Uyen, Nguyen, Tran, & Nguyen, 2020). Furthermore, the reaction can be conducted at elevated temperatures and pressures to promote crystallization, while minimizing the loss of reactants due to volatilization. However, the hydrothermal method also has certain drawbacks. For instance, it often requires specialized high-pressure, temperature-controlled equipment, which is expensive and requires professional maintenance. Additionally, the method consumes a large amount of energy to maintain the high temperature and pressure conditions,

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leading to high energy consumption. This high energy demand not only imposes significant economic cost but also conflicts with contemporary green and circular economy strategies, thus limiting the method’s broader applicability.

2.2.4. Sol-gel method

The sol-gel method is an advanced technique for synthesizing LDHs, integrating both sol chemistry and the gelation process (Sokol, et al., 2019). The basic procedure involves hydrolysis, precipitation, washing, and drying steps, during which metal precursors are dissolved in an appropriate solvent, typically containing metal salts such as nitrates or chlorides. Then, the gelation process is initiated by adjusting the pH value and adding precipitants, such as sodium hydroxide or ammonia solution, which transform the sol into a gel network. Subsequent heat treatment (such as drying or calcination) is performed to obtain the LDHs (Rahman, et al., 2024). The sol-gel method offers excellent controllability and is capable of synthesizing high-purity, large surface area, and highly uniform LDHs at relatively low temperatures (Valeikiene, et al., 2020). Furthermore, by adjusting parameters such as the precursor concentration, the type of precipitant, and the reaction conditions, the properties of the final product can be precisely tailored (Murath, 2019). This method is particularly suitable for the preparation of complex multi-metal LDHs and for specific functional modifications, thus catering to a wide range of application needs.

2.2.5. Template method

The template method for synthesizing LDHs involves the use of template materials to guide the formation of specific morphologies and structures, thereby enabling precise control over the resulting structure (Moloudi, et al., 2023). The process begins by selecting an appropriate template, such as nanospheres, microporous materials, or self-assembled organic molecules. The surface or pores of the template are then immersed in a solution containing metal ions, and a precipitation reaction is carried out under strictly controlled conditions to form the precursor layered structure of the LDHs. Finally, the template is removed through physical or chemical methods (such as acid etching or solvent dissolution), yielding LDHs with the desired shape and structure.

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The distinctive feature of the template method lies in its ability to precisely control the morphology, pore structure, and dispersibility of the LDHs by adjusting the shape and size of the template material, thus enabling highly customized material design (Jiang, et al., 2024). This approach is particularly suitable for the preparation of catalysts, energy storage materials, and sensors with complex morphologies and high performance. However, improper selection of templates or incomplete removal can lead to defects in the final product, such as uneven pore size distribution, structural instability, or contamination.

2.2.6. Interlayer anion exchange method

The interlayer anion exchange method is one of the key techniques for synthesizing LDHs. It utilizes the ion-exchange reaction characteristics to introduce various cations into the layered structure, thereby adjusting and optimizing the properties of LDHs (Z. Xu, et al., 2024). Initially, the base LDHs are synthesized by using conventional methods, typically containing metal ions and hydrated interlayer anions. Next, these LDHs are immersed in a solution containing different cations for ion-exchange. The target cations replace the original interlayer anions and become embedded within the interlayer regions of the LDHs, thereby altering their chemical composition and functional properties (Sotiles, Baika, Grassi, & Wypych, 2019). By controlling the ion-exchange conditions (i.e., the time, temperature, and ion concentration), the structure and performance of LDHs can be regulated precisely (P. Wang, et al., 2023). However, the ion-exchange method faces challenges in practical applications, including high costs, poor stability, and strong environmental dependence.

2.2.7. Electrodeposition method

The electrodeposition method is an effective technique for synthesizing LDHs; it involves the deposition of metal precursors onto a substrate surface to construct the layered structure. This method primarily includes chemical vapor sedimentation and electrochemical sedimentation (Ho, et al., 2020). In the former process, gaseous precursors react with the substrate at high temperatures to form thin-film LDHs, a technique that enables the preparation of high-purity, uniform, and large-area materials.

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In electrochemical deposition, an electric current is applied in an electrochemical cell, including the reduction of metal ions and their subsequent deposition onto the electrode surface to form LDHs (J. Wang, et al., 2022). This method is simple to operate, cost-effective, and allows for precise control over the deposition rate and thickness. Additionally, the key to the deposition method lies in the regulation of reaction conditions, including temperature, pressure, and solution composition, all of which directly influence the structure and properties of the LDHs. Furthermore, the optimization of these parameters enables the synthesis of LDHs with tailored interlayer anions and crystal morphologies, meeting specific application requirements.

3. The activity source and performance regulation of LDHs

The catalytic activity of LDHs is primarily attributed to their unique layered structure and tunable interlayer composition, which specifically manifests in three aspects: metal ion activity, interlayer anion activity, and surface/interface activity. Based on this, we can further achieve precise performance modulation through the control of catalytic activity sources (Figure 5).

3.1. The activity sources of LDHs

Metal cations, anions, and surface-active groups in the layered structure of LDHs themselves contribute greatly to their catalytic performance (Lijiao, et al., 2017; Lijiao Ma, et al., 2017; L. Ma, et al., 2016; Wang, Dengfeng, Zhang, & Xuelan, 2015; D. Wang, Zhang, Cong, Liu, & Zhou, 2018; H. Wang, Bing, Chen, Yang, & Wei, 2020).

3.1.1. Metal ion activity

The layered structure of LDHs contains alternating divalent and trivalent metal ions, which serve as the primary source of their catalytic activity. Specifically, the catalytic mechanism involves synergistic electron transfer pathways between mixed-valence metal ions, where the divalent ions (e.g., Mg^{2+}, Fe^{2+}) act as electron donors while trivalent ions (e.g., Al^{3+}, Fe^{3+}, Zr^{4+}) function as electron acceptors, creating localized charge polarization that facilitates redox reactions. Metal ions such as Mg^{2+}, Al^{3+}, and Fe^{2+}/Fe^{3+} can provide catalytic activity through their electronic structures and changes in oxidation states. For example, the impact of various transition metal ions

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(Fe^{3+}, Cr^{3+}, Cu^{2+}, Co^{2+}, Ni^{2+}, Mn^{2+}, Zn^{2+}, and Zr^{4+}) on the active sites of MgAl-LDHs has been investigated; MgAl-Zr-LDHs exhibited the highest catalytic activity (Wang, et al., 2015; D. Wang, et al., 2018) (Figure 6A). Similarly, doping with metal ions such as Ga^{3+} and indium (In^{3+}) can significantly enhance the catalytic activity of LDHs. The larger ionic radius and lower electronegativity of In^{3+} compared to Al^{3+} weaken the In–O bonds, promoting electron delocalization across the LDH layers and accelerating proton-coupled electron transfer (PCET) processes during catalysis (H. Wang, et al., 2020) (Figure 6B-C). The catalytic performance is further governed by the interplay between metal ratios and their electronic hybridization. For instance, Wang et al. (L. Wang, et al., 2010) used CoMgFe-LDHs as precursors and obtained CoFe/MgO alloy samples through reduction in a hydrogen atmosphere. Density functional theory (DFT) studies reveal that a Co: Fe ratio of 3:1 optimizes the overlap between Co^{3+} eg orbitals and Fe^{3+} t2g orbitals, enabling efficient intermetallic charge transfer (IMCT) during redox reactions. By adjusting the Co/Fe metal ratio in the LDHs precursor, they significantly improved the catalytic activity of the resulting material. Based on the uniform dispersion of metals within the main layers of LDHs, there is a strong interaction between the active metal and the oxide support. Compared with monometallic supported catalysts, bimetallic catalysts offer tunable geometric, electronic, and interfacial structures, and the synergistic catalytic effects can significantly enhance the performance of metal catalysts. By introducing two catalytically active transition metal elements simultaneously into the Mg-based LDHs layers, a bimetallic catalyst supported on MMO can be obtained. Gao et al. (W. Gao, et al., 2014) used NiMgFe-LDHs as precursors to prepare a bimetallic NiFe/MgO alloy catalyst. The Ni–Fe dual sites establish a bidirectional electron transfer pathway: electrons from hydrazine (N_2H_4) dehydrogenation are transferred to Ni via σ-donation, while Fe^{3+} withdraws electrons through π-backbonding, collectively weakening N–N and N–H bonds. This catalyst could rapidly and completely decompose hydrazine to generate hydrogen gas at room temperature, and its catalytic activity was significantly higher than that of monometallic catalysts.

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In summary, metal atoms such as Fe, Co, Ni, and Cu, supported on the LDHs layers, are key to their high catalytic activity. Through in-depth studies of the activity of metal ions, it is possible to further optimize the performance of LDHs and to expand their range of applications.

3.1.2. Interlayer anion activity

The interlayer anions in LDHs play a vital role in their catalytic activity. The presence of these anions not only stabilizes the layered structure but also regulates the reaction environment. Specifically, anions such as SO_4^{2-}, CO_3^{2-}, and Cl^- are intercalated between the metal hydroxide layers, where they interact with metal ions through electrostatic forces. This interaction prevents the delamination or disintegration of the layered structure, thus ensuring its stability. Additionally, the interlayer anions can participate in ion exchange processes during catalytic reactions, thereby influencing both the reaction rate and selectivity. In practical applications, the interlayer anions in LDHs can exchange with ions from the surrounding solution. This ion exchange process not only enables the fine-tuning of the material's properties, but also facilitates the selective removal or capture of specific ions.

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For instance, Ma et al. (Lijiao, et al., 2017; Lijiao Ma, et al., 2017; L. Ma, et al., 2016) synthesized MgAl-LDHs intercalated with tetrathiomolybdate (MoS_4^{2-}) anions. They found that these materials exhibited excellent selective adsorption properties toward heavy metal cations (such as lead [Pb^{2+}], mercury [Hg^{2+}], cadmium [Cd^{2+}], and silver [Ag^+], among others) and metal-oxygen anions (such as $HAsO_3^{2-}$/$HAsO_4^{2-}$, and chromate [CrO_4^{2-}]) (Figure 6D). The LDHs demonstrated selective adsorption of the target heavy metals as well as resistance to oxidation, thereby enabling efficient removal of the target ions. In addition, Wang et al. (J. Wang, et al., 2015) synthesized nanosheets of graphene oxide-LDHs (MgAl-LDHs/GO). Due to the rich functional groups (-C=O, -COOH, -OH, etc.) on the surface of graphene oxide, the graphene sheets acquire a negative charge. This negative charge facilitates the adsorption of positively charged cations into the interlayer of the graphene sheets, enabling ion exchange. Exploiting this property, the material allows for the exchange and detection of a variety of ions.

In conclusion, interlayer anions in LDHs play a crucial role in contributing to the structural stability of the material and also in influencing its catalytic activity, ion-exchange capacity, and charge balance. These anions preserve the integrity of the layered structure, facilitate ion exchange processes, and enhance the overall performance of LDHs in applications such as selective adsorption, catalysis, and environmental remediation.

3.1.3. Surface and interface activity

The surface and interface activity of LDHs also plays a critical role in their overall reactivity. Firstly, due to their high specific surface area, LDHs provide more opportunities for the contact between molecules and ions. The interface regions of LDHs can create a specific chemical environment that alters the adsorption properties of reactants and modifies their reaction pathways. Lei et al. (X. Lei, et al., 2011) synthesized MgAl-LDHs using two methods: the nucleation/crystallization separation method (SNAS) (Evans & Xue, 2006) and the urea decomposition method. They found that the LDHs synthesized via the SNAS method had smaller, more uniform crystal sizes, a larger specific surface area, and more exposed active sites on the surface. At a reaction temperature of 120°C, the catalytic conversion rate of the reactants reached 87%. When LDHs are exfoliated into monolayer nanosheets, they allow for the maximum exposure of active sites on the surface of the LDHs catalyst. Ruiz et al. (Cesar, et al., 2014) also found that monolayer MgAl-LDHs catalysts possess abundant Lewis basic sites on their surface, which contribute to outstanding catalytic activity during catalytic applications. Additionally, the structural defects and amorphous regions on the surface of LDHs act as preferential adsorption sites for reactants or active sites for catalytic reactions, enhancing both the surface and interface activity of the LDHs. The surface and interface activity of LDHs are critical factors contributing to their excellent performance in a variety of applications. By optimizing the surface and interfacial

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structure of LDHs, their performance can be further improved, leading to more efficient application outcomes.

In conclusion, the activity of LDHs can be attributed to three main sources, metal ions, interlayer anions, and the surface and interface activity. By understanding and mastering these sources of activity, they provide a foundation for optimizing the structural design strategies of LDHs, enabling the preparation and regulation of high-performance LDHs.

3.2. The activity regulation of LDHs

Although the unique structural characteristics of LDHs have found applications in various fields, limitations such as low activity and limited versatility hinder further in-depth research. Thus, enhancing and optimizing the catalytic performance of LDHs has become a focal point in current studies. Among the most promising strategies for tuning their properties are cation doping, anion exchange, and the formation of LDHs composites (Taviot-Gueho, et al., 2018; P. Wang, et al., 2023).

3.2.1. Cation doping

Cation doping of LDHs refers to the incorporation of different metal ions into the original layered structure of LDHs, thereby enabling modulation of their catalytic performance. In this process, the dopant ions must be similar to the original metal ions in terms of the ionic radius and valence state to maintain the structural stability of the material. Yang et al. (Y. Yang, Ou, Yang, & Wei, 2019) employed a cation doping strategy and introduced vanadium (V^{5+}) ions into the CoFe-LDHs during the hydrothermal synthesis process to produce CoFeV-LDHs. Through density functional theory (DFT) simulations, they discovered that doping with high-valent V^{5+} ions enhance the ability of Co or Fe to form covalent bonds with oxygen in hydroxyl groups. This in turn promotes the transfer of electrons from the oxygen atom to the metal atoms, reducing the energy required for charge transfer during the oxygen evolution reaction (OER), thus facilitating the OER catalytic process. Additionally, Ding et al. (Y. Ding, Du, & Zhang, 2020) successfully synthesized Mo^{2+}-doped CoFe-LDHs nanorods via a hydrothermal method. The incorporation of Mo^{2+} ions was shown to modulate the

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electronic structure of the material, the Mo^{2+}-doped CoFe-LDHs exhibited reduced water adsorption energy compared with pure CoFe-LDHs, with the adsorption energy decreasing from -2.58 eV to -0.86 eV. Furthermore, DFT calculations indicated that the introduction of Mo^{2+} promotes the formation of an amorphous phase, thereby accelerating electron transfer and enhancing the electrocatalytic performance of CoFeMo LDHs. Meanwhile, Yu et al. (Jie Yu, et al., 2021) employed the fast nanoscale precipitation (FNP) technique to prepare NiFe-LDHs, and achieved La^{3+} doping through simple mechanical grinding. La^{3+} ions could adsorb onto the surface of the LDHs without disrupting the overall structure. When the doping concentration exceeded 5%, the DFT calculations revealed that La-NiFe-LDHs exhibited a significantly reduced Gibbs free energy for the OOH* adsorption in the rate-determining step (RDS) of the OER four-electron process compared with NiFe-LDHs. This reduction in Gibbs free energy enhanced the overall OER, making La-NiFe-LDHs a more effective electrocatalyst.

At the same time, La-NiFe-LDHs demonstrated a lower Gibbs free energy in the Volmer step (hydrogen adsorption process) of the hydrogen evolution reaction (HER), indicating stronger hydrogen adsorption behavior, less hydrogen generation, and a lower desorption barrier. This suggests that the doping of La^{3+} significantly enhances the intrinsic catalytic performance of LDHs. Tang et al. (Y. T. A, A, B, A, & B, 2020) developed NiFe-LDHs nanosheets doped with V^{5+} and found that the doping of V^{5+} significantly enhanced the HER performance of NiFe-LDHs, showing excellent catalytic activity during electrocatalytic reactions (Figure 7A). Meanwhile, Xiong et al. (B, et al., 2020) explored the doping of various trivalent or tetravalent metal cations (titanium [Ti^{4+}], Ga^{3+}, or Al^{3+}) into ZnM-LDHs (Figure 7C). Their study revealed that such doping could enable precise and directional tuning of the catalytic properties of ZnM-LDHs.

In brief, by employing metal ion doping strategies such as V^{5+}, Mo^{2+}, and La^{3+}, the catalytic activity of LDHs can be effectively enhanced and tuned. These strategies provide a promising pathway for optimizing LDH performance across various

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electrocatalytic reactions, offering significant insights and opportunities for future research and applications.

