

Progress of Cds/Nife-Ldh Heterostructure and Its Applications in Photocatalysis

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Abstract: As a renewable and environmentally friendly energy conversion technique, photocatalytic technology has a wide range of potential uses. Because of their suitable energy band structure and exceptional light-absorbing capacity, CdS materials exhibit significant promise in photocatalysis. However, challenges like the high rate of photogenerated carrier complexes and photocorrosion limit the practical application of CdS materials. Building composite heterostructures can help address the drawbacks of CdS materials. Of them, NiFe-LDH is the ideal material to enhance CdS's photocatalytic activity due to its layered structure, high stability, and large number of active sites. This review highlights the great potential of the CdS/NiFe-LDH heterostructure in the photocatalytic field by reviewing the fundamental properties of CdS and NiFe-LDH, as well as the preparation process, photocatalytic mechanism, and applications in the fields of carbon dioxide reduction, organic pollutant degradation, and photocatalytic hydrogen precipitation. The future development direction of this heterostructure and the difficulties (material stability, preparation cost, etc.) in practical application are finally discussed in this work. Future studies should focus on improving the properties of the substance and creating more effective preparation techniques in order to support its broad application in clean energy and environmental preservation and to lay the foundation for the industrial use of CdS/NiFe-LDH heterostructures.

Keywords: CdS, NiFe-LDH, Heterostructure, Photocatalysis

1. Introduction

Global environmental pollution and the energy shortage have grown worse since the 21st century owing to the fast growth of industry and population. These issues pose a serious threat to human survival and raise the standard for the development of new green energy technologies [1, 2]. It is anticipated that solar energy will replace conventional fossil fuels as a sustainable and renewable energy source [3-5]. Since Fujishima and Honda's initial successful breakdown of aqueous hydrogen using TiO₂ under light circumstances in 1972 [6], semiconductor photocatalytic technology has been continuously developed. Recent years have seen a lot of interest in solar photocatalytic technology based on semiconductor materials, which has been used in the fields of photocatalytic hydrogen precipitation, organic pollutant degradation, and carbon dioxide reduction. This technology offers the benefits of low energy consumption, high efficiency, and clean energy [7, 8]. These applications not only address the issue of the scarcity of resources, but they also advance environmental preservation to some extent. The limited utilization of visible light sources, low stability, and high electron-hole

complexation rate [9] are some of the practical limitations of photocatalytic technology that still restrict its use and promotion. Finding novel semiconductor materials with great stability and efficiency is therefore crucial. By building composite structures and combining the distinct qualities of each component, researchers have greatly increased the photocatalytic effectiveness in recent years. This has allowed them to efficiently separate photogenerated electrons and holes and lower the compounding rate. The main issues and future directions of this technology are outlined and discussed in this review, which also looks at the development of CdS/NiFe-LDH heterostructures in photocatalysis. The narrow bandgap semiconductor material CdS is combined with the layered double hydroxide material NiFe-LDH to form a heterostructure.

2. Structure and properties of CdS and NiFe-LDH

2.1. CdS

One common semiconductor material of groups II-VI is CdS, which typically has two crystal structures: cubic sphalerite and hexagonal fibrillar zincite. In this instance, a stable structure is formed by the tetrahedral coordination of each Cd atom with four neighboring S atoms. The stability of the two crystal forms varies with temperature due to their differing atomic configurations. For instance, it has been demonstrated that the cubic sphalerite structure is more stable at high temperatures, whereas the hexagonal sphalerite structure is most stable at ambient temperature (Figure 1). It is possible to vary CdS's stability, specific surface area, and light absorption capacity by altering its crystal structure and morphological characteristics. These changes result in distinct optical and electrical characteristics in photocatalytic reactions [10].

