



SPECIAL TOPIC: Advanced Photocatalytic Materials

Nanostructured CdS for efficient photocatalytic H₂ evolution: A review

Rongchen Shen^{1†}, Doudou Ren^{1†}, Yingna Ding², Yatong Guan², Yun Hau Ng^{3*}, Peng Zhang^{4*} and Xin Li^{1*}

ABSTRACT Cadmium sulfide (CdS)-based photocatalysts have attracted extensive attention owing to their strong visible light absorption, suitable band energy levels, and excellent electronic charge transportation properties. This review focuses on the recent progress related to the design, modification, and construction of CdS-based photocatalysts with excellent photocatalytic H₂ evolution performances. First, the basic concepts and mechanisms of photocatalytic H₂ evolution are briefly introduced. Thereafter, the fundamental properties, important advancements, and bottlenecks of CdS in photocatalytic H₂ generation are presented in detail to provide an overview of the potential of this material. Subsequently, various modification strategies adopted for CdS-based photocatalysts to yield solar H₂ are discussed, among which the effective approaches aim at generating more charge carriers, promoting efficient charge separation, boosting interfacial charge transfer, accelerating charge utilization, and suppressing charge-induced self-photocorrosion. The critical factors governing the performance of the photocatalyst and the feasibility of each modification strategy toward shaping future research directions are comprehensively discussed with examples. Finally, the prospects and challenges encountered in developing nanostructured CdS and CdS-based nanocomposites in photocatalytic H₂ evolution are presented.

Keywords: solar fuel, nanostructured cadmium sulfide-based photocatalysts, modification strategies, hydrogen production, photocharge utilization

INTRODUCTION

Rapid urbanization and industrialization have led to a shortage of global energy resources, while significant amounts of harmful and toxic chemical pollutants are being discharged into the environment [1–5]. This necessitates research on environment-friendly technologies to solve the aforementioned environmental and energy crises. Tremendous efforts have been devoted toward developing technologies for producing clean and sustainable energy [6–9]. Among the available renewable energy options, solar energy conversion into chemical fuels has been extensively studied in recent decades [10–14]. Solar energy can become the greenest and most abundant energy source on earth if efficient harvesting and conversion are enabled [15–20]. Among the proposed technologies, visible-light-activated semiconductor photocatalytic technology has been considered one of the most promising strategies for simultaneously overcoming the challenges of environmental pollution and global energy shortage [21–26]. Visible-light semiconductor photocatalytic technologies can convert solar energy into valuable chemical fuels, such as clean H₂ and renewable hydrocarbon fuel, from water splitting, as well as from the photoreduction of CO₂ [27–36]. Concurrently, photocatalytic technology can be used for environmental purification *via* photocatalytic degradation of various harmful chemical pollutants [37–51]. The increasing public awareness of the impacts of global warming and energy shortages has led to efforts that promote research

¹ Key Laboratory of Energy Plants Resource and Utilization, Ministry of Agriculture and Rural Affairs, College of Forestry and Landscape Architecture, South China Agricultural University, Guangzhou 510642, China

² College of Materials and Energy, South China Agricultural University, Guangzhou 510642, China

³ School of Energy and Environment, City University of Hong Kong, Hong Kong SAR, China

⁴ State Center for International Cooperation on Designer Low-Carbon & Environmental Materials (CDLCEM), School of Materials Science and Engineering, Zhengzhou University, Zhengzhou 450001, China

† These two authors contributed equally to this work.

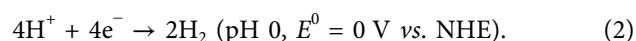
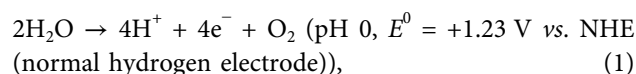
* Corresponding authors (emails: Xinliscau@yahoo.com (Li X); yunhau.ng@cityu.edu.hk (Ng Y); zhangp@zzu.edu.cn (Zhang P))

on new sustainable energy sources [52–55]. Hydrogen (H_2)—a clean energy carrier—has attracted widespread attention as an alternative to fossil fuel to reduce the current environmental pollution and energy crisis [45,56–61]. Carbon-free H_2 can either be used in a typical internal combustion engine to power vehicles or to generate electricity through fuel cells. Presently, H_2 is mainly acquired through the steam reforming of methane using natural gas, which relies on the use of nonrenewable fossil products [62–64].