3.2.2. Anionic substitution

Due to the interlayer anion exchangeability characteristic of LDHs, the interlayer anions can be replaced by various inorganic anions, polyoxometalates, heteropoly acid anions, and other species from the external environment. This ion exchange process allows for the tuning of the material's properties and enables selective removal or capture of specific ions. This unique feature of LDHs makes them highly versatile for a range of applications, such as environmental remediation, ion filtration, and catalysis. For instance, Yan et al. (Hou, Yan, Li, Yang, & Zhao, 2019; Jing, et al., 2019; J. Li, Yu, Zhang, Zhu, & Yan, 2020) incorporated ethylenediaminetetraacetic acid (EDTA) into MgAl-LDHs to construct high-performance LDHs (EDTA-MgAl-LDHs) capable of effectively removing heavy metals from aqueous solutions (Figure 7B). The detection mechanism is based on the electrostatic interaction between negatively charged CrO_4^{2-} and the surface of EDTA-MgAl-LDHs, leading to a complexation reaction. CrO_4^{2-} undergoes interlayer anion exchange with the -COOH groups of EDTA, resulting in the reduction of Cr^{6+} to the non-toxic and environmentally benign Cr^{3+}. This interlayer anion exchange process modifies the activity of the LDHs, thereby enabling their application in the detection of foodborne contaminants. Furthermore, during the anion exchange process, the efficiency of the anion exchange process is influenced by the nature of the exchanged anions. The ease of anion substitution is closely related to the ionic radius and charge density of the anions. Specifically, smaller ionic radii and higher charge densities tend to facilitate the replacement of interlayer anions in LDHs, the exchangeability of the anion A^- and the exchange capacity of the incoming B^- ion. In the common inorganic anions, they can be exchanged in the order of $NO_3^- > Cl^- > SO_4^{2-} > CO_3^{2-}$ (Jiang, et al., 2024).

The unique layered structure of LDHs allows for the interchangeability of interlayer anions. By selecting the appropriate anion species, the catalytic properties of LDHs can be effectively tuned, offering promising potential for widespread application

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in detection processes. This tunability, inherent to the layered architecture, provides a versatile approach for optimizing LDHs in various catalytic and sensing contexts.

3.2.3. Composite

The enhancement of catalytic activity in LDHs composite is primarily achieved by constructing specialized structures. These composites can be integrated with various materials such as transition metal oxides (MOn), transition metal atoms, transition metal sulfides (MXenes), transition metal phosphides, and carbon-based materials. The strategy aims to increase the density of active sites and reduce the adsorption energy of intermediate products, thereby improving the overall catalytic efficiency of the material. Gao et al. (Z. W. Gao, et al., 2018) synthesized a novel CoO/CoFe-LDHs composite by depositing CoO nanoclusters onto CoFe-LDHs nanosheets. Compared with the individual CoO and CoFe-LDHs components, the CoO/CoFe-LDHs composite demonstrated significantly enhanced electrocatalytic performance. This improved catalytic activity can be attributed to the larger interfacial area between the two phases formed during the composite synthesis, which facilitates more efficient catalytic interactions. Meanwhile, Wang et al. (Y. Wang, Wang, Zhang, Liu, & Sun, 2020) also demonstrated that the composite strategy can significantly enhance the catalytic performance of LDHs. They successfully prepared CuO nanofibers coated with CoFe-LDHs nanosheets (CuO@CoFe-LDHs/CF) via a deposition method. Thanks to this three-dimensional (3D) composite structure, CuO@CoFe-LDHs/CF exhibited a large number of active sites, which greatly improved its electrocatalytic performance, with a remarkable 14-fold enhancement compared with the non-composite material. This enhancement can be attributed to three main factors: first, the highly conductive substrate material in the composite significantly promotes electron transfer. Second, the 3D network structure exposes a high density of active sites, accelerating surface catalytic reactions. Third, the strong interactions between CoFe-LDHs and the composite materials induce changes in the electronic structure, reducing the adsorption energy of intermediate products. These synergistic effects highlight the potential of

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composite strategies in optimizing the catalytic properties of LDHs for advanced applications.

In summary, cation doping, anion substitution, and composite strategies can all significantly regulate the catalytic performance of LDHs. Selecting the appropriate modification method based on the specific application requirements is crucial to optimize the material's performance and to meet practical demands.

4. The applications of LDHs in food safety detection

Due to their unique layered structure and adjustable catalytic activity, LDHs exhibit numerous advantages that enable them to demonstrate broad potential in the field of food safety detection. LDHs have shown exceptional sensing and recognition capabilities in the detection of common food contaminants, such as heavy metal ions, pesticide residues, and toxins (Table 2).

4.1. Heavy metal ions

The exceptional ion-exchange capacity, high specific surface area, and adsorption properties of LDHs demonstrate remarkable potential in the detection of heavy metal ions. The sensing and recognition mechanisms of LDHs towards heavy metal ions differ. They primarily recognize these ions through two processes: reversible adsorption and irreversible mineralization. The former involves the adsorption of heavy metal ions onto the surface or interlayers of the LDHs, while the latter immobilizes these ions within the lamellar structure through ion exchange, leading to the formation of new LDHs. On the other hand, irreversible mineralization is more effective than reversible adsorption, achieving the goal of ultra-stable mineralization. For instance, Lian et al. (Jiajia Lian, 2022) employed a one-pot hydrothermal method for *in situ* loading of CeO_2 nanoparticles onto the surface of LDHs. By leveraging the high specific surface area of LDHs and the uniformly dispersed CeO_2 nanoparticles, they collaboratively developed a colorimetric sensor for Cu^{2+} detection, with a limit of detection (LOD) of 9.80 µM. When Cu^{2+} ions are introduced into the system, this color change is due to the formation of a complex between Cu^{2+} and LDH-CeO_2 composite, which results in alterations to the optical properties of the composite. The colorimetric change can be quantitatively

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measured, providing a method for Cu^{2+} detection. Similarly, the team of Sankarlinkam et al. (Sankarlinkam, Suresh, & Hariharan, 2024) developed an electrochemical sensor for Fe^{2+} detection by constructing a MgAl-LDHs composite-modified glassy carbon electrode. The sensor primarily relies on the multilayered structure of MgAl-LDHs, which catalytically oxidizes Fe^{2+} to Fe^{3+} during the detection process, thus enabling the detection of Fe^{2+} (Figure 8A-B). The sensor demonstrated a linear range from 0.1 to 102.1 μM, with a sensitivity of 0.061 μA μM^{-1} and a LOD of 50 nM. Even in the presence of other interfering metal ions, it achieved efficient, rapid, and intelligent detection of Fe^{2+}. In addition, Mansour et al. (Salem, Khan, Manea, Qashqoosh, & Alahdal, 2023) synthesized a composite functional material of polyaniline/layered double hydroxides@carbon nanotubes (PANI/LDHs@CNT) and constructed a heavy metal ion sensor based on this material for detecting manganese (Mn^{7+}) and Cr^{6+} via a fluorescence ‘quenching’ response and Al^{3+} via a fluorescence ‘turn-on’ response (Figure 8C-F). The detection mechanism primarily involves the phenomenon of fluorescence quenching due to the internal filtration effect when Mn^{7+} and Cr^{6+} are present. In contrast, when Al^{3+} is introduced, the fluorescence intensity is restored and enhanced through a metal ion chelation effect. Under optimal conditions, the LODs for Mn^{7+}, Cr^{6+}, and Al^{3+} were 51 nM, 59 nM, and 81 nM, respectively. Guo et al. (Guo, Li, Chen, Yu, & Ma, 2020) synthesized a tunable luminescent composite functional material, SDC/OS Mg/Al-LDHs, based on LDHs. SDC refers to 4,4’-styrene dicarboxylic acid, and OS is sodium 1-octanesulfonate. Using this material, they developed a fluorescence sensor for the efficient, portable, and rapid detection of Fe^{3+} (Figure 8G-H). Within the Fe^{3+} concentration range of 0.18~1.78 μM (0.01~0.1 ppm), the quenching constant of the composite was 2.34×10^{5} M^{-1}, with a LOD of 6.12×10^{-8} M. The detection mechanism can be attributed to the binding reaction between Fe^{3+} metal ions and ligands in SDC/OS Mg/Al-LDHs structure, which causes the quenching of SDC/OS Mg/Al-LDHs fluorescence and finally realizes the detection. Yu et al. (Y. Yu & Yang, 2022) prepared a composite functional material, MnMgFe-LDHs/BC, by co-precipitating Mn-Mg-Fe-LDHs onto biochar (BC). They applied this material for

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the detection of Cd^{2+}. The detection mechanism primarily involves the abundant hydroxyl groups and interlayer anions on the surface of MnMgFe-LDHs, which enable the capture of Cd^{2+}. Additionally, the porous structure of the conductive biochar provides channels for the adsorption of Cd^{2+}. The detection method achieved a LOD for Cd^{2+} as low as 0.03 ng/L and demonstrated high-efficiency Cd^{2+} detection in water samples. Chen et al. (Chen, et al., 2023) used a hydrothermal method for *in situ* synthesizing and assembly of Fe-Co-LDHs rods around MXene, resulting in the preparation of a 3D Fe-Co-LDHs/MXene composite material. Due to the well-designed 3D structure and excellent conductivity, this material exhibited outstanding performance in detecting As^{3+}. Where the As^{3+} ions interact with the material's surface, causing a measurable change in conductivity that can be used for sensitive detection. Within a linear detection range of 1~1000 ppt, the current response on the Fe-Co-LDHs/MXene/GCE electrode showed a strong linear correlation with the As^{3+} concentration. The sensitivity was 0.22 μA ppt^{-1} cm^{-2}, and the LOD was 0.9 ppt. Liu et al. (M. Liu, et al., 2017) successfully loaded the organic dye acridine orange (AO) onto LDHs, endowing the LDHs with unique fluorescent properties. This modification allowed the material to achieve high-sensitivity detection of Hg^{2+} in a linear range from 0.5 to 150 mM.

Overall, LDHs have demonstrated considerable promise in detecting heavy metal ions, thanks to their superior ion exchange properties, large specific surface area, and unique structural characteristics. Moving forward, research should explore the catalytic behavior of LDHs under diverse environmental conditions, aiming to optimize their sensitivity, selectivity, and stability for more reliable heavy metal detection. By advancing these properties and incorporating innovative materials or technologies, LDHs could significantly enhance environmental monitoring, offering a more sustainable and efficient solution for global pollution management. This approach would pave the way for their practical and widespread application.

4.2. Pesticide residue detection

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The excellent structural properties of LDHs also make them highly promising for applications in pesticide residue detection. For example, Liang et al. (Liang, Miao, & Gong, 2012) developed a green and simple method to synthesize a novel Ni/Al-LDH graphene nanosheets (LDHs-GNs) composite material. This material was synthesized on a graphene nanosheet cathode substrate. The LDHs-GNs composite demonstrated efficient and specific capture of organophosphate pesticides (OPs) (Figure 9A-B), primarily due to the high selective adsorption properties of LDHs and the large surface area and high electrical conductivity of graphene nanosheets. The unique structural characteristics promoted the enrichment of OPs on the surface, allowing for highly sensitive detection of methyl parathion, with a LOD of 0.6 ng/mL. Gong et al. (Gong, Guan, & Song, 2013) utilized LDHs as a matrix for immobilizing acetylcholinesterase (AChE) to detect OPs (Figure 9C-D). Due to the regular structure, good mechanical properties, and thermal stability of LDHs, they provide an excellent biocompatible microenvironment that helps maintain the bioactivity of AChE. By combining flow injection analysis (FIA) with amperometric detection, the AChE-LDHs-modified electrode significantly promoted the oxidation of thiocholine products generated by the enzymatice reaction, thereby enhancing the sensitivity for the detection of methyl parathion. In the linear ranges of 0.005~0.3 μg/mL and 0.3~4.0 μg/mL, the LOD was 0.6 ng/mL (S/N = 3). The developed detection method exhibited good reproducibility and stability. Zhao et al. (Y. Zhao, et al., 2022) developed a MI-NiAl-LDHs material with specific molecular recognition ability for glyphosate by electro-deposition and low-temperature plasma methods on Ni nanorod arrays (Ni NRAs) (Figure 9E-G). This material demonstrated high sensitivity and strong recognition ability towards glyphosate. The catalytic active centers were primarily located at the Ni^{3+} sites in the MI-NiAl-LDHs, which at relatively low anodic potentials, triggered the formation of highly active NiOOH sites. These sites significantly enhanced the electrocatalytic oxidation ability of the NiAl-LDHs interface for the C-N bond. The LOD was 3.1 nmol/L within the glyphosate linear range of 10.0 nmol/L to 1.0 μmol/L. Zhang et al. (C. Zhang, Liang, Lu, Li, & Xu, 2020) developed a novel copper-aluminum metal

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hydroxide-doped graphene nanoprobes (CuAl-LDHs/Gr NC) to detect trace amounts of glyphosate (Figure 9H-I). The constructed sensor exhibited strong enrichment capability for glyphosate, with a logarithmic linear detection range from 2.96×10^{-9} to 1.18×10^{-6} mol/L and a LOD of 1×10^{-9} mol/L. The detection mechanism relies on the interaction between the CuAl-LDHs/Gr NC and glyphosate. Glyphosate adsorbs onto the material's surface, causing a change in its intrinsic properties, thereby enabling detection. Additionally, the sensor demonstrated excellent reproducibility, stability, and resistance to interference, making it a promising tool for sensitive and reliable glyphosate detection.

In light of the findings, the composition and structural properties of LDHs can be strategically tailored to meet the specific needs of pesticide detection, allowing for precise recognition and selective identification of various pesticide types. Furthermore, integrating LDHs with advanced pre-treatment methods, such as electrochemical modification or nanomaterial doping, can significantly enhance detection sensitivity, selectivity, and overall efficiency. These tailored strategies not only improve the accuracy and reliability of pesticide residue detection but also pave the way for more robust and adaptable sensing platforms in environmental monitoring and safety assessments.

4.3. Mycotoxin detection

LDHs have also demonstrated significant potential in the detection of mycotoxins. For example, a research team led by Zhang et al. (X. Zhang, et al., 2022) developed an electrochemical aptamer sensor for the detection of ochratoxin A (OTA) by using MXene-Au as the electrochemical sensing platform and platinum (Pt) nanoparticle anchored hollow NiCo layered double hydroxides (Pt@NiCo-LDHs) as the signal amplification material (Figure 10A-B). In this system, the authors first assembled complementary DNA (cDNA) onto the MXene-Au/working electrode (WE) via Au-S bonds, followed by hybridization with an aptamer exhibiting excellent peroxidase (POD)-like activity. Subsequently, they introduced Pt@NiCo-LDHs to trigger the ‘signal on’ state, generating a significant electrochemical signal. Upon the presence of

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OTA, the aptamer-cDNA hybrid dissociates, leading to a ‘signal off’ state. This ‘signal on/off’ strategy enables highly sensitive detection of OTA, with a linear range spanning from 20 fg/mL to 100 ng/mL and a LOD of 8.9 fg/mL (S/N = 3). This detection technology holds great promise for efficient, intelligent, and portable OTA monitoring in field applications. The research team led by Zhang et al. (X. Zhang, et al., 2023) developed an electrochemical-colorimetric dual-mode microfluidic paper-based analytical device (μPAD) for the detection of aflatoxin B1 (AFB1). In this system, the researchers anchored gold nanoparticles in Ni-Co layered double hydroxide nanocages (Au/Ni-Co LDHs NCs) to amplify the signal, facilitating sensitive AFB1 detection (Figure 10C-D). The sensor operates through the hybridization of an aptamer with cDNA, which leads to the binding of Au/Ni-Co LDHs NCs with excellent POD-like activity to the target DNA nanostructures. This binding catalyzes the production of hydroxyl radicals from hydrogen peroxide (H_2O_2), which in turn catalyze the substrate, 3,3’,5,5’-tetramethylbenzidine (TMB), to generate a blue-colored product, thus providing a colorimetric signal output. However, when the target AFB1 binds to the aptamer, it induces the dissociation of the aptamer/cDNA complex, leading to a decrease in the signal. The electrochemical-colorimetric dual-mode sensor demonstrated high sensitivity for AFB1 detection, with a linear range of 0.2 pg/mL to 100 ng/mL in the electrochemical mode and 50 pg/mL to 100 ng/mL in the colorimetric mode.

In summary, the application of LDHs for mycotoxin detection showcases their exceptional potential in delivering rapid, accurate, and intelligent detection solutions. The unique properties of LDHs, such as high surface area and selective adsorption, enable the development of highly sensitive and specific sensors for mycotoxins. By further optimizing the composition and structure of LDHs, along with incorporating innovative detection technologies, this approach can be significantly advanced to offer more robust, real-time, and reliable systems for mycotoxin monitoring in food safety and environmental assessments.