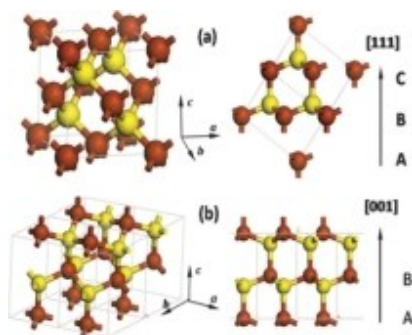


Figure 1: Schematic diagrams of zinc-blende and wurtzite crystal structures of CdS [10]

CdS quantum dots were produced by Jayanta Barman et al. [11] using the chemical deposition approach in a polyvinyl alcohol matrix, and the particle size was regulated by varying the pH. As the particle size shrank, it was found that band gap defect states dominated the luminescence, indicating that the surface defects significantly impacted the optical characteristics. Using a hydrothermal process, Zhao et al. [12] created CdS nanowires, which were discovered to have a high specific surface area and porosity as well as good photocatalytic hydrogen production capabilities when exposed to visible light. By loading CoMnO₄ nanoparticles onto CdS nanosheets using a liquid-phase technique, Bao [13] created two-dimensional CdS nanosheets and CoMnO₄/CdS heterojunctions. It was discovered that the 2% CoMnO₄/CdS composites had a photocatalytic hydrogen generation rate of 3138 $\mu\text{mol}\cdot\text{g}^{-1}\cdot\text{h}^{-1}$, which was 5.8 times greater than that of pure CdS. Through the creation of heterojunctions to increase the separation efficiency of photogenerated carriers and, therefore, boost the photocatalytic activity, the work above highlights the notable benefits of CdS structures with varying dimensions in photocatalytic performance.

CdS's low prohibited bandwidth, strong light absorption coefficient, and appropriate energy band structure are the primary causes of its broad concern [14]. When exposed to visible light, CdS valence band electrons are stimulated to the conduction band and produce holes in the valence band, creating electron-hole pairs that have the redox potential to support photocatalytic processes [15, 16]. However, unmodified CdS's use in photocatalysis is restricted due to issues including photocorrosive effects and high photogenerated carrier complexation rates [17,18]. Researchers built heterostructures, added co-catalysts, and used various tactics to increase the photocatalytic activity of CdS in order to address these issues [19-27].

2.2. NiFe-LDH

With the chemical formula $[M_{1-x}^{2+}M_x^{3+}(\text{OH})_2]^{x+} [A_{x/n}^{n-} m\text{H}_2\text{O}]$, layered double hydroxide (LDH) is a hydromagnesite layered material. The interlayer anion is A_n^{n-} , and the divalent and trivalent metal cations are represented by M^{2+} and M^{3+} , respectively.

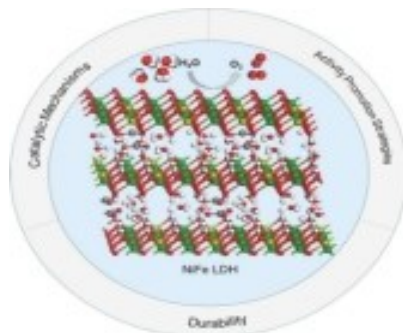


Figure 2: Schematic illustration of NiFe-LDH structure [28]

Because of its regular layered structure, strong ion exchange capacity, and tunability (Figure 2), the LDH material has a promising future in photocatalysis [29-33]. Because of its high activity and inexpensive cost, NiFe-LDH has drawn a lot of attention recently among the many LDH materials [34-36]. Positively charged metal hydroxide layers and interlayer anions make up the crystal structure of NiFe-LDH. These elements mix through electrostatic contact to create a stable layered structure [28]. NiFe-LDH's layered structure not only gives it a lot of active sites and a big specific surface area [37], but it also makes it simple to control the interlayer anions and layer spacing, which enhances its physicochemical characteristics [38,39]. Furthermore, NiFe-LDH has special electrical characteristics due to the synergistic interaction between Ni and Fe. The two elements, Fe^{3+} and Ni^{2+} , respectively, increase the material's redox capacity and facilitate the separation of photogenerated electron-hole pairs.