Since Fujishima and Honda [65] first discovered the photoelectrochemical splitting of H_2O using a Pt/ TiO_2 electrode in 1972, solar H_2 generation *via* artificial photosynthesis has rapidly emerged as a promising method to convert and store solar energy in the form of carbon-free H_2 . Over the past few decades, photocatalytic water splitting for H_2 evolution has been studied in several systems, including organic–inorganic hybrid, molecular material-based homogeneous, and semiconductor-based heterogeneous systems [19,66–70]. The field of solar H_2 is rapidly expanding and involves multidisciplinary approaches, including the following branches: (1) understanding the fundamental photocatalytic mechanisms and constructing efficient photocatalytic systems [71–76]; (2) developing heterogeneous photocatalytic and homogeneous molecular-photosensitizer systems [19,77–79]; and (3) designing novel semiconductor photocatalysts with unique structures and properties [80–82]. The synergy among all these disciplines plays a key role in improving the efficiencies of photocatalytic systems.

The overall photocatalytic H_2O splitting reaction ($2H_2O \rightarrow 2H_2 + O_2$) requires a positive Gibbs energy change of $237.13 \text{ kJ mol}^{-1}$, corresponding to a minimum energy of 1.23 eV [83]. The basic processes of overall photocatalytic

water splitting include three steps (Fig. 1a): (1) light absorption, (2) charge excitation from the valence band (VB) to the conduction band (CB) of the semiconductors to form electron–hole (e^-h^+) pairs, and (3) migration of photoexcited charge carriers (e^-h^+ pairs) to surface active sites to react with H_2O to generate O_2 and H_2 [10,84–86]. During step 3, e^-h^+ recombination readily occurs, resulting in poor photocatalytic activity for overall water splitting. Notably, the CB, VB, and bandgap energies play crucial roles in influencing the photocatalytic performance of semiconductor photocatalysts [11,87]. The standard reduction potentials for the half-reactions of O_2 and H_2 evolution are described in Equations (1) and (2), respectively.



Over the past few years, numerous semiconductor photocatalysts have been developed for photocatalytic H_2 evolution water splitting, such as metal-free compounds (g- C_3N_4 [11,88–94], SiC [78,95,96], and 6,13-pentacenequinone [97]), inorganic solid solutions ($Zn_xCd_{1-x}S$ [98–103], $ZnIn_2S_4$ [104–108], $Bi_{1-x}In_xTaO_4$ [109], $HNb_xTa_{1-x}WO_6$ [59], and $Mn_xCd_{1-x}S$ [110,111]), oxynitrides ($BaTaO_2N$ [112], $LaTaON_2$, and $TaON$ [113]), sulfides (ZnS [114–117], $NiCo_2S_4$ [118], Cu_2ZnSnS_4 [119], MoS_2 [120–123], $CuInS_2$ [124–127], and CdS [128–130]), selenides (WSe_2 [131], $CoSe_2$ [132], $CdSe$ [133–135]), and metal oxides ($SrTiO_3$ [136–138], Ga_2O_3 [139], Fe_2O_3 [140–142], Cu_2O [143,144], ZrO_2 [145], TiO_2 [146–149], ZnO [150–152], Ta_2O_5 [153,154], and CoO_x [155]). Among them, CdS is considered one of the most promising photocatalysts for H_2 production due to its ex-