4.4. Antibiotic detection

LDHs, when combined with conductive materials such as carbon quantum dots (CQDs) or graphene, enable highly sensitive detection of antibiotics. For instance, the research team led by Sanny et al. (Carvalho, et al., 2019) demonstrated the controlled doping of CdTe quantum dots (QDs) onto the surface of pristine LDHs, resulting in a novel multi-functional LDHs-based composite material. This material was subsequently used to develop a highly sensitive and selective electrochemical method for detecting ciprofloxacin. Within a linear range of 2.5×10^{-8}~1.2×10^{-5} mol/L, the LOD was 4.2×10^{-8} mol/L, while the limit of quantification (LOQ) was 1.3×10^{-7} mol/L. The detection mechanism is that ciproloxacin binds carboxyl group (-COOH) and ketone group (-C=O) to the Cd^{2+} site on the quantum dot, stabilizing the adsorbed intermediate and reducing the oxidation overpotential. Remarkably, it maintained 94% accuracy even in the presence of interfering substances such as Zn^{2+}, Fe^{2+}, Cu^{2+}, citric acid, and ascorbic acid. In addition, the research team led by Chelladurai et al. (Karuppiah, et al., 2022) developed a novel 2D nanosheet structure of zinc ammonium molybdate layered double hydroxides (AZnMo-LDHs) using a combination of co-precipitation and ultrasonic methods. This material exhibited a strong response to dimetridazole (DMZ), with a linear range of 0.25~570 μM. The sensitivity was 1 $\mu A\ \mu M^{-1}\ cm^{-2}$, and the LOD was 0.021 μM. Furthermore, this method has been applied successfully for the detection of DMZ in real samples, including eggs and milk (Figure 11A-B). The detection mechanism is primarily attributed to the strong adsorption of DMZ onto the AZnMo-LDHs, which results in changes in the electrochemical signal. Additionally, Zheng et al. (Zheng, et al., 2024) developed an efficient and stable catalyst (RuZnAl-LDHs) for the high-performance detection and degradation of tetracycline hydrochloride (Figure 11C-D). The detection mechanism relies on the uniform distribution of single ruthenium (Ru) atoms on the ZnAl-LDHs, which exhibits excellent photocatalytic properties under visible light. The catalyst demonstrates a fast response, a low LOD (1.0 mg/L), and strong anti-interference capability. This study has provided inspiration for the design of multifunctional single-atom catalysts and broadened the potential applications of single-atom catalysts in environmental and analytical fields. Building

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on this, the research team led by Duan et al. (Duan, et al., 2024) developed a visual electrochemical immunosensor by using CoMn-LDHs derived from CoMn-ZIF as the sensitive material and ink to modify screen-printed electrodes (SPE). This sensor, in collaboration with a smartphone, enables rapid and portable detection of Tylenol (TYL) (Figure 11E). The detection mechanism primarily relies on the large specific surface area, wide pore size, rich oxygen vacancies, and exposed metal sites of CoMn-LDHs. The sensor operated within a linear range of 1.0 fg/mL to 1.0 ng/mL, with a LOD of 0.89 fg/mL for TYL. The constructed electrochemical immunosensor exhibited high selectivity, good reproducibility, and stability. This study provided a novel electrochemical immune sensing strategy for the efficient detection of antibiotics in food samples.

In conclusion, LDHs hold remarkable potential for antibiotic detection, offering highly sensitive and efficient platforms for identifying a wide range of antibiotics. By strategically modifying the structure and composition of LDHs, it becomes feasible to develop multiplexed detection systems capable of simultaneously detecting multiple antibiotics, thus enhancing detection versatility. This approach not only improves detection speed and accuracy but also offers a promising solution for rapid screening in clinical diagnostics, food safety, and environmental monitoring, addressing growing concerns about antibiotic resistance and contamination.

4.5. Carcinogens detection

The unique structural characteristics of LDHs continue to demonstrate promising application potential in the detection of carcinogenic substances in food. For instance, Zhao et al. (Z. Zhao & Zheng, 2024) synthesized CoFe-LDHs and modified it onto a glassy carbon electrode to construct a bifunctional sensor (CoFe-LDHs/GCE) capable of simultaneously detecting dopamine (DA) and acetaminophen (AP) (Figure 12A-B). This sensor exhibited a wide linear detection range for DA and AP (2~900 μM), with a LODs of 0.08 and 0.37 μM, respectively, and a sensitivity of 0.54 and 0.55 $\mu A\ \mu M^{-1}\ cm^{-2}$, respectively. Additionally, it demonstrated excellent reproducibility, selectivity, and stability, highlighting its significant research value for the detection of carcinogenic

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substance AP. The detection mechanism is based on the electrochemical interactions between the CoFe-LDHs/GCE and the target molecules (DA and AP), where adsorption onto the electrode surface leads to changes in the electrochemical signal, enabling simultaneous and sensitive detection of both substances. Prasanna et al. (Prasanna, et al., 2024) developed a composite functional material (AuNP/Ni-Fe LDHs/BPNSs) by loading black phosphorus nanosheets (BPNSs) onto nickel-iron layered double hydroxides (Ni-Fe LDHs). The AuNP/Ni-Fe LDHs/BPNSs composite exhibited a large specific surface area, high electrical conductivity, excellent electrocatalytic activity, and rapid electron transfer characteristics. These properties are crucial for its role in detecting diphenylamine (DPA) in food matrices (Figure 12C-D). Within a linear range of 0.0125~3.825 μM, the sensor demonstrated a LOD of 4.63 nM and a sensitivity of 0.399 μA μM^{-1} cm^{-2} for DPA. The sensor based on this material showed excellent selectivity, stability, reproducibility, and repeatability, and was applied successfully to monitor DPA in apples, sweet tomatoes, pears, and grapes, with satisfactory detection recoveries. In parallel, Ayesha Aziz et al. (Aziz, et al., 2022) synthesized CuNiAl-LDHs, which exhibited excellent electrocatalytic performance for nitrate detection within a voltage range of -0.7~+0.7 V (Figure 12E). The sensor demonstrated linear detection ranges of 5 nM~40 μM and 75 μM~2.4 mM, with a LOD of 0.02 nM (S/N = 3). The sensitivity was 830.5 ± 1.84 μA mM^{-1} cm^{-2}, and the response time was 3 seconds. The CuNiAl- LDHs serve as electrocatalysts, facilitating the electrochemical reduction of nitrate ions. The interaction between nitrate and the electrocatalytic surface of CuNiAl-LDHs leads to electron transfer at the electrode surface, which generates a measurable electrochemical signal. The developed sensor enabled portable, rapid, and efficient nitrate detection in mineral water, tap water, and Pepsi beverages. Furthermore, Liu et al. (M. Liu, et al., 2024) successfully synthesized a Ru/NiFe-LDHs-MXene/SPCE electrode through a one-step electrodeposition method and applied it for the detection of nitrofurantoin (NFT) (Figure 12F-G). By combining 2D MXene materials with multifunctional LDHs, the two components worked synergistically to enhance the electrochemical performance. The incorporation of noble metals and nanoclusters

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significantly boosted the electrochemical properties by promoting metal-support interactions and optimizing the reorganization of the electronic structure. The constructed Ru/NiFe-LDHs-MXene/SPCE sensor exhibited excellent sensing performance for NFT, with high sensitivity (152.44 μA μM^{-1} cm^{-2}) and a LOD of 2.2 nM, enabling highly sensitive detection of the NFT content in food samples. However, Yang et al. (S. Yang, KaiqiangZhang, JianBai, ChaopengPeng, JiaxiFang, ZhiliXu, Wenyuan, 2021) designed and synthesized a novel hetero structured material based on silver-doped zinc oxide nanorods (Ag-ZnO NRs) and nickel-cobalt layered double hydroxides (NiCo-LDHs) through a hydrothermal method. This material exhibited strong adsorption and sensing capabilities for the carcinogen quercetin, attributed primarily to the surface plasmon resonance of Ag nanoparticles and the synergistic effect of the heterostructure (Figure 12H-I). The sensor demonstrated a linear detection range from 1.0×10^{-10} to 5.0×10^{-6} mol/L, with a LOD of 0.075 nmol/L for quercetin. The sensing system displayed high selectivity, stability, and reproducibility for quercetin detection.

4.6. Others

LDHs have attracted significant attention as versatile and efficient materials in the field of food safety detection in recent years. Beyond their ability to detect common food contaminants, LDHs have shown significant potential for the detection and monitoring of a variety of substances related to food safety, including glucose (W. Y. A, et al., 2019; Farhat, 2021; Fenfen, et al., 2018; Jian Xin Xie, 2017; L. Wu, et al., 2021; Y. Y. Xu, Wensheng, 2017; W. Zhang, Ding, Zhang, & Shang, 2022a; J. Zhu, Wang, Chen, & Liu, 2023; Y. Zhu, et al., 2024), H_2O_2 (Jian Xin Xie, 2017; Y. Y. Xu, Wensheng, 2017), insulin (Khataee, Dehghan, Shaghaghi, Khataee, & Amini, 2024), ascorbic acid (Erika, Scavetta, Domenica, & Tonelli, 2005; Kong, Shi, Zhao, Wei, & Duan, 2011; Shi, Chen, & Niu, 2021; W. Yang, et al., 2020), trace phenolic compounds (Zhen Li 2021), xanthine (Shan, et al., 2009), DNA (Q. Ding, et al., 2023; Yuanyi, et al., 2023), folic acid (Y. L. Liu, Lei, 2016), and hydrogen sulfide (H_2S) (W. Zhang, Ding, Zhang, & Shang, 2022b). The excellent properties of LDHs make them highly effective in sensing

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low concentrations of these contaminants, offering great promise for improving food safety monitoring.

In addition to the aforementioned contaminants, LDHs have shown great promise in detecting food adulterants—substances added to food products to fraudulently increase volume, weight, or appearance. Detecting food adulteration is critical for ensuring the authenticity and safety of food products. LDHs' unique properties, such as high surface area, tunable interlayer spacing, and selective anion intercalation, make them ideal for the detection of various food adulterants, such as chemical additives, synthetic dyes, and preservatives. For example, LDHs can be employed to detect organic adulterants, such as illegal food colorants, by incorporating recognition sites into their structures, which selectively bind to specific molecules and trigger measurable changes in material properties (Q. Ding, et al., 2023; Yuanyi, et al., 2023). The ability of LDHs to detect food adulterants is further enhanced by their capacity to be incorporated into portable, low-cost sensors. These sensors can be used on-site by food inspectors, regulatory bodies, or even consumers to test for adulteration, enabling real-time monitoring and ensuring that adulterated food products do not reach consumers.

According to the literature, LDH-based detection technologies expand the scope of food safety testing and enhance the ability to monitor food quality and safety. This ability should help prevent issues such as food adulteration, excess contamination by harmful substances, and excessive spoilage, thereby providing robust technological support for improving food quality control and safeguarding public health. Furthermore, in response to practical testing requirements, the rational and precise design of LDHs structures aims to meet specific detection needs, facilitating the efficient detection of contaminants within complex food matrices.

5. The limitations of LDHs in food safety detection

LDHs exhibit significant advantages in the detection of food contaminants due to their high surface area, strong adsorption capacity, and structural tunability. However, they also possess certain limitations.

Journal Pre-proof

5.1 Stability in complex food matrices

One of the primary challenges of using LDHs in food sensing is their stability when exposed to complex food matrices. Foods consist of a variety of components, including proteins, lipids, carbohydrates, and other organic and inorganic substances. These components can interact with the LDHs, potentially altering their structural integrity and functionality. For example, food matrices with high moisture content or acidic environments can destabilize LDHs, reducing their effectiveness in sensing specific analytes. This interaction could lead to a decreased sensitivity or inaccurate detection, particularly in liquid-based food products like beverages or soups.

5.2 Limited selectivity

LDHs have the ability to adsorb a broad range of ions and molecules, but this property can also be a drawback in food sensing applications. In real-world food products, there is often a wide range of ions and compounds present, many of which could interfere with the sensing process. LDHs may lack the selectivity required to differentiate between target analytes and other substances in complex food samples. For example, when using LDHs to detect microbial contamination or foodborne pathogens, the presence of other ions or substances might interfere with the sensor’s response, leading to false positives or false negatives.

5.3 Environmental sensitivity

The performance of LDH-based sensors can be highly sensitive to environmental factors such as pH, temperature, and ionic strength. Foods can have varying pH levels, particularly acidic products like fruits, dairy, or fermented foods. These fluctuations in pH can cause changes in the LDH's structure or lead to the leaching of interlayer anions, potentially compromising the sensor’s accuracy. Similarly, temperature variations during food storage, transportation, or preparation can alter the behavior of LDHs, affecting their ability to reliably sense target molecules. Such environmental sensitivity makes it difficult to maintain consistent and accurate performance across a wide range of food types and storage conditions.

5.4 Complex sensor design and fabrication

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LDH-based sensors often require sophisticated design and fabrication processes to ensure that they function effectively in food sensing applications. The process of synthesizing LDHs with the correct structure, particle size, and surface properties can be labor-intensive and costly. Additionally, integrating LDHs into sensors that can be used in real-world food testing often requires additional steps, such as surface modification or the use of stabilizing agents, to ensure that the sensors remain functional under various conditions. This adds complexity to the overall sensing system, which could limit its practicality for large-scale or routine food safety testing.

5.5 Reusability and long-term stability

Another limitation of using LDHs in food sensing is the issue of reusability. LDHs are often designed for single-use applications, meaning that once they interact with target molecules or ions in food, their ability to function as a sensor may be compromised. The interlayer anions may be exchanged or degraded, reducing the sensor's performance over time. For food safety monitoring, sensors that can be reused multiple times would be more cost-effective and practical, but the inherent limitations of LDHs in terms of their reusability and long-term stability could pose challenges.

Given the existing limitations, we can deepen the interdisciplinary integration of materials science, food science, engineering and manufacturing, and smart sensing to strengthen the reliability of LDHs applications in food safety monitoring.

6. Conclusions

In summary, LDHs represent a highly promising class of materials for the detection of food contaminants, driven by their unique structural properties and versatile functionality. The exceptional potential of LDHs in this field is primarily attributed to three key factors: their intrinsic structural features, ease of functionalization, and outstanding stability, safety, and reusability. The inherent layered structure of LDHs, characterized by high surface area, tunability, and selective adsorption capabilities, allows for effective interaction with target analytes, significantly enhancing the sensitivity and specificity of detection methods. These characteristics make LDHs highly effective for the detection of a wide range of

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contaminants in complex food matrices, including pesticides, heavy metals, and microbial pathogens. Moreover, the ability to easily modify the surface properties of LDHs through functionalization further increases their selectivity for specific contaminants. By incorporating various functional groups or forming hybrid composites, LDHs can be tailored to suit diverse detection requirements, thereby enabling their integration into various sensing platforms such as colorimetric, electrochemical, and fluorescent detection systems. This adaptability enhances their versatility and positions them as a key tool for reliable food safety monitoring.

Equally important, the chemical and thermal stability, biocompatibility, and low toxicity of LDHs make them particularly suitable for food-related applications. Their reusability without significant performance degradation further promotes their long-term use, offering a sustainable and cost-effective solution for continuous monitoring. These features collectively highlight the great promise of LDHs in food safety applications, where both accuracy and sustainability are crucial. Despite these promising attributes, further research is needed to optimize the performance of LDHs in detecting food contaminants. Advancing synthesis methods, refining functionalization techniques, and expanding their detection capabilities are essential for enhancing their practical applications. A deeper understanding of the interaction mechanisms between LDHs and specific analytes will further improve the efficiency and precision of LDH-based detection systems. As research in this field progresses, LDHs are expected to play a pivotal role in ensuring food safety, offering fast, reliable, and economical solutions for the detection of foodborne contaminants.

7. Challenges and future trends

Despite the promising prospects of LDHs in food contaminant detection, they still face numerous challenges. To further enhance the effectiveness of LDHs in food safety detection, a series of optimization and innovative research efforts are required. First, improving the application performance of existing LDHs in various food matrices remains a key challenge. The complex composition of food can influence the properties of LDHs, making it a significant challenge to maintain their high stability and

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selectivity in diverse and dynamic food matrices. Therefore, a key direction for future research is to explore strategies that ensure the stability and selectivity of LDHs in such complex environments. Then, in response to practical application requirements, it is essential to precisely design the structure of LDHs to meet the detection needs of different contaminants. For instance, by controlling the interlayer spacing, doping with metal species that present excellent catalytic performance, and surface functionalization, it is possible to achieve highly selective detection of specific contaminants, thereby enhancing the sensitivity and accuracy of the detection system. Lastly, the development of related industrial products and portable detection devices will require the integration and collaboration of multiple disciplines and fields. Such interdisciplinary efforts are pivotal for achieving rapid, portable, and intelligent food safety detection.

Furthermore, LDHs, with their unique structural versatility, high surface area, and tunable properties, present a wide range of possibilities for advancement in numerous industries beyond food safety, including environmental monitoring, healthcare, and energy storage. In the food safety sector, LDHs’ ability to detect biomarkers of spoilage, such as biogenic amines, volatile organic compounds (VOCs), and microbial metabolites, has been demonstrated. However, their potential goes far beyond meat products. The future research trajectory should explore the application of LDHs for detecting freshness and contamination in various food matrices, from dairy products to fruits and vegetables, with a focus on improving the sensor's adaptability and efficiency in different food environments, Then, the healthcare industry holds promising prospects for LDHs. Their ability to selectively interact with certain biomolecules opens doors for diagnostic tools, drug delivery systems, and biosensors for monitoring health conditions. Future research could leverage their biocompatibility and ability to respond to specific biochemical markers to create more effective medical sensors, diagnostics, and targeted therapies. In the realm of environmental monitoring, LDHs are already being explored for their capacity to detect pollutants such as heavy metals and toxins in water and soil. Expanding this application to broader environmental challenges, such as air quality monitoring and detecting climate-related changes-will become

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increasingly important as we face global environmental crises. LDHs could become essential tools in creating real-time, portable sensors for monitoring pollutants and providing early warnings about ecological shifts. Furthermore, the energy sector could benefit from the unique properties of LDHs in energy storage and conversion devices. Their ability to function as efficient catalysts in fuel cells, supercapacitors, and batteries makes them ideal candidates for enhancing the performance of sustainable energy technologies. Research focused on optimizing their conductive properties and scalability could lead to major advances in energy storage solutions, contributing to the ongoing transition to renewable energy sources.