Numerous studies have tried to use NiFe-LDH as co-catalysts or co-catalysts because of the aforementioned qualities. The latter speeds up the photocatalytic reaction by trapping photogenerated electrons through the Ni and Fe active sites on its own surface, while the former uses the layered structure and numerous surface-active sites to combine with other photocatalytic materials to form heterojunctions [40-44], which motivate the separation of photogenerated electron-hole pairs.

3. Strategies for the construction of CdS/NiFe-LDH heterostructures

3.1. Hydrothermal and solvothermal preparations

The most common and economical techniques for creating CdS/NiFe-LDH composites are hydrothermal and solvothermal. The term "hydrothermal method" denotes the process of creating

materials by a chemical reaction at high temperatures and high pressures in an aqueous solution. The solvothermal technique, which is typically used to regulate the material's size and shape, entails a comparable reaction in an organic solvent. CdS/NiCoAl-LDH composites were made by Yang et al. [45] using a hydrothermal process. Pure CdS/NiCoAl-LDH composites were obtained by dispersing CdS in distilled water, adding $\text{CoSO}_4 \cdot 7\text{H}_2\text{O}$, $\text{NiSO}_4 \cdot 6\text{H}_2\text{O}$, and $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$, adjusting the reaction conditions using urea and NH_4F , and then heat-treating the mixture for 24 h at 120°C . The CdS/MgAl-LDH heterostructure was created by Gao et al. [46] using a solvothermal technique. To create the CdS/LDH composites, the MgAl-LDH nanosheets were first produced hydrothermally, then reacted with cadmium nitrate and thiourea in ethylenediamine. The mixture was then maintained at 150°C for 24h. The benefits of both of these methods include the ability to grow nanomaterials in mild environments and the capacity to optimize the materials' structure and characteristics by varying reaction temperature, time, solvent concentration, and precursor concentration. However, there are disadvantages, including the need for expensive equipment and lengthy reaction times.

3.2. Preparation by electrochemical deposition method

High efficiency and controllability are two advantages associated with using electrochemical deposition, a technique that deposits materials on the electrode surface through an electrochemical reaction. In order to create ZnO/CdS/NiFe-LDH heterostructures, Liu et al. [47] employed electrochemical deposition to cultivate NiFe-LDH nanosheets on ZnO/CdS nanowires. The hydrothermal approach was used for producing ZnO. The SILAR technique was used to deposit CdS nanoparticles onto ZnO, and finally, electrodeposition in an electrolyte containing $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ and $\text{NiSO}_4 \cdot 6\text{H}_2\text{O}$ was used to prepare NiFe-LDH nanosheets. The experimental findings demonstrated the high charge separation efficiency and photocatalytic activity of the ZnO/CdS/NiFe-LDH heterostructures made by electrochemical deposition. Using electrochemical deposition, Pirkarami et al. [48] created CdS on NiCo-LDH/NF electrodes. In order to make NiCo-LDH nanosheets, the NF surface was first pretreated using the chemical oxidation method. Next, the NF surface was electrodeposited in an electrolyte containing $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ and $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ electrodeposition solution. Lastly, CdS was deposited in a solution containing $\text{Cd}(\text{CH}_3\text{COO})_2 \cdot 2\text{H}_2\text{O}$ and thiourea to create a CdS/NiCo-LDH heterostructure. The findings demonstrate the heterostructure's strong photocatalytic activity and charge separation efficiency. By adjusting the deposition potential and time, the morphology and thickness of the material can be precisely adjusted to maximize the photocatalytic performance of the heterostructures. However, excessive process parameters suggest that the equipment requirements are high, time-consuming, and challenging to produce on a large scale, which restricts the method's widespread use.