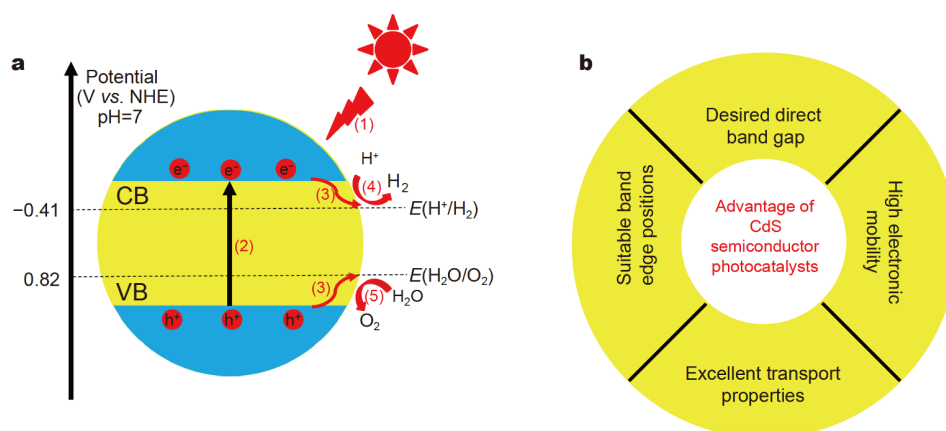


Figure 1 (a) Schematic of the main processes for photocatalytic H_2O splitting on a semiconductor photocatalyst that typically include: (1) light absorption, (2) charge excitations, (3) charge transfer, (4) H_2 evolution, and (5) O_2 evolution. (b) Advantages of CdS semiconductor photocatalysts.

cellent photoresponse in visible light, which is empowered by its suitable bandgap, high stability, as well as low material and synthesis costs [156–158]. A few review papers have reported the progress in the use of inorganic cocatalysts, construction of heterojunctions, and charge transfer behaviors in heterogeneous photocatalytic H₂ production systems [159,160]. In recent years, some review papers have also discussed different classes of semiconductor photocatalysts, including g-C₃N₄ [12] and TiO₂ [161,162], for photocatalytic H₂ production. However, there are only limited comprehensive reviews on the development of photocatalytic H₂ generation systems using CdS-based photocatalysts.

CdS—a classical and vital II–VI photo-semiconductor with a bandgap of 2.4 eV—has been extensively investigated as a promising photocatalyst owing to its several fundamental advantages, including excellent charge transport properties, ideal direct bandgap, high electronic mobility, and suitable band edge positions [17,163,164] (Fig. 1b). The CdS surface can be conveniently functionalized with a variety of ligands, which not only render the CdS-based photocatalysts hydrophobicity or hydrophilicity, but also offer selectivity for interaction with other relevant functional materials [165]. Furthermore,

CdS evinces efficient photocatalytic performances under visible light (wavelength <520 nm). It also has excellent charge carrier transportation capacities, which can efficiently facilitate the migration of photoexcited h⁺ and e⁻ in a timely manner, thereby prolonging the lifetime of the photoexcited carriers and resulting in higher photocatalytic activity [166].

It is evident from Fig. 2a that research on H₂ has rapidly expanded in recent years due to its attractiveness and sustainability. Notably, CdS has recently emerged as a hot research topic owing to its application in nanoscale optoelectronic devices and the potential merits of its known fundamental physical properties (Fig. 2b). The development of CdS photocatalysts has been a significant component of energy and environmental research over the past few years. In 1981, Kalyanasundaram *et al.* [167] reported the production of photocatalytic H₂ and oxygen (O₂) in stoichiometric proportions from overall water splitting in an aqueous suspension of CdS loaded with Pt and RuO₂ under visible light. In 2007, one-dimensional (1D) CdS nanowires (NWs) synthesized *via* a solvothermal method in ethylenediamine were found to exhibit a higher rate of photocatalytic H₂ production in the mixed aqueous solution of Na₂S and Na₂SO₃ as sacrificial re-