In conclusion, the future development of LDHs is vast and multifaceted. By expanding their applications, enhancing their performance, and integrating them with advanced technologies, LDHs will continue to push the boundaries of innovation, fostering improvements in a wide range of industries. This broad perspective ensures that their potential is not confined to a single domain but is explored for its transformative contributions across multiple fields.

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CRediT authorship contribution statement

Guangchun Song: Methodology, Investigation, Formal analysis, Data curation, Writing-original draft. **Pengxiang Yuan:** Conceptualization, Supervision, Validation, Writing-review & editing. **Xiaochun Zheng:** Conceptualization, Validation, Writing-review & editing. **Marie-Laure Fauconnier:** Investigation, Formal analysis. **Cheng Li:** Writing-review & editing. **Li Chen:** Conceptualization, Supervision, Writing-review & editing. **Dequan Zhang:** Supervision, Validation, Writing-review & editing, Funding acquisition.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data will be made available on request.

References

Yang, W., Li, J., Liu, M., Dickon, H., Liu, Y., Sun, X., & Yang, J. (2019). Bioinspired hierarchical CoAl-LDH/MFe_2O_4 (Ni, Zn, Co) as peroxidase mimics for colorimetric detection of glucose. *Applied clay science, 181*, 105238-105250.

Tang, Y., Liu, Q., Dong, L., Wu, H., & Yu, X. (2020). Activating the hydrogen evolution and overall water splitting performance of NiFe LDH by cation doping and plasma reduction. *Applied Catalysis B: Environmental, 266*, 118627-118636.

Ashiq, J., Saeed, U., Li, Z., & Nawaz, M. H. (2024). Advances in meat spoilage detection: A review of methods involving 2D-based nanomaterials for detection of spoiled meat. *Journal of Food Composition and Analysis, 132*, 106295-106328.

Aziz, A., Asif, M., Ashraf, G., Iftikhar, T., Hu, J., Xiao, F., & Wang, S. (2022). Boosting electrocatalytic activity of carbon fiber@fusiform-like copper-nickel LDHs: Sensing of nitrate as biomarker for NOB detection. *Journal of Hazardous Materials, 422*, 126907-126918.

Xiong, X., Zhao, Y., Shi, R., Yin, W., Zhao, Y., Waterhouse, G., & Zhang, T. (2020). Selective photocatalytic CO_2 reduction over Zn-based layered double hydroxides containing tri or tetravalent metals. *Science Bulletin, 65*, 987-994.

Carvalho, S., Santana, T., Matos, C., Costa, L., Sussuchi, E., & Gimenez, I. (2019). Synthesis of Hydrotalcite-Supported CdTe Semiconductor Nanocrystals for Electrochemical Detection of Ciprofloxacin. *Journal of the Brazilian Chemical Society, 30*, 1266-1275.

Hidalao, J., Sanchidrian, C., & Ruiz, J. (2014). Delaminated layered double hydroxides as catalysts for the Meerwein-Ponndorf-Verley reaction. *Applied Catalysis A: General, 470*, 311-317.

Chen, L., Xiao, P., Hu, B., Wang, Y., Yang, J., & Liu, J. (2023). Preparation of Fe−Co−LDH/MXene modified electrode for sensitive determination of arsenic

Journal Pre-proof

(III) in aquatic system. *Electroanalysis, 35*, 202200521-202200531.

Fang, C., Dai, Y., Chen, L., Wang, Z., Lu, Q., Li, Y., Cai, L., Liu, B., Zhang, Y., Li, Y., & Wang, L. (2022). One-step coprecipitation synthesis of Cl− intercalated Fe_3O_4@SiO_2 @MgAl LDH nanocomposites with excellent adsorption performance toward three dyes. *Separation and Purification Technology, 295*, 121227-121241.

Kumari, S., Soni, S., Sharma, A., Kumar, A., Sharma, V., Jaswal, V., Bhatia, S., & Sharma, A. (2024). Layered double hydroxides based composite materials and their applications in food packaging. *Applied Clay Science,247*, 107216-107231.

Sohrabi, H., Arbabzadeh, O., Falaki, M., Majidi, M., Han, N., Yoon, Y., & Khataee, A. (2022). Electrochemical layered double hydroxide (LDH)-based biosensors for pesticides detection in food and environment samples: A review of status and prospects. *Food and Chemical Toxicology, 164*, 113010-113022.

Stanley, M., Sriram, B., Wang, S., Sherlin V, A., Kogularasu, S., & George, M. (2025). Preserving food quality: Electrochemical detection of synthetic food antioxidant, propyl gallate in processed foods using ternary component layered double hydroxide/graphene aerogel synergy. *Materials Today Sustainability, 29*, 101061-101072.

Muthukumaran, M., Govindaraj, M., Kogularasu, S., Siriram, B., Raja, B., Wang, S., Chien, G., & Selvi J, A. (2024). Recent advances in metal-organic frameworks for electrochemical sensing applications. *Talanta Open, 11*, 100396-100421.

Sakthivel, K., Balasubramanian, S., Chien, G., Wang, S., Ahammad., Billey, W., & Platero, J. (2024). Editors' Choice—Review—Advances in Electrochemical Sensors: Improving Food Safety, Quality, and Traceability. *ESC Sensors Plus, 3*, 020605-020630.

Stanley, M., Sriram, B., Wang, S., Sherlin V, A., Kogularasu, S., Lee, Y., & Geotge. (2024). Rational confinement of ternary layered double hydroxide within three-dimensional graphene aerogel: An electrochemical tool for propyl gallate sensing. *Chemical Engineering Journal, 485*, 149682-149693.

Journal Pre-proof

Kogularasu, S., Sriram, B., Wang, S., & Sheu, J. (2022). Superlattice Stacking by Confinement of the Layered Double Hydroxide/Vanadium Carbide Hybrid Composite. The effect on interlayer anions (SO_4^{2-} and CO_3^{2-}) for comparing the electrochemical sensing of a food adulterant. *ACS Sustainable Chemistry & Engineering, 10*, 15115-15123.

Cui, W., Jiao, Q., Zhao, Y., & Zhou, M. (2012). Preparation of carboxylate-intercalated layered double hydroxides using mixed hydroxides or oxides. *Synthesis and Reactivity in Inorganic and Metal-Organic Chemistry, 42*, 579-582.

Ding, Q., Li, J., Chen, X., Yin, J., Li, S., & Li, H. (2023). Fabrication of the CNTs/NiAl–LDH nanocomposite as a carrier of a luminescent sensor for DNA detection. *Materials Science and Engineering: B, 298*, 116920-116926.

Ding, Y., Du, X., & Zhang, X. (2020). Controllable synthesis of CoFeMo layered double hydroxide nanoarrays for promoting the oxygen evolution reaction. *Dalton Transactions, 49*, 15417-15424.

Duan, F., Wang, Y., Hu, B., Diao, J., Shang, X., Guo, C., He, L., & Wang, M. (2024). Smartphone-assisted portable amperometric immunosensor based on CoMn-LDH for the visualized detection of tylosin. *Microchemical Journal, 204*, 110970-110978.

Scavetta, E., & Tonelli, D. (2005). Amperometric Sensors Based on Synthetic Hydrotalcites and Their Application for Ethanol Detection in Beer. *Electroanalysis, 17*, 363-370.

Evans, D., & Xue, D. (2006). Preparation of layered double hydroxides and their applications as additives in polymers, as precursors to magnetic materials and in biology and medicine. *Chemical Communications, 37*, 485-496.

Fan, G., Li, F., Evans, D., & Duan, X. (2015). Catalytic applications of layered double hydroxides: recent advances and perspectives. *Chemical Society Reviews, 45*, 7040-7066.

Farhat, H., Celier, J., Forano, C., & Mousty, C. (2021). Evaluation of hierarchical glucose oxidase/Co3Mn-CO3 LDH modified electrodes for glucose detection.

Journal Pre-proof

Journal Pre-proof

Electrochimica Acta, 376, 138050-138058.

Wang, F., Zhang, Y., Liang, W., Chen, L., Li, Y., & He, X. (2018). Non-enzymatic Glucose Sensor with High Sensitivity Based on Cu-Al Layered Double Hydroxides. *Sensors and Actuators B: Chemical, 273*, 41-47.

Figueiredo, M., Suarez, E., Petrilli, H., Leroux, F., Taviot-Gueho, C., & Leopoldo Constantino, V. (2022). Limiting content of trivalent iron to form organic-inorganic single-phase layered double hydroxides hybrids by coprecipitation. *Applied clay science, 228*, 106642-106652.

Gao, W., Li, C., Chen, H., Wu, M., He, S., Wei, M., Evans, D., & Duan, X. (2014). Supported nickel-iron nanocomposites as a bifunctional catalyst towards hydrogen generation from $N_2H_4H_2O$. *Green Chemistry, 16*, 1560-1568.

Gao, Z., Ma, T., Chen, X., Liu, H., Cui, L., Qiao, S., Yang, J., & Du, X. (2018). Strongly Coupled CoO Nanoclusters/CoFe LDHs Hybrid as a Synergistic Catalyst for Electrochemical Water Oxidation. *Small, 14*, 1800195-1800203.

Ge, X., Gu, C., Yin, Z., Wang, X., Tu, J., & Li, J. (2016). Periodic stacking of 2D charged sheets: Self-assembled superlattice of Ni-Al layered double hydroxide (LDH) and reduced graphene oxide. *Nano Energy, 20*, 185-193.

Gong, J., Guan, Z., & Song, D. (2013). Biosensor based on acetylcholinesterase immobilized onto layered double hydroxides for flow injection/amperometric detection of organophosphate pesticides. *Biosensors & Bioelectronics, 39*, 320-323.

Guo, R., Li, J., Chen, L., Yu, Z., & Ma, S. (2020). SDC/OS-LDH composite for highly sensitive fluorescence detection of Fe3+ at much low concentration. *Dalton Transactions, 49*, 10413-10420.

Ho, P., Ospitali, F., Luna, G., Fornasari, G., Vaccari, A., & Benito, P. (2020). Coating of Rh/Mg/Al Hydrotalcite-Like Materials on FeCrAl Fibers by Electrodeposition and Application for Syngas Production. *Energy Technology, 8*, 1901018-1901022.

Hou, T., Yan, L., Li, J., Yang, Y., & Zhao, Y. (2019). Adsorption performance and mechanistic study of heavy metals by facile synthesized magnetic layered double oxide/carbon composite from spent adsorbent. *Chemical Engineering Journal, 384*, 123331-123341.

Inayat, A., Klumpp, M., & Schwieger, W. (2011). The urea method for the direct synthesis of ZnAl layered double hydroxides with nitrate as the interlayer anion. *Applied clay science, 51*, 452-459.

Lian, J., Liu, P., & Liu, Q. (2022). Nano-scale minerals in-situ supporting CeO_2 nanoparticles for off-on colorimetric detection of L–penicillamine and Cu^{2+} ion. *Journal of Hazardous Materials, 433*, 128766-128776.

Qin, J., Huang, Y., Shi, M., Wang, H., Han, M., Yang, X., Li, Fei., & Ma, L. (2020). Aqueous-phase detection of antibiotics and nitroaromatic explosives by an alkali-resistant Zn-MOF directed by an ionic liquid. *RSC Advances, 10*, 1439-1446.

Xie, J., Chen, W., Wu, X., Wu, Y., & Lin, H. (2017). Enhanced luminol chemiluminescence by Co-Fe LDH nanoplates and its application in H_2O_2 and glucose detection. *Analytical Methods, 9*, 974-979.

Jiang, S., Zhang, M., Xu, C., Liu, G., Zhang, K., & Zhang, Z. (2024). Recent Developments in Nickel-Based Layered Double Hydroxides for Photo(-/)electrocatalytic Water Oxidation. *Acs Nano, 18*, 16413-16449.

Li, J., Yan, L., Yang, Y., Zhang, X., Zhu, R., & Yu, H. (2019). Insight into the adsorption mechanisms of aqueous hexavalent chromium by EDTA intercalated layered double hydroxides: XRD, FTIR, XPS, and zeta potential studies. *New Journal of Chemistry, 43*, 15915-15923.

Karuppiah, C., Babulal, S., Chen, T., Chen, S., Hsu, L., Farraj, D., Ramaraj, S., Elshikh, M., & Yang, C. (2022). A novel ammonium zinc molybdate layered double hydroxide nanoflakes/vapor grown carbon fibers nanomaterials based electrocatalyst for the monitoring of dimetridazole drug in real samples. *Journal of Environmental Chemical Engineering, 10*, 108227-108241.

Khataee, S., Dehghan, G., Shaghaghi, Z., Khataee, A., & Amini, M. (2024). A novel

Journal Pre-proof

bifunctional electrochemical nanosensor for simultaneous detection of glucose and insulin based on NiO/Co_3O_4@CuAl LDH-MWCNT nanocomposite-modified carbon paste electrode. *Microchemical Journal, 201*, 110644-110656.

Kong, X., Shi, W., Zhao, J., Wei, M., & Duan, X. (2011). Layer-by-layer assembly of electroactive dye/inorganic matrix film and its application as sensor for ascorbic acid. *Talanta, 85*, 493-498.

Kou, X., Li, C., Zhao, Y., Wang, S., & Ma, X. (2018). CO 2 sorbents derived from capsule-connected Ca-Al hydrotalcite-like via low-saturated coprecipitation. *Fuel Processing Technology, 177*, 210-218.

Kumari, S., Sharma, A., Kumar, S., Thakur, A., Thakur, R., Bhatia, S. K., & Sharma, A. K. (2022). Multifaceted potential applicability of hydrotalcite-type anionic clays from green chemistry to environmental sustainability. *Chemosphere, 306*, 135464-135499.

Lei, X., Lu, W., Peng, Q., Li, H., Chen, T., Xu, S., & Zhang, F. (2011). Activated MgAl-layered double hydroxide as solid base catalysts for the conversion of fatty acid methyl esters to monoethanolamides. *Applied Catalysis A General, 399*, 87-92.

Lei, Z., & Guo, B. (2021). 2D Material-Based Optical Biosensor: Status and Prospect. *Advanced Science, 9*, 2102924-2102967.

Li, J., Yu, H., Zhang, X., Zhu, R., & Yan, L. (2020). Crosslinking acrylamide with EDTA-intercalated layered double hydroxide for enhanced recovery of Cr(VI)and Congo red:Adsorptive and mechanistic study. *Frontiers of Environmental Science & Engineering, 14*, 14.

Li, R., Li, Y., Jia, X., Yang, J., Miao, X., Shao, D., Feng, L., Wang, S., Wu, J., & Song, H. (2023). In-situ grown of NiAl-LDHs for self-healing fabric with flame-retardant, UV-protection and antifouling performance. *Ceramics International, 49*, 14635-14644.

Li, Y., Wu, T., Shen, H., Yang, S., Qin, Y., Zhu, Z., Zheng, L., Wen, X., Xia, M., & Yin, X. (2022). Flexible MXene-based Janus porous fibrous membranes for sustainable solar-driven desalination and emulsions separation. *Journal of*

Journal Pre-proof

cleaner production, 347, 131324-131333.

Liang, H., Miao, X., & Gong, J. (2012). One-step fabrication of layered double hydroxides/graphene hybrid as solid-phase extraction for stripping voltammetric detection of methyl parathion. *Electrochemistry Communications, 20*, 149-152.

Ma, L., Islam, S., Xiao, C., Zhao, J., Liu, H., Yuan, M., Sun, G., Li, H., Ma, S., & Kanatzidis, M. (2017). Rapid Simultaneous Removal of Toxic Anions $[HSeO_3]^-$, $[SeO_3]^{2-}$, and $[SeO_4]^{2-}$, and Metals Hg^{2+}, Cu^{2+}, and Cd^{2+} by MoS_4^{2-} Intercalated Layered Double Hydroxide. *Journal of the American Chemical Society, 139*, 12745-12757.

Liu, M., Lv, G., Mei, L., Wei, Y., Liu, J., Li, Z., & Liao, L. (2017). Fabrication of AO/LDH fluorescence composite and its detection of Hg^{2+} in water. *Sci Rep, 7*, 13414-13423.

Liu, M., Zhe, T., Li, F., Zhu, L., Ouyang, S., & Wang, L. (2024). An ultrasensitive electrochemical sensor based on NiFe-LDH-MXene and ruthenium nanoparticles composite for detection of nitrofurantoin in food samples. *Food Chemistry: X, 461*, 140915-140925.

Liu, Q., Ma, J., Wang, K., Feng, T., Peng, M., Yao, Z., Fan, C., & Komarneni, S. (2017). BiOCl and TiO2 deposited on exfoliated ZnCr-LDH to enhance visible-light photocatalytic decolorization of Rhodamine B. *Ceramics International, 43*, 5751-5758.