3.3. Other preparation methods

In situ growth is another efficient technique to produce CdS/NiFe-LDH heterostructures, in addition to hydrothermal and electrochemical deposition methods. CdS/NiFe-LDH heterostructures were developed by Zhou et al. [49] using the in situ growing technique. First, the surface of NiFe-LDH was sprayed with $\text{Cd}(\text{NO}_3)_2$ solution. Next, the surface was sprayed with Na_2S , which caused Cd^{2+} to react with S^{2-} on the surface of NiFe-LDH to produce CdS nanoparticles. The findings show a considerable increase in the hydrogen output of the CdS/NiFe-LDH heterostructure when exposed to visible light. The technique offers the benefits of closely regulating CdS loading and distribution, guaranteeing intimate contact between CdS and NiFe-LDH, and successfully preventing interfacial contamination during the post-treatment phase.

The ultrasonic process is also frequently employed in material preparation. By using the liquid-phase pulsed laser ablation (PLAL) method to ablate Ni foils in aqueous FeCl_3 solution, Lee et

al. [50] created NiFe-LDH nanosheets. These were then incorporated into CdS nanorods and combined using an ultrasonic technique to create CdS/NiFe-LDH nanocomposites. The experimental findings showed that the NiFe-LDH nanostructures produced using the PLAL method were firmly bonded to the CdS nanorods' edges. This close contact at the two interfaces greatly accelerated the rate of photocatalytic hydrogen production and efficiently facilitated carrier transport.

4. Photocatalytic mechanism of Cds/Nife-Ldh heterostructure

4.1. Interface charge transfer mechanism

When CdS is photoexcited in the CdS/NiFe-LDH heterostructure, the holes stay in the valence band while the photogenerated electrons move from the valence band to the conduction band (Figure 3). Because NiFe-LDH has a more negative conduction band position than CdS, photogenerated electrons will move quickly from the CdS conduction band to the NiFe-LDH conduction band. At the same time, the holes stay in the CdS valence band. This effectively separates photogenerated electron holes and lowers their complexation rate, extending the lifetimes of the photogenerated carriers. Furthermore, the catalytic activity and electron transport are further enhanced by the redox of $\text{Ni}^{3+}/\text{Ni}^{2+}$ and $\text{Fe}^{3+}/\text{Fe}^{2+}$ in NiFe-LDH [51].

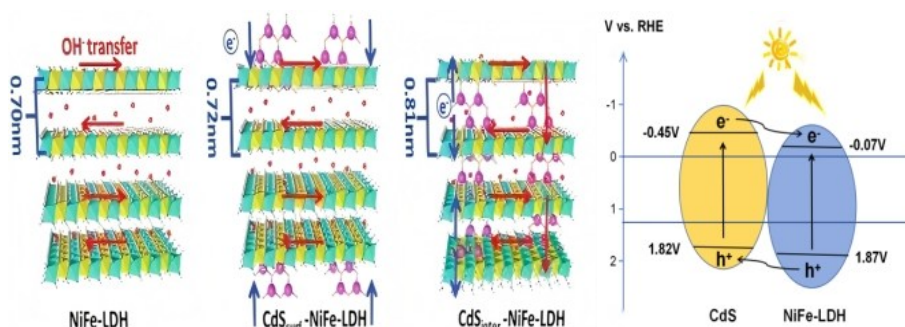


Figure 3: (a) Schematic illustration of the CdS/NiFe-LDH heterojunction structure [52]; (b) schematic band structure diagram of the CdS/NiFe-LDH heterostructure

The photocurrent response test allows one to examine how the heterostructure promotes the photogenerated carrier separation efficiency. The photocurrent densities of the CdS/NiFe-LDH composites are substantially higher than those of the pure CdS and NiFe-LDH, as illustrated in Fig. 4(a). This suggests that the heterostructure formation effectively suppresses the electron-hole complexation while promoting the generation and migration of photogenerated carriers. Furthermore, as illustrated in Fig. 4(b), the transient photocurrent test demonstrates that the photocurrent density of CdS/NiFe-LDH falls more slowly upon light cessation, further demonstrating the extension of the lives of photogenerated carriers and the decrease in recombination rate. The findings above clarified the interfacial charge transfer mechanism of heterostructure and amply validated its function in fostering charge separation [49].