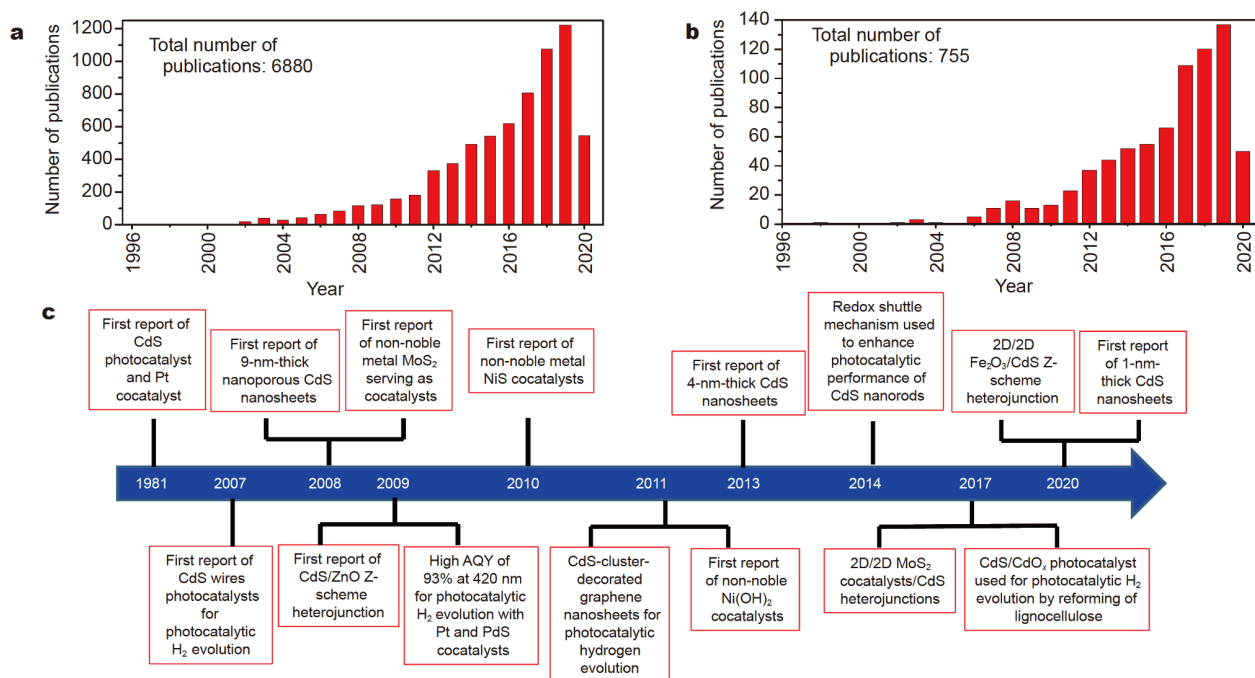


Figure 2 (a) Number of publications since 1996 on photocatalytic H₂-production cocatalysts involving the use of “photo* or light*” and “hydrogen* or H₂ or H-2”. (b) Number of publications since 1996 on photocatalytic CdS-based H₂-production cocatalysts involving the use of “CdS* or Cadmium sulfide*” and “hydrogen* or H₂ or H-2”. (Reorganized from ISI Web of Science Core Collection, date of information search: May 16, 2020). (c) The roadmap of the development of CdS-based photocatalysts for H₂ generation.

agents [168]. Meanwhile, CdS nanosheets with engineered thicknesses have been fabricated by different methods [169–172]. Subsequently, various noble-metal or non-noble-metal cocatalysts, such as Pt–PdS [173], MoS₂ [166], NiS [174], Ni(OH)₂ [175] and Ni₃C [176], have been applied in photocatalytic H₂ evolution over CdS-based photocatalysts. Additionally, various CdS-based composite photocatalysts, such as ZnO/CdS [177], CdS/WO₃ [178], CdS/ α -Fe₂O₃ Z-scheme heterojunction [179], CdS-cluster/graphene [180] and two-dimensional (2D) layered hybrid CdS nanosheets/MoS₂ heterojunctions [66], have also been reported for enhanced photocatalytic H₂ evolution. Despite the significant development, there are only a handful of reviews on CdS-based semiconductor photocatalysts [17,181]. Therefore, a comprehensive review on the strategies to promote CdS-based semiconductors' photocatalytic performances is essential for further development of solar H₂.

An excellent CdS-based photocatalyst must be inexpensive