Liu, P., Liu, D., Liu, Y., & Li, L. (2016). ANTS-anchored Zn-Al-CO_3-LDH particles as fluorescent probe for sensing of folic acid. *Journal of Solid State Chemistry, 241*, 164-172.

Ma, L., Islam, S. M., Liu, H., Zhao, J., Sun, G., Li, H., Ma, S., & Kanatzidis, M. G. (2017). Selective and Efficient Removal of Toxic Oxoanions of As(III), As(V), and Cr(VI) by Layered Double Hydroxide Intercalated with MoS42–. *Chemistry of Materials, 29*, 3274-3284.

Ma, L., Wang, Q., Islam, S. M., Liu, Y., Ma, S., & Kanatzidis, M. G. (2016). Highly

Journal Pre-proof

Journal Pre-proof

Selective and Efficient Removal of Heavy Metals by Layered Double Hydroxide Intercalated with the MoS4(2-) Ion. *Journal of the American Chemical Society, 138*, 2858-2866.

Moloudi, M., Noori, A., Rahmanifar, M., Shabangoli, Y., El-Kady, M., Mohamed, N., Kaner, R., & Mousavi, M. (2023). Layered Double Hydroxide Templated Synthesis of Amorphous NiCoFeB as a Multifunctional Electrocatalyst for Overall Water Splitting and Rechargeable Zinc–Air Batteries. *Advanced Energy Materials, 13*, 2203002-2203018.

Murath, S., Somosl, Z., Kukovecz, A., Konya, Z., Slipos, P., Palinko, I. (2019). Novel route to synthesize CaAl- and MgAl-layered double hydroxides with highly regular morphology. *Journal of Sol-Gel Science and Technology, 89*, 844-851.

Nguyen, H., Uyen, D., Nguyen, D., Tran, V., & Nguyen, T. (2020). Characterization and Evaluation of Ca/Al LDHs Adsorbents Synthesized by a One-Step Hydrothermal Method for Congo Red Removal. *Materials Science Forum, 977*, 195-200.

Prasanna, S., Lin, Y., Ramaraj, S., Dhawan, U., Liu, X., Tung, C., Sakthivel, R., & Chung, R. (2024). 2D/2D heterostructure Ni-Fe LDH/black phosphorus nanosheets with AuNP for noxious substance diphenylamine detection in food samples. *Food Chemistry, 432*, 137295-137308.

Rahman, F., Yang, X., Motswaiso, F., Takanashi, I., Kameda, T., Rahman, M. T., Saito, Y., Kumagai, S., & Yoshioka, T. (2024). Effective synthesis of ethylene urea from CO_2 adsorbed cerium doped Mg–Al layered double hydroxide. *Journal of cleaner production, 434*, 140191-140200.

Salem, M., Khan, A., Manea, Y., Qashqoosh, M., & Alahdal, F. (2023). Highly efficient iodine capture and ultrafast fluorescent detection of heavy metals using PANI/LDH@CNT nanocomposite. *Journal of Hazardous Materials, 447*, 130732-130749.

Sangtam, A., Saikia, P., Goswamee, R., Sinha, D., & Sinha, U. (2022). Green synthesis of mesoporous Ni-Co layered double hydroxide and its application for removal

Journal Pre-proof

of 2,4-dinitrophenol from water: A theoretical study complemented by the first principle density functional theory-Monte-Carlo approach. *Journal of Environmental Chemical Engineering, 10*, 108378-108396.

Sankarlinkam, S., Suresh, I., & Hariharan, G. N., NoelKulandaisamy, Arockia JayalathaRayappan, John Bosco Balaguru. (2024). Ethylenediaminetetraacetic acid intercalated MgAl-layered double-hydroxides nanocomposite as an efficient platform in the development of electrochemical sensor for the detection of iron (II). *Journal of Applied Electrochemistry, 54*, 309-321.

Shan, D., Wang, Y., Zhu, M., Xue, H., Cosnier, S., & Wang, C. (2009). Development of a high analytical performance-xanthine biosensor based on layered double hydroxides modified-electrode and investigation of the inhibitory effect by allopurinol. *Biosensors & Bioelectronics, 24*, 1171-1176.

Shi, H., Chen, L., & Niu, N. (2021). An off-on fluorescent probe based on graphene quantum dots intercalated hydrotalcite for determination of ascorbic acid and phytase. *Sensors and Actuators B: Chemical, 345*, 130353-130353.

Sokol, D., Ivanov, M., Salak, A. N., Grigalaitis, R., Banys, J., & Kareiva, A. (2019). Dielectric properties of Bi-substituted LDHs synthesized by co-precipitation and sol-gel methods. *Materials Science-Poland, 37*, 190-195.

Song, W., Zhang, X., Zhang, L., Yu, Z., Li, X., Li, Y., Cui, Y., Zhao, Y., & Yan, L. (2023). Removal of various aqueous heavy metals by polyethylene glycol modified MgAl-LDH: Adsorption mechanisms and vital role of precipitation. *Journal of Molecular Liquids, 375*, 121386-121395.

Sotiles, A., Baika, L., Grassi, M., & Wypych, F. (2019). Cation Exchange Reactions in Layered Double Hydroxides Intercalated with Sulfate and Alkaline Cations $(A(H_2O)_6)[M\sim(2+)_6Al_3(OH)(18)(SO_4)_2]\cdot 6H_2O$ ($M\sim(2+)$ = Mn, Mg, Zn; $A\sim+$ = Li, Na, K). *Journal of the American Chemical Society, 141*, 531-540.

Staal, L. B., Pushparaj, S. S. C., Forano, C., Prevot, V., Ravnsb?K, D., Bjerring, M., & Nielsen, U. G. (2017). Competitive reactions during synthesis of zinc aluminum layered double hydroxides by thermal hydrolysis of urea. *Journal of Materials*

Journal Pre-proof

Chemistry A, 5, 21795-21806.

Tan, Y., Yi, H., & Zhao, X. T. Y. G. L. W. Z. K. (2024). Layered double hydroxides for air pollution control: Applications, mechanisms and trends. *Journal of cleaner production, 436*, 140635.140631-140635.140620.

Tang, J., Mu, B., Zong, L., & Wang, A. (2018). One-step synthesis of magnetic attapulgite/carbon supported NiFe-LDHs by hydrothermal process of spent bleaching earth for pollutants removal. *Journal of cleaner production, 172*, 673-685.

Taviot-Gueho, C., Prevot, V., Forano, C., Renaudin, G., Mousty, C., & Leroux, F. (2018). Tailoring Hybrid Layered Double Hydroxides for the Development of Innovative Applications. *Advanced Functional Materials, 28*, 1703868.1703861-1703868.1703833.

Valeikiene, L., Roshchina, M., Grigoraviciute-Puroniene, I., Prozorovich, V., Zarkov, A., Ivanets, A., & Kareiva, A. (2020). On the Reconstruction Peculiarities of Sol–Gel Derived Mg2−xMx/Al1 (M = Ca, Sr, Ba) Layered Double Hydroxides. *Crystals, 10*, 470-490.

Wang, D., Zhang, X., Liu, C., Cheng, T., Wei, W., & Sun, Y. (2015). Transition metal-modified mesoporous Mg-Al mixed oxides: Stable base catalysts for the synthesis of diethyl carbonate from ethyl carbamate and ethanol. *Applied Catalysis A: General, 505*, 478-486.

Wang, D., Zhang, X., Cong, X., Liu, S., & Zhou, D. (2018). Influence of Zr on the performance of Mg-Al catalysts via hydrotalcite-like precursors for the synthesis of glycerol carbonate from urea and glycerol. *Applied Catalysis A: General, 555*, 36-46.

Wang, H., Bing, W., Chen, C., Yang, Y., & Wei, M. (2020). Geometric effect promoted hydrotalcites catalysts towards aldol condensation reaction. *Chinese Journal of Catalysis, 41*, 1279-1287.

Wang, J., Mei, X., Huang, L., Zheng, Q., Qiao, Y., Zang, K., Mao, S., Yang, R., Zhang, Z., & Gao, Y. (2015). Synthesis of layered double hydroxides/graphene oxide

Journal Pre-proof

nanocomposite as a novel high-temperature CO2 adsorbent. *Journal of Energy Chemistry, 24*, 127-137.

Wang, J., Sun, X., Hu, H., Shen, T., Liu, G., Li, Z., Cao, D., & He, L. (2022). Electrodeposition of Defect-Rich Ternary NiCoFe Layered Double Hydroxides: Fine Modulation of Co(3+) for Highly Efficient Oxygen Evolution Reaction. *Chemistry - A European Journal, 28*, e202103601-e202103608.

Wang, L., Liu, J., Zhou, Y., Song, Y., He, J., & Evans, D. G. (2010). Synthesis of CoFe alloy nanoparticles embedded in a MgO crystal matrix using a single-source inorganic precursor. *Chemical Communications, 46*, 3911-3913.

Wang, P., Zhang, X., Zhou, B., Meng, F., Wang, Y., & Wen, G. (2023). Recent advance of layered double hydroxides materials: Structure, properties, synthesis, modification and applications of wastewater treatment. *Journal of Environmental Chemical Engineering, 11*, 111191-111231.

Wang, Q., & O'Hare, D. (2012). Recent advances in the synthesis and application of layered double hydroxide (LDH) nanosheets. *Chem Rev, 112*, 4124-4155.

Wang, Y., Wang, T., Zhang, R., Liu, Q., & Sun, X. (2020). CuO@CoFe Layered Double Hydroxide Core–Shell Heterostructure as an Efficient Water Oxidation Electrocatalyst under Mild Alkaline Conditions. *Inorganic Chemistry, 59*, 9491–9495.

Wu, H., Wan, X., Niu, J., Cao, Y., Wang, S., Zhang, Y., Guo, Y., Xu, H., Xue, X., & Yao, J. (2024). Enhancing iron content and growth of cucumber seedlings with MgFe-LDHs under low-temperature stress. *J Nanobiotechnology, 22*, 1-20.

Wu, L., Zhou, X., Wan, G., Tang, Y., Shi, S., Xu, X., & Wang, G. (2021). Novel hierarchical CuNiAl LDH nanotubes with excellent peroxidase-like activity for wide-range detection of glucose. *Dalton Transactions, 50*.

Xu, M., & Wei, M. (2018). Layered Double Hydroxide-Based Catalysts: Recent Advances in Preparation, Structure, and Applications. *Advanced Functional Materials, 28*, 1802943.1802941-1802943.1802920.

Xu, L., Lian, M., Chen, X., Lu, Y., & Yang, W. (2017). Amperometric sensing of

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hydrogen peroxide via an ITO electrode modified with gold nanoparticles electrodeposited on a CoMn-layered double hydroxide. *Mikrochimica Acta: An International Journal for Physical and Chemical Methods of Analysis, 184*, 3989-3996.

Xu, Z., Wu, Y., Wang, Y., Hu, J., Ma, Y., Zhang, Z., Huang, H., Wei, J., Shi, C., & Yu, Q. (2024). Insight into ion exchange behavior of LDHs: Asynchronous chloride adsorption and intercalated ions release processes. *Cement & concrete composites, 147*, 105433-105449.

Yang, S., Deng, K., Zhang, J., Bai, C., Peng, J., Fang, Z., & Xu, W. (2021). Synergy effect of Ag plasmonic resonance and heterostructure construction enhanced visible-light photoelectrochemical sensing for quercetin. *Electrochimica Acta, 371*, 137772-137782.

Yang, W., Li, J., Wang, M., Sun, X., Liu, Y., Yang, J., & Ng, D. H. L. (2020). A colorimetric strategy for ascorbic acid sensing based on the peroxidase-like activity of core-shell Fe(3)O(4)/CoFe-LDH hybrid. *Colloids Surf B Biointerfaces, 188*, 110742-110751.

Yang, Y., Ou, Y., Yang, Y., & Wei, X. (2019). Modulated transition metal-oxygen covalency in the octahedral sites of CoFe layered double hydroxides with vanadium doping leading to highly efficient electrocatalysts. *Nanoscale, 11*, 23296-23303.

Yu, J., Lu, K., Wang, C., Wang, Z., Fan, C., Bai, G., Wang, G., & Yu, F. (2021). Modification of NiFe layered double hydroxide by lanthanum doping for boosting water splitting. *Electrochimica Acta, 390*, 138824-138835.

Yu, J., Wang, Q., O'Hare, D., & Sun, L. (2017). Preparation of two dimensional layered double hydroxide nanosheets and their applications. *Chemical Society Reviews, 46*, 5950-5974.

Yu, Y., & Yang, W. (2022). In Situ Synthesis of MnMgFe-LDH on Biochar for Electrochemical Detection and Removal of Cd(2+) in Aqueous Solution. *Molecules, 27*, 7875-7890.

Journal Pre-proof

Journal Pre-proof

Wu, Y., Liu, S., Xiao, Y., Li, Q., Xiang, Q., Song, X., Wu, S., Wang, K., & Yang, Y. (2023). A versatile fluorescence sensor for DNA detection based on layered double hydroxides and exonuclease III. *New Journal of Chemistry, 47*, 13228-13234.

Zhang, C., Liang, X., Lu, Y., Li, H., & Xu, X. (2020). Performance of CuAl-LDH/Gr Nanocomposite-Based Electrochemical Sensor with Regard to Trace Glyphosate Detection in Water. *Sensors (Basel), 20*, 4146-4161.

Zhang, J., Yu, L., Chen, Y., & Lou, X. W. D. (2020). Designed Formation of Double-Shelled Ni–Fe Layered-Double-Hydroxide Nanocages for Efficient Oxygen Evolution Reaction. *Advanced Materials, 32*, 1906432-1906438.

Zhang, W., Ding, M., Zhang, X., & Shang, H. (2022a). Biosynthesis-mediated Ni-Fe-Cu LDH-to-sulfides transformation enabling sensitive detection of endogenous hydrogen sulfide with dual-readout signals. *Analytic Chimica Acta, 1250*, 340981-340989.

Zhang, W., Ding, M., Zhang, X., & Shang, H. (2022b). Biosynthesis-mediated Ni-Fe-Cu LDH-to-sulfides transformation enabling sensitive detection of endogenous hydrogen sulfide with dual-readout signals. *Analytica Chimica Acta, 1229*, 340390-340396.

Zhang, W., Wu, Y., Qi, J., Chen, M., & Cao, R. (2017). A Thin NiFe Hydroxide Film Formed by Stepwise Electrodeposition Strategy with Significantly Improved Catalytic Water Oxidation Efficiency. *Advanced Energy Materials, 7*, 1602547-1602553.

Zhang, X., Wang, F., Li, Z., Hu, B., Zheng, Q., Piao, Y., Feng, L., & Cao, J. (2023). Dual-mode electrochemical/colorimetric microfluidic sensor integrated tetrahedral DNA nanostructures with Au/Ni-Co LDH NCs nanozyme for ultrasensitive detection of aflatoxin B1. *Sensors and Actuators B: Chemical, 393*, 134322-134330.

Zhang, X., Wang, F., Zhi, H., Zhao, J., Wan, P., & Feng, L. (2022). Electrochemical "Signal On/Off" Paper-Based Aptasensor for Ochratoxin a Detection Based on

Mxene-Au and Pt@Nico-Ldh-Catalyzed Signal Amplification. *Social Science Electronic Publishing, 368*, 132161-132169.

Zhao, Y., Yan, Y., Liu, C., Zhang, D., Wang, D., Ispas, A., Bund, A., Du, B., Zhang, Z., Schaaf, P., & Wang, X. (2022). Plasma-Assisted Fabrication of Molecularly Imprinted NiAl-LDH Layer on Ni Nanorod Arrays for Glyphosate Detection. *Acs Applied Materials & Interfaces, 14*, 35704-35715.

Zhao, Z., & Zheng, L. (2024). Ultrasound assisted synthesis of CoFe-LDH for the simultaneous electrochemical detection of dopamine and acetaminophen. *Inorganic Chemistry Communications, 164*, 112435-112442.

Li, Z., Zeng, H., Cao, X., Li, H., Long, Y., Feng, B., & Lv, S. (2021). High-sensitive sensor for the simultaneous determination of phenolics based on multi-walled carbon nanotube/NiCoAl hydrotalcite electrode material. *Mikrochim Acta, 188*, 308-321.

Zheng, J., Fan, C., Li, X., Yang, Q., Wang, D., Duan, A., Pan, S., & You, F. (2024). Intelligent multifunctional ruthenium monoatomic/ZnAl-LDH photocatalysts for simultaneous detection and rapid degradation of antibiotics. *Journal of Environmental Management, 353*, 120156-120174.

Zhong, H., Liu, T., Zhang, S., Li, D., Tang, P., Alonso-Vante, N., & Feng, Y. (2019). Template-free synthesis of three-dimensional NiFe-LDH hollow microsphere with enhanced OER performance in alkaline media. *Journal of Energy Chemistry, 33*, 130-137.

Zhu, J., Wang, F., Chen, J., & Liu, C. (2023). An efficient biosensor using a functionalized microneedle of Cu(2)O-based CoCu-LDH for glucose detection. *RSC Advances, 13*, 32558-32566.