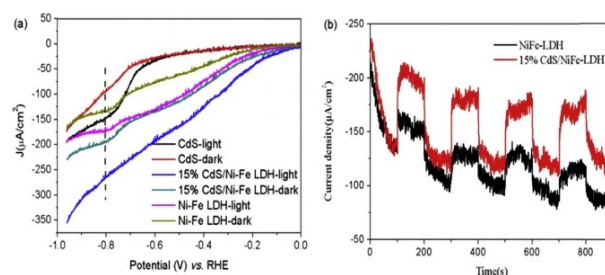


Figure 4: (a) LSV plots of CdS/NiFe-LDH and their typical composites in dark and under light irradiation; (b) the on-off J-t photocurrent curves under light irradiation (on) and in dark (off) [49]

4.2. Energy band structure modulation

The CdS/NiFe-LDH heterostructure has the following energy band arrangement: a typical n-type heterojunction is formed when the valence band position of CdS is below that of NiFe-LDH, and the conduction band position of CdS is higher than that of NiFe-LDH. The heterostructure has good photoresponsive properties and improves the light-absorbing ability in the visible and even near-infrared ranges when exposed to light because it efficiently prevents the compounding of photogenerated electron-hole pairs, increases the carrier lifetime, and simultaneously broadens the light-absorbing range [32, 53].

The absorption intensity of the CdS/NiFe-LDH composite in the visible region is higher than that of the single component, which efficiently uses the solar spectrum, according to the UV-vis diffuse reflectance spectroscopy test, as illustrated in Fig. 5(a). Blueshift phenomena developed when the bulk of CdS on LDH increased, further demonstrating the heterostructure's improved photoresponsive qualities. Additionally, the bandgap of the composite is between that of CdS (2.4 eV) and NiFe-LDH (1.8 eV), and it progressively approaches that of CdS as the proportion of CdS increases, based on an analysis of the bandgap of Fig. 5(b) [49].

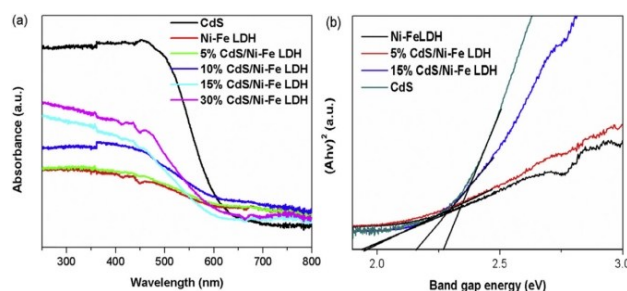


Figure 5: (a) UV-Vis DRS spectra of CdS/NiFe-LDH and its typical composites; (b) the plot of $(A \cdot hv)^2$ versus energy ($h\nu$) for typical samples [49]

4.3. Synergistic effect

Excellent photocatalytic performance and stability are demonstrated by the CdS/NiFe-LDH composite, which combines the characteristics of CdS and NiFe-LDH. In particular, NiFe-LDH offers operational charge transfer channels that allow the quick transfer of photogenerated electrons to the conduction band, thereby drastically lowering the electron-hole pair complexation rate. In contrast, CdS has outstanding light absorption capacity and efficient optical properties in the visible range. Furthermore, NiFe-LDH's layered structure offers a large number of active sites, which improves overall performance while preserving chemical stability.