Zhu, Y., Qian, J., Xu, K., Ouyang, W., Yang, J., & Yang, N. (2024). Hollow nanocages heterostructured NiCo-LDH/MWCNTs electrocatalyst for highly sensitive and non-invasive detection of saliva glucose. *Chemical Engineering Journal, 485*, 149795-1497806.

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Captions for all Figures

Figure 1. Statistics on the amounts of publications related to the topics of A) food contaminant detection, B) nano-sensors, and C) LDHs in different countries in the last decade. Summary of annual publications in D) food contaminant detection, E) nano-sensors, and F) LDHs in different countries from 2014 to present. G) Statistics of publications related to LDHs sensing in different years and fields (Source of data: Web of Science).

Figure 2. Layered structure and composition of LDHs (Kumari, et al., 2022; P. Wang, et al., 2023).

Figure 3. Lamellar structure of LDHs. A. Major divalent and trivalent metal types (R. Li, et al., 2023; Q. Wang & O'Hare, 2012; H. Wu, et al., 2024). B. Band structure of LDHs with different metal compositions (Y. Li, et al., 2022).

Figure 4. Synthesis strategies of LDHs (X. Ge, ChangdongYin, ZongyouWang, XiuliTu, JiangpingLi, Ju, 2016; Inayat, et al., 2011; Sangtam, et al., 2022; P. Wang, et al., 2023; J. Zhang, et al., 2020; Wei Zhang, et al., 2017; Zhong, et al., 2019).

Figure 5. Activity source and regulation of LDHs (Taviot-Gueho, et al., 2018; Wang, Dengfeng, Zhang, & Xuelan, 2015; D. Wang, Zhang, Cong, Liu, & Zhou, 2018; P. Wang, et al., 2023).

Figure 6. The activity sources of LDHs (Lijiao, et al., 2017; Lijiao Ma, et al., 2017; L. Ma, et al., 2016; Wang, et al., 2015; D. Wang, et al., 2018; H. Wang, et al., 2020).

Figure 7. The activity regulation of LDHs (Y. T. A, et al., 2020; B, et al., 2020; Hou, et al., 2019; Jing, et al., 2019; J. Li, et al., 2020).

Figure 8. Heavy metal ions detection. A-B. Fe^{3+} (Sankarlinkam, et al., 2024). C-F. Cr^{6+}, Mn^{3+}, and Al^{3+} (Salem, et al., 2023). G-H. Fe^{3+} (Guo, et al., 2020).

Figure 9. Pesticide residues detection. A-B. methyl parathion(Liang, et al., 2012). C-D. organophosphate pesticides(Gong, et al., 2013). E-G. Glyphosate(Y. Zhao, et al., 2022). H-I. Glyphosate(C. Zhang, et al., 2020).

Figure 10. Mycotoxins detection. A-B. Ochratoxin a (X. Zhang, et al., 2022). C-D. aflatoxin B1 (X. Zhang, et al., 2023).

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Figure 11. Antibiotics detection. A-B. dimetridazole drug (Karuppiah, et al., 2022). C-D. Tetracycline (Zheng, et al., 2024). E. Tylenol (Duan, et al., 2024).

Figure 12. Carcinogens detection. A-B. Dopamine and acetaminophen (Z. Zhao & Zheng, 2024). C-D. Diphenylamine (Prasanna, et al., 2024). E. Nitrite oxidizing bacteria (Aziz, et al., 2022). F-G. Nitrofurantoin (M. Liu, et al., 2024). H-I. Quercetin (S. Yang, KaiqiangZhang, JianBai, ChaopengPeng, JiaxiFang, ZhiliXu, Wenyuan, 2021).

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Table 2. Applications of LDHs in the detection of food contaminant.

Type of traget	Tragets	LDHs	Synthesis strategies	Detection method	Linear range	LOD	References
Heavy metal ions	Cu^{2+}	CeO_2-LDHs	Hydrothermal method	Colorimetric method	/	9.80 μM	(Jiajia Lian, 2022)
	Fe^{2+}	MgAl-LDHs	Hydrothermal method	Electrochemical method	0.1~102.1 μM	50 nM	(Sankarlinkam, Suresh, & Hariharan, 2024)
	Mn^{7+}, Cr^{6+}, and Al^{3+}	PANI/LDHs@CNT	Coprecipitation method	Fluorescent method	/	51, 59, and 81 nM	(Salem, Khan, Manea, Qashqoosh, & Alahdal, 2023)
	Fe^{3+}	SDC / OS Mg / Al-LDHs	/	Fluorescent method	0.18~1.78 μM（0.01~0.1 ppm）	6.12×10^{-8} M	(Guo, Li, Chen, Yu, & Ma, 2020)
	Cd^{2+}	MnMgFe-LDHs/BC	Coprecipitation method	/	/	0.03 ng/L	(Y. Yu & Yang, 2022)

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	As^{3+}	3D Fe-Co-LDHs/MXene	Hydrothermal method	Electrochemical method	1~1000 ppt	0.9 ppt	(Chen, et al., 2023)
	Hg^{2+}	AO-LDHs	/	Fluorescent method	0.5~150 mM	/	(M. Liu, et al., 2017)
Pesticide residue	Methyl parathion (MP)	LDHs-GNs	/	Electrochemical method	/	0.6 ng/mL	(Liang, Miao, & Gong, 2012)
	Methyl parathion (MP)	AChE-LDHs	/	Electrochemical method	0.005~0.3 µg/mL	0.6 ng/mL	(Gong, Guan, & Song, 2013)
	Glyphosate	MI-NiAl-LDHs	Electrodeposition method	Electrochemical method	10.0 nmol/L~1.0 µmol/L	3.1 nmol/L	(Y. Zhao, et al., 2022)
	Glyphosate	CuAl-LDHs/GrNC	/	Electrochemical method	2.96×10^{-9}~1.18×10^{-6} mol/L	1×10^{-9} mol/L	(C. Zhang, Liang, Lu, Li, & Xu, 2020)
	Glyphosate	NiAl-LDHs	Coprecipitation method	Electrochemical method	0.1~1µM and 3~100 µM	0.081 µM and 0.08 µM	(L. Ge, et al., 2022)
Mycotoxin	Ochratoxin A (OTA)	Pt@NiCo-LDH	/	Electrochemical method	20 fg/mL~100 ng/mL	8.9 fg/mL	(X. Zhang, et al., 2022)
	Aflatoxin B1	Au/Ni-Co LDHs	/	Electrochemical-	0.2 pg/mL~100	/	(X. Zhang, et

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	(AFB$_1$)	NC		colorimetric method	ng/mL and 50 pg/mL~100 ng/mL		al., 2023)
Antibiotic	Ciprofloxacin (CPX)	QDs-LDHs	/	/	2.5×10^{-8}~1.2×10^{-5} mol/L	4.2×10^{-8} mol/L	(Carvalho, et al., 2019)
	Dimetronidazole（DMZ）	AZnMo-LDHs	Coprecipitation method	/	0.25~570 μM	0.021 μM	(Karuppiah, et al., 2022)
	Tetracycline hydrochloride (TC)	RuZnAl-LDHs	/	/	/	1.0 mg/L	(Zheng, et al., 2024)
	Tylenol (TYL)	CoMn-LDHs	/	Electrochemical-immunoassay method	1.0 fg/mL~1.0 ng/mL	0.89 fg/mL	(Duan, et al., 2024)
Carcinogens	Dopamine (DA), and P-acetyl amino (AP)	CoFe-LDH/GCE	/	Electrochemical method	2~900 μM	0.08 μM and 0.37 μM	(Z. Zhao & Zheng, 2024)
	Diphenylamine (DPA)	AuNP/Ni-Fe LDHs/BPNSs	/	Electrochemical method	0.0125~1003.825 μM	4.63 nM	(Prasanna, et al., 2024)

Others	Nitrates	CuNiAl LDHs	/	Electrochemical method	5 nM~40 µM and 75 µM~2.4 mM	0.02 nM	(Aziz, et al., 2022)
	Nitrofurantoin (NFT)	Ru/NiFe-LDHs-MXene/SPCE	Electrodeposition method	Electrochemical method	/	2.2 nM	(M. Liu, et al., 2024)
	Quercetin	NiCo-LDHs	Hydrothermal method	Ionospheric resonance method	1.0×10^{-10}~5.0×10^{-6} mol/L	0.075 nmol/L	(S. Yang, KaiqiangZhang, JianBai, ChaopengPeng, JiaxiFang, ZhiliXu, Wenyuan, 2021)
	Glucose	CuAl-LDHs/GCE	Coprecipitation method	Electrochemical method	0.1~240 µmol/L	0.02 µmol/L	(Fenfen, et al., 2018)
	Glucose and H_2O_2	Co-Fe-LDHs	/	Chemiluminescence method	1.0×10^{-8}~3.0×10^{-6} mol/L	5.0×10^{-9} mol/L and 2.0×10^{-8}	(Jian Xin Xie, 2017)

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					mol/L	
H_2O_2	CoMn-LDHs	Coprecipitation method	Electrochemical method	0.1 μM~1.27 mM	0.06 μM	(Y. Y. Xu, Wensheng, 2017)
Glucose	CuNiAl LDHs	Coprecipitation method	Colorimetric method	10~200 μM	2.9 μM	(L. Wu, et al., 2021)
Glucose	CoCu-LDHs	/	Electrochemical method	0.03~0.40 mM and 0.40~6.00 mM	0.46 μM	(J. Zhu, Wang, Chen, & Liu, 2023)
Glucose	NiCo-LDHs	/	Electrochemical method	0.1~3000 μM and 3000~9231.8 μM	0.03 μM	(Y. Zhu, et al., 2024)
Glucose	Co_3Mn-CO_3-LDHs	/	/	/	0.02 mM	(Farhat, 2021)
Glucose	CoAl-LDHs/MFe_2O_4	/	Colorimetric method	25~400 μM	13 μM	(W. Y. A, et al., 2019)

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	Glucose and insulin	NiO/CoO CuAl LDHs@MWCNT	/	Electrochemical method	0.01~26.5 mM and 0.02~70.0 nM	2.8 µM and 17.3 pM	(Khataee, Dehghan, Shaghaghi, Khataee, & Amini, 2024)
	Ascorbic acid	MgAl-CO_3-LDHs	/	Chemiluminescence method	5.0~5000 nM	0.5 nM	(Erika, Scavetta, Domenica, & Tonelli, 2005)
	Ascorbic acid	GQDs-LDHs	Hydrothermal method	Fluorescent method	5~300 µmol/L	1.73 µmol/L	(Shi, Chen, & Niu, 2021)
	Ascorbic acid	NGB-LDHs	/	Fluorescent method	1.2~55.2 µM	0.51 µM	(Kong, Shi, Zhao, Wei, & Duan, 2011)
	Ascorbic acid	Fe_3O_4/CoFe-LDHs	/	Colorimetric method	0.5~10 µM	0.2 µM	(W. Yang, et al., 2020)
	P-hydroquinone (HQ), P-catechol (CC),	NiCoAl-LDHs	/	Electrochemical method	1.50~600 µM and 5.00~1.03 × 10^3	0.4, 0.8, and 6 × 10^{-3} µM	(Zhen Li 2021)

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	and bisphenol A(BPA)				μM		
	Xanthine	XnOx-LDHs	Anion exchange method	Electrochemical method	1×10^{-6}~2×10^{-4} M	1×10^{-7} M	(Shan, et al., 2009)
	DNA	Ni/Al-LDHs	/	Fluorescent method	10~400 nM	1.38 nM	(Yuanyi, et al., 2023)
	DNA	CNTs/NiAl-LDHs	Hydrothermal method	Fluorescent method	4.4~26.3 μg/mL	6.09×10^{-8} g/mL	(Q. Ding, et al., 2023)
	Folic acid	Zn-Al-CO_3-LDHs	/	/	1~200 μM	0.1 μM	(Y. L. Liu, Lei, 2016)
	Hydrogen sulfide (H_2S)	Ni-Fe-Cu LDHs	/	Electrochemical-photothermal method	0.1~90 μM and 50~400 μM	0.09 μM	(W. Zhang, Ding, Zhang, & Shang, 2022b)

Note: The symbol of " / " indicates not stated in the original study.

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Table 1. The comparison of the advantages and disadvantages of LDHs synthesis strategies.

Synthesis strategies	**Influencing factors**	**Advantages**	**Disadvantages**	**References**
Coprecipitation method	pH value	Simple and easy to operate, etc.	Uneven particle size and poor crystallinity, etc.	(Chengqian, et al., 2022; Kou, et al., 2018; Song, et al., 2023; P. Wang, et al., 2023)
Urea hydrolysis method	Temperature	Uniform particle size distribution and high crystallinity, etc.	Prone to the formation of interfering substances such as CO_3^{2-}, etc.	(Inayat, et al., 2011; Staal, et al., 2017; P. Wang, et al., 2023)
Hydrothermal method	Temperature and pressure	High crystallinity, uniform morphology and size, etc.	Low yield and high cost, etc.	(Nguyen, et al., 2020; Tang, et al., 2018; Zhong, et al., 2019)
Sol-gel method	pH value and type of precipitating agent	High purity, large specific surface area, strong uniformity, and ease of functionalization and regulation, etc.	High cost and long duration, etc.	(Murath, 2019; Rahman, et al., 2024; Sangtam, et al., 2022; Sokol, et al., 2019; Valeikiene, et al., 2020)

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Template method	Types of templates	Easy to achieve functionalization and customization regulation, etc.	Uneven particle size, low purity, and unstable structure, etc.	(Jiang, et al., 2024; Moloudi, et al., 2023; J. Zhang, et al., 2020)
Interlayer anion exchange	Types of ions	Easy to achieve functionalization and regulation, etc.	Poor stability, high cost, and strong environmental dependency, etc.	(X. Ge, ChangdongYin, ZongyouWang, XiuliTu, JiangpingLi, Ju, 2016; Sotiles, et al., 2019; P. Wang, et al., 2023; Z. Xu, et al., 2024)
Electrodeposition method	Temperature, pH value, and pressure	Simple operation, and low cost, etc.	Uneven morphology, time-consuming, and complicated operation, etc.	(Ho, et al., 2020; J. Wang, et al., 2022; Wei Zhang, et al., 2017)

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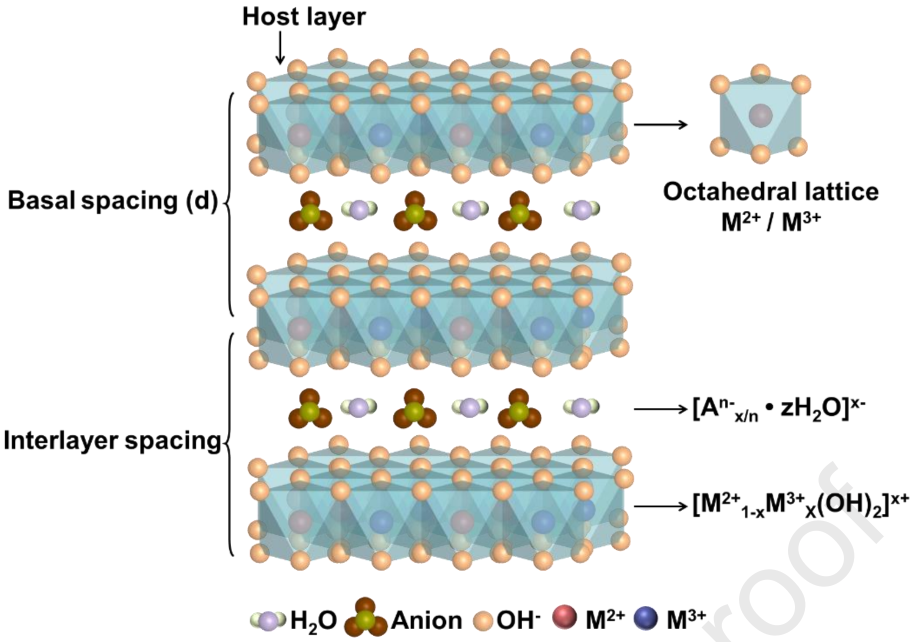

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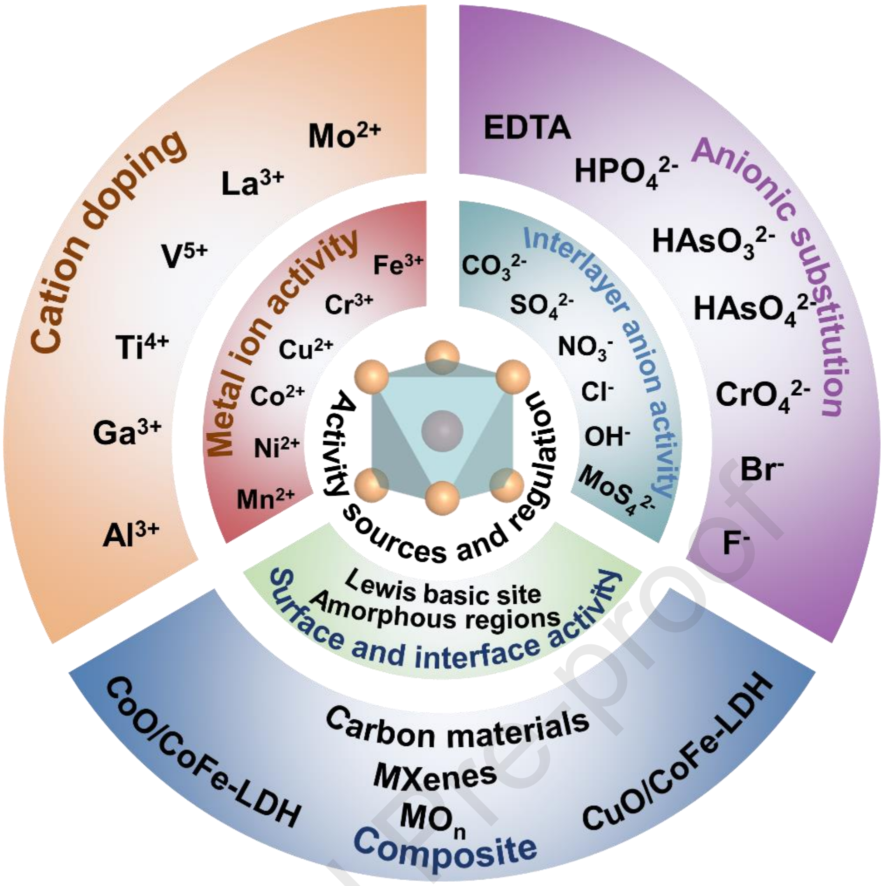

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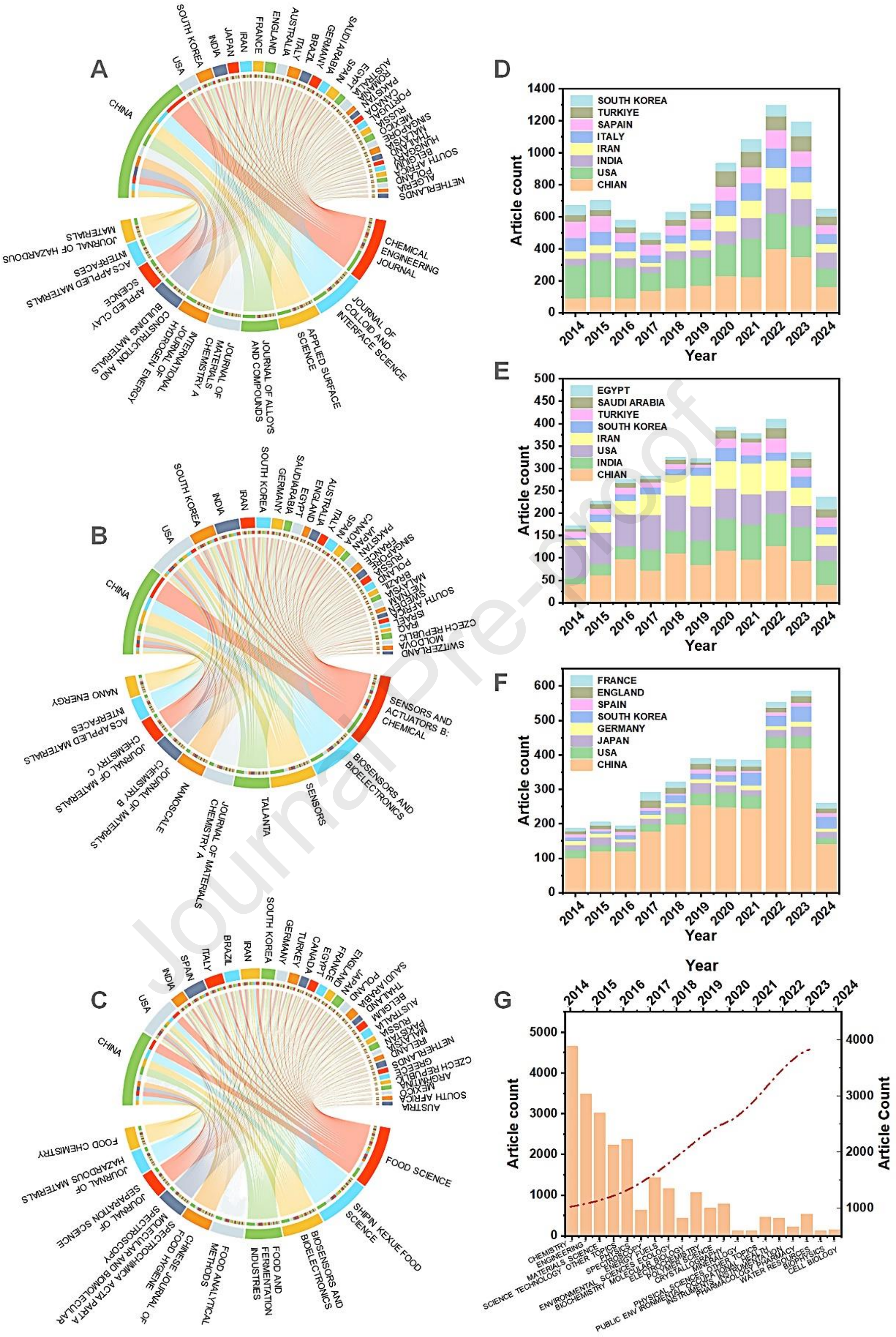

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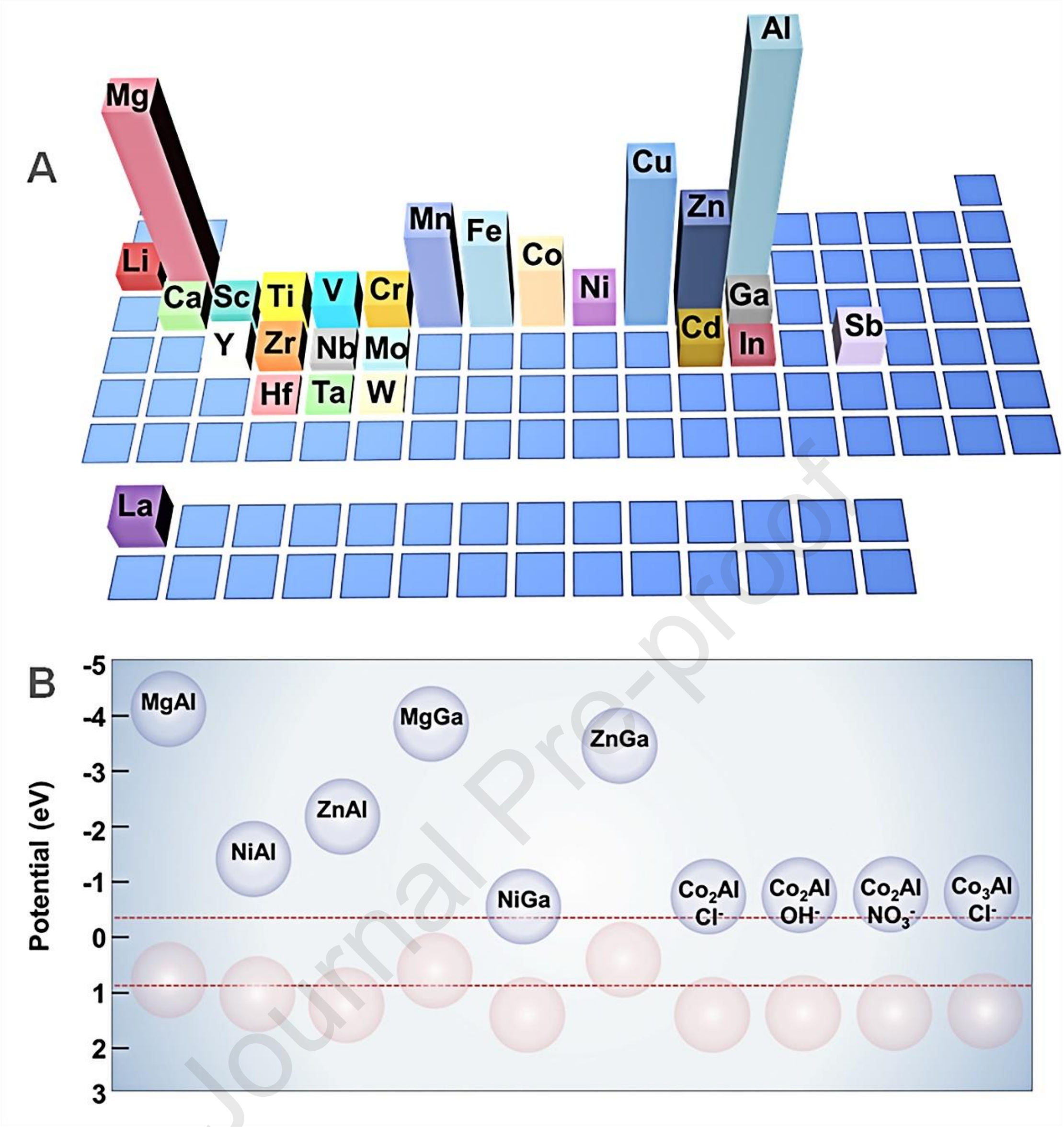

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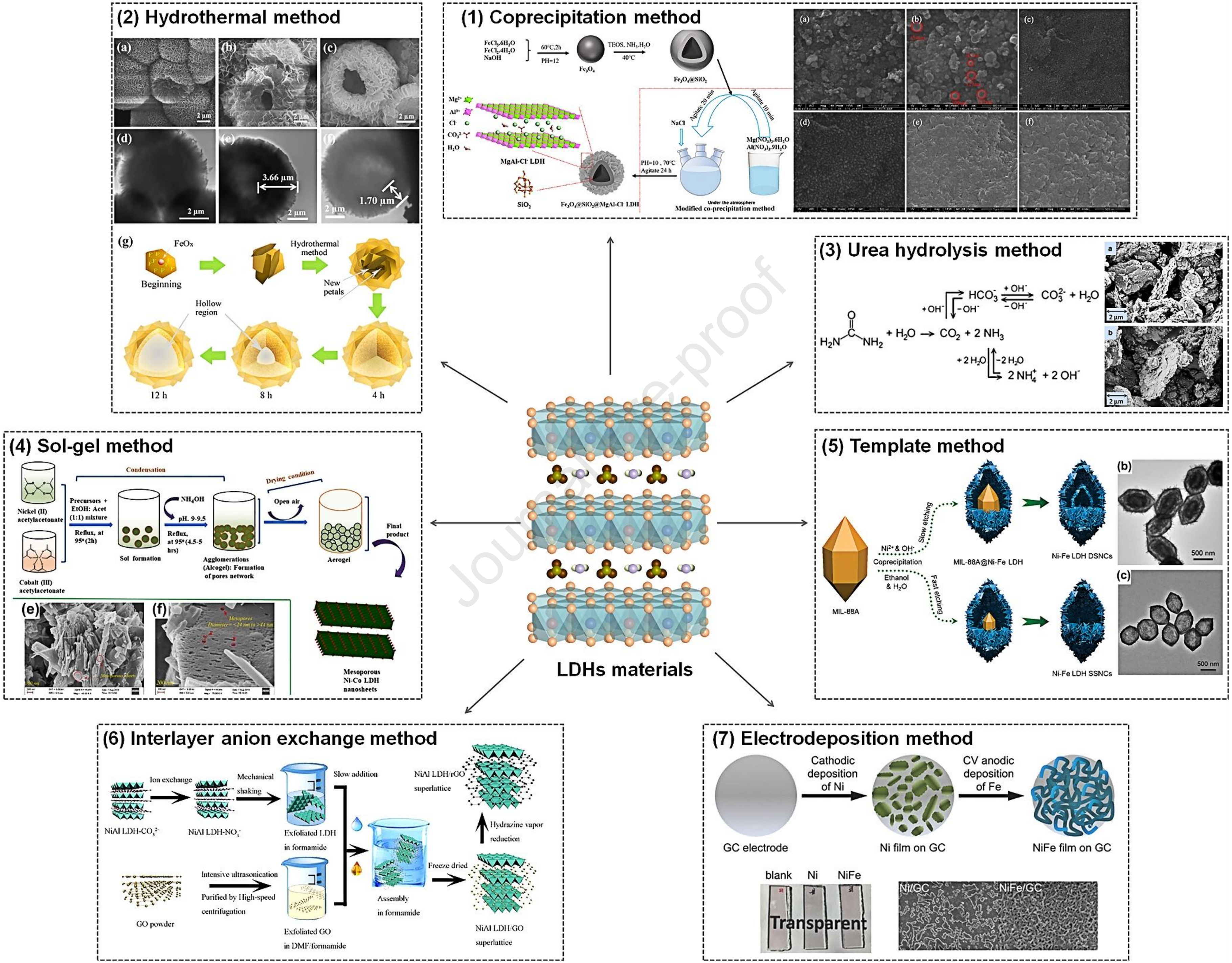

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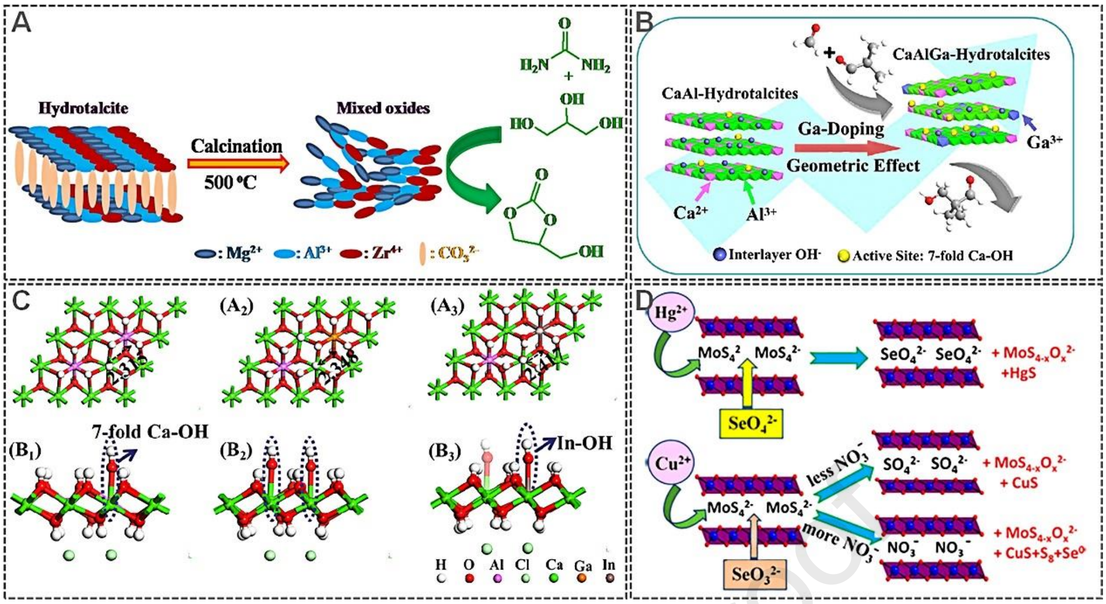

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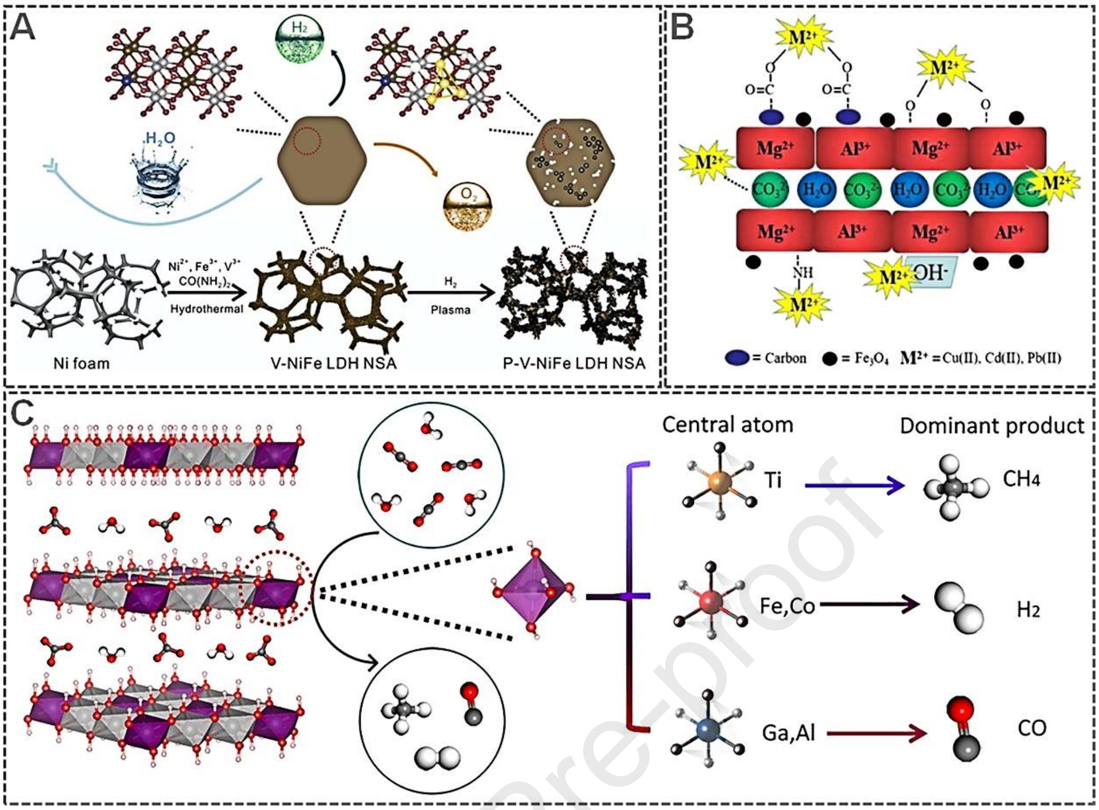