As illustrated in Fig. 6(a), the photocatalytic hydrogen production performance test demonstrated that the CdS/NiFe-LDH heterostructure's hydrogen production performance was significantly higher

than that of the CdS and NiFe-LDH materials alone. As seen in Fig. 6(b) [49], cycling stability studies demonstrated that the 15% CdS/NiFe-LDH composite had good stability and sustained strong photocatalytic activity after multiple cycles. Through FETEM analysis, Li et al. [50] discovered that the heterostructure did not experience any discernible structural changes during the catalytic reaction. This further confirmed the heterostructure's structural stability by showing the establishment of a strong interfacial between CdS and NiFe-LDH contact.

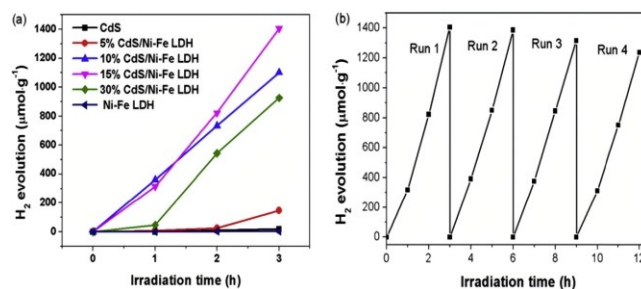


Figure 6: (a) Photocatalytic hydrogen evolution activity of CdS/NiFe-LDH and its representative composites; (b) cycle runs for the photocatalytic hydrogen production over 15% CdS/NiFe-LDH sample [49]

5. CdS/NiFe-Ldh heterostructures for photocatalysis applications

5.1. Photocatalytic hydrogen precipitation

As a clean, sustainable, and renewable resource, hydrogen energy has garnered a lot of interest [54-57]. Direct conversion of solar energy into hydrogen energy is possible with photocatalytic hydrogen precipitation technology, which offers a wide range of potential uses [58]. However, the actual uses of single CdS photocatalysts have been restricted due to issues including high photogenerated carrier complexation rate and severe photocorrosion. Building a heterostructure by composite with NiFe-LDH, which has the benefits of a large specific surface area, strong stability, and low cost, may effectively optimize its performance and increase photocatalytic activity. This makes it an ideal photocatalytic material.

CdS/NiFe-LDH heterostructures were used in photocatalytic hydrogen precipitation by Zhou et al. [49]. In the results, the composite's photocatalytic hydrogen precipitation rate was significantly higher than that of CdS and NiFe-LDH alone, reaching $469 \mu\text{mol g}^{-1} \text{ h}^{-1}$ at 15wt% CdS loading. According to the Mott-Schottky curves, the valence band positions of both CdS and NiFe-LDH are more positive than the water oxidation potential. In contrast, the conduction band positions are more or less negative than the water reduction potentials. This suggests that the composites may have the capacity to precipitate hydrogen photocatalytically. Furthermore, the electron microscopy pictures demonstrated that the CdS nanoparticles were evenly distributed across the NiFe-LDH nanosheet surface, which improved the material's photostability in addition to its photocatalytic activity.

Using a hydrothermal process, Yang et al. [45] synthesized CdS/NiCoAl-LDH heterostructures and examined their potential use in photocatalytic hydrogen precipitation. According to the findings, the 50-CNCAL composite produced hydrogen at a rate of $2116.1 \mu\text{mol g}^{-1} \text{ h}^{-1}$ when exposed to visible light, which was 21.8 and 9.3 times greater than that of pure NiCoAl-LDH and CdS, respectively. Furthermore, the 50-CNCAL composite demonstrated good stability after four-cycle testing, retaining 94% of its photocatalytic activity. The substantial synergistic impact of CdS and NiCoAl-LDH, which increases light absorption and interfacial charge transfer efficiency, is primarily responsible for the increased performance.

5.2. Degradation of organic pollutants

The environment and human health are increasingly at risk from organic contaminants as industrialization picks up speed. Because of its environmentally friendly and sustainable features, photocatalytic technology has emerged as an effective strategy for breaking down organic contaminants [59]. However, the short lifetime and high photogenerated carrier complex rate are issues with conventional photocatalytic technology. Building CdS/NiFe-LDH heterostructures can effectively decrease energy loss and improve photocatalytic performance. The visible light-absorbing capacity of CdS and the benefit of NiFe-LDH's lamellar structure, which can effectively and steadily break down organic contaminants, can be combined in CdS/NiFe-LDH heterostructures.