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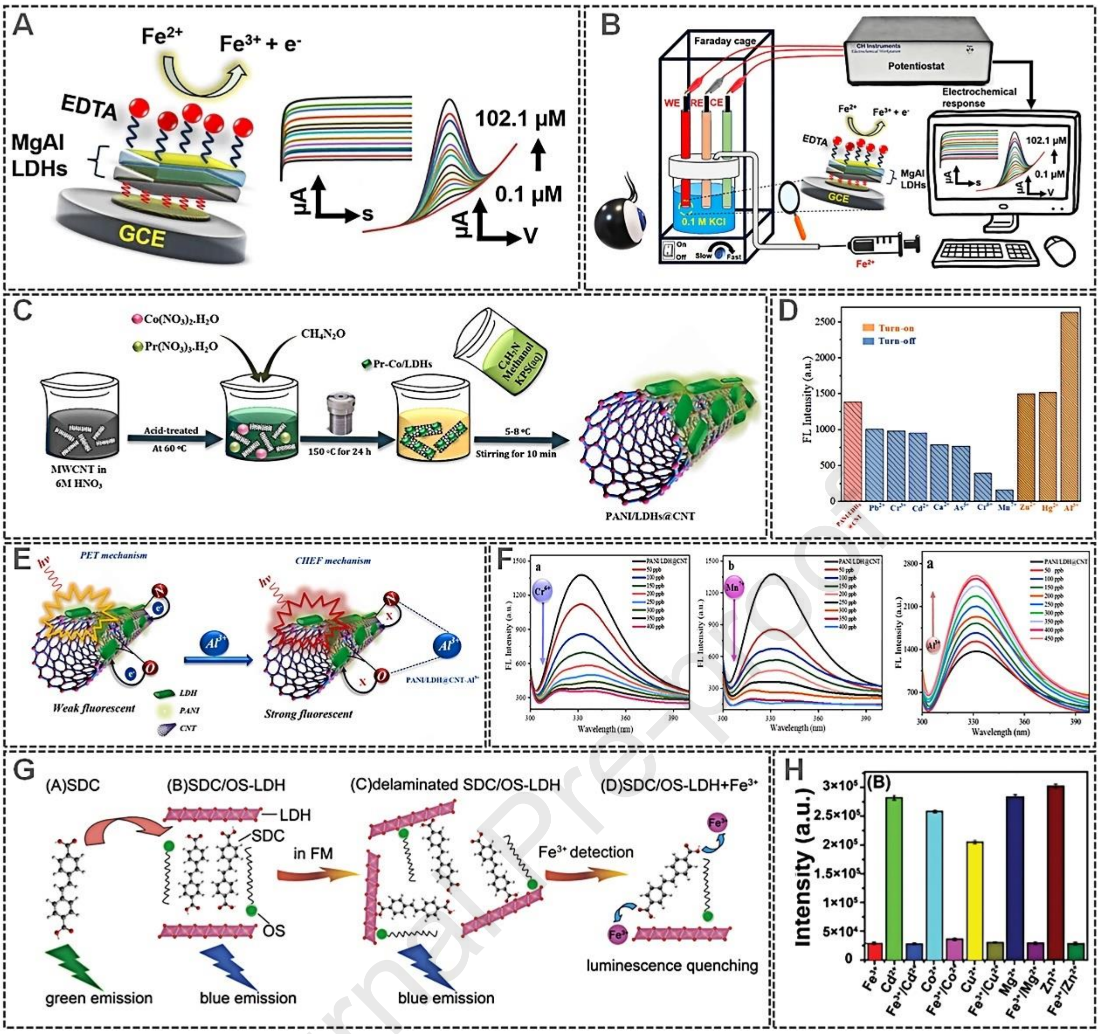

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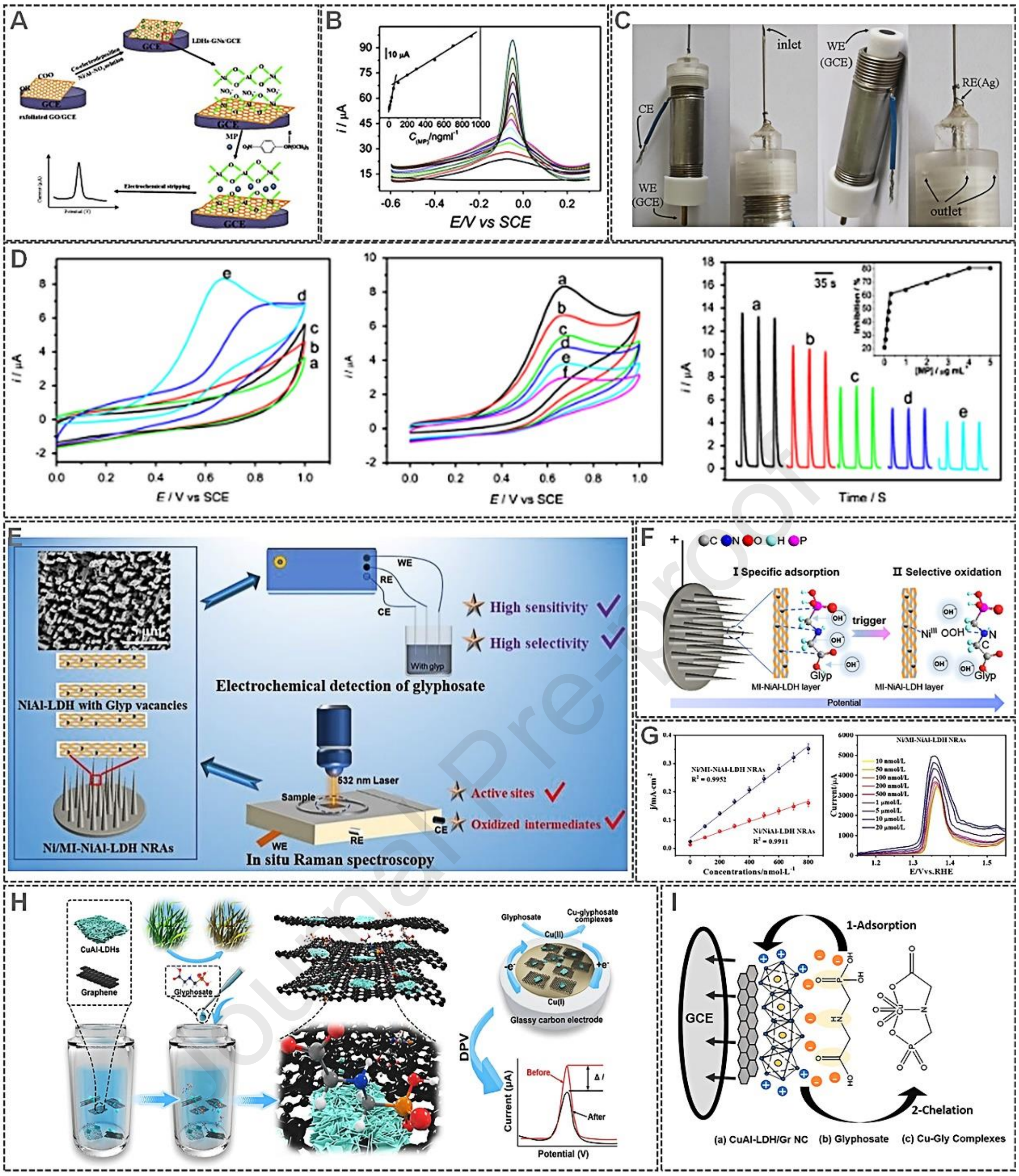

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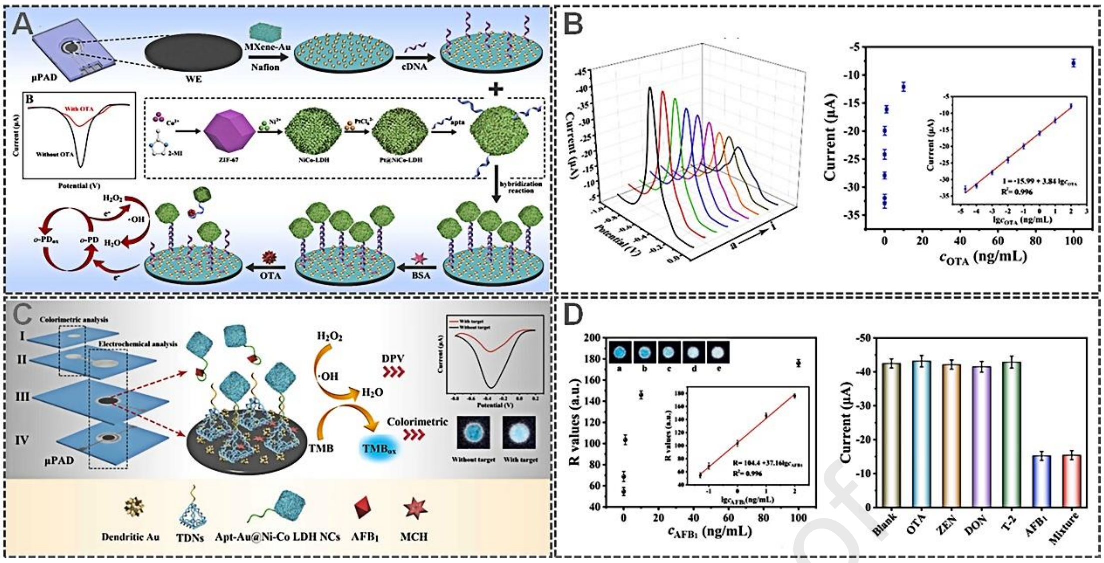

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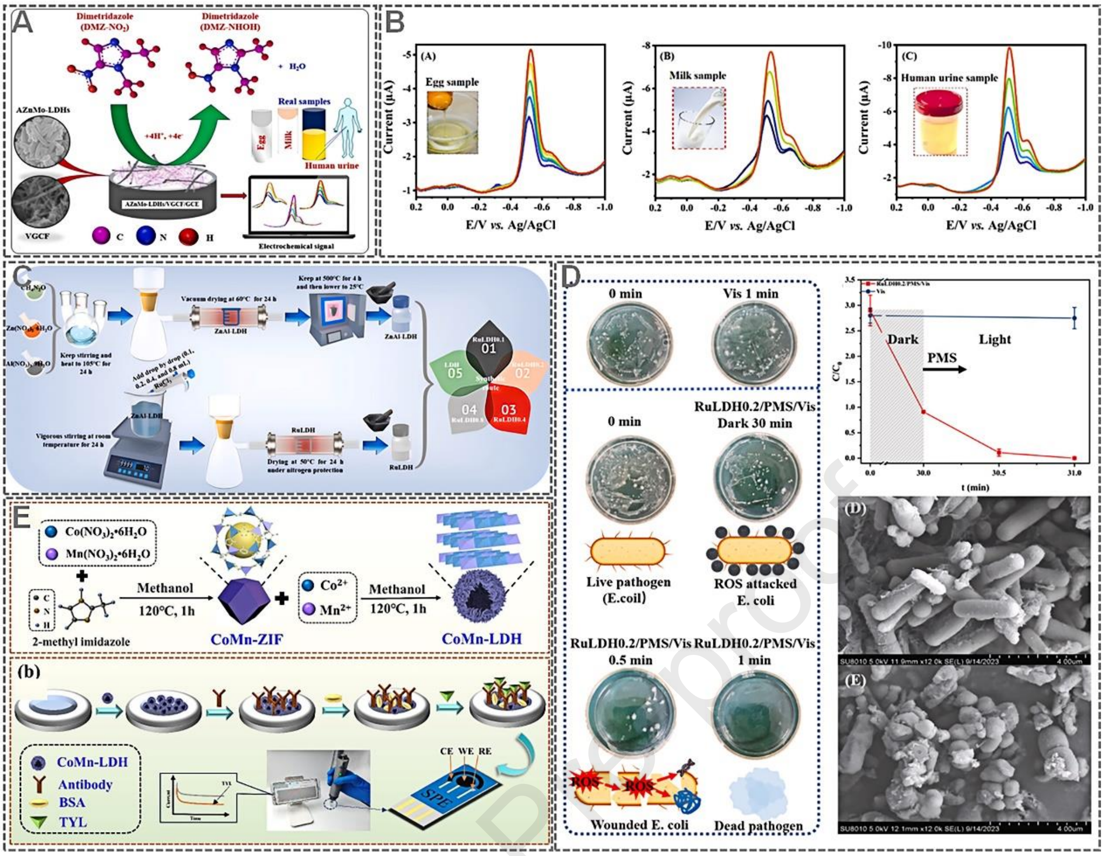

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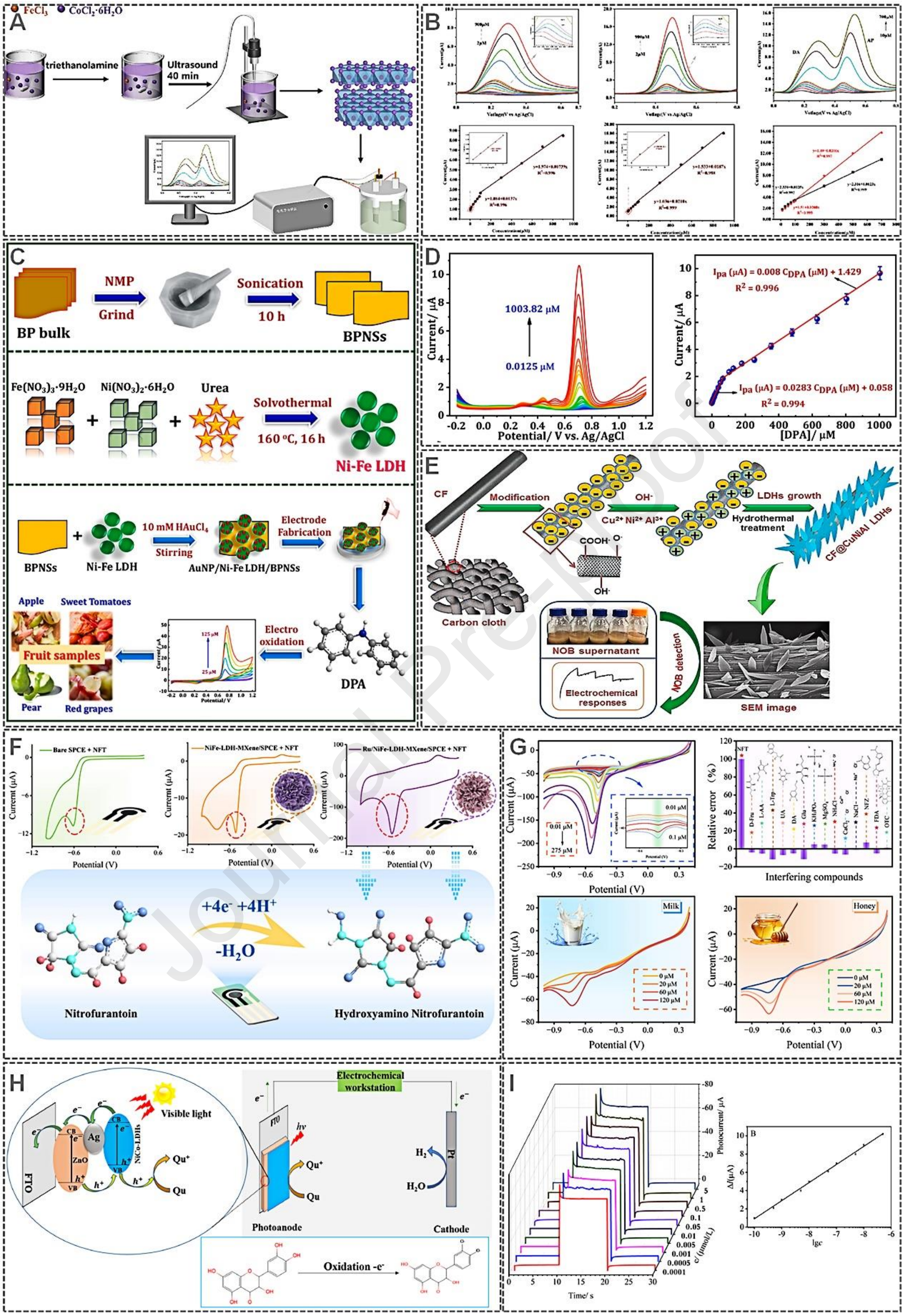

Highlights

- The synthesis methods, activity sources, and properties of LDHs were explained.
- Comprehensive regulation strategies for LDH activity were thoroughly discussed.
- LDHs demonstrate significant potential for detecting food contaminants.
- The challenges and future trends of LDHs application were reviewed.

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