The CdS/MgAl-LDH heterostructure was used by Gao et al. [46] to degrade tetracycline (TC) and hexavalent chromium (Cr(VI)) in concert. According to the experimental findings, CdS/MgAl-LDH eliminated 92.7% of Cr(VI) in 60 min and completely degraded TC in 40 mins. The heterogeneous structure, which facilitates the efficient separation of photogenerated electrons and holes and raises the efficiency of the oxidation and reduction reactions, is responsible for this effective degrading performance.

5.3. Carbon dioxide reduction

The ability of photocatalytic CO₂ reduction technology to transform greenhouse gases into highly valuable compounds is significant for both the environment and energy [60-62]. Because of its narrow band gap and superior absorption of visible light, CdS has been utilized extensively for CO₂ reduction; nevertheless, its efficiency is limited by its high rate of photogenerated carrier complexation and severe photo corrosion. The efficiency of photocatalytic CO₂ reduction is greatly increased by creating a heterostructure in complex with NiFe-LDH, which can efficiently encourage the separation of photogenerated charges and supply a large number of active sites through the layered structure of NiFe-LDH.

By using a solvothermal technique, Zhang et al. [63] created NiAl-LDH/CdS heterostructures, which they then used for the photocatalytic reduction of CO₂. The findings demonstrated that, when exposed to visible light, the heterostructures could effectively convert CO₂ to methanol and methane. The photocatalytic performances of the heterostructures were 3.3 and 1.6 times greater than those of pristine NiAl-LDH and CdS, respectively.

6. Conclusion

The CdS/NiFe-LDH heterostructure and its advancements in photocatalysis research were reviewed in this paper. Its fundamental characteristics, construction method, photocatalytic mechanism, and uses in carbon dioxide reduction, organic pollutant degradation, and photocatalytic hydrogen precipitation were also covered. Through efficient charge separation and band modulation, the CdS/NiFe-LDH heterostructure has greatly increased photocatalytic efficiency. The layered structure and numerous active sites of NiFe-LDH work in concert with the narrow band gap and high optical absorption coefficient of CdS to make this heterostructure useful for photocatalytic hydrogen precipitation, organic pollutant degradation, and carbon dioxide reduction. The excellent performance of this heterostructure in photocatalytic hydrogen precipitation, organic pollutant degradation, and carbon dioxide reduction is the result of the synergistic effect of the layered structure and abundant active sites of NiFe-LDH with the narrow band gap and high light absorption coefficient of CdS. This suggests new ideas and paths for the problems of environmental pollution and energy shortage. Despite their considerable potential in photocatalysis, CdS/NiFe-LDH heterostructures still confront numerous obstacles. First, there is still a photo corrosion issue with CdS, and the material's long-term durability is inadequate, particularly in situations with extended

exposure. Second, it is challenging to achieve large-scale production due to the material's high preparation costs, lengthy reaction time, and expensive equipment requirements. Future studies can be enhanced in the following ways: First, by doping stabilizing elements or adding the right number of flaws, the materials' photocatalytic activity and stability can be further improved. For example, additional metal elements are added to NiFe-LDH to strengthen its antioxidant reduction ability. In contrast, transition metal elements are doped in CdS to increase their resistance to photo corrosion [64]. Second, for large-scale preparation, inexpensive and ecologically favorable synthesis pathways are created [8]. To cut down on reaction time and energy usage, for instance, microwave-assisted synthesis or ultrasound-assisted synthesis can be investigated. To satisfy the demands of a sustainable and green energy supply as well as environmental enhancement, future research must consistently offer fresh perspectives to establish the groundwork for the practical use of CdS/NiFe-LDH heterostructures.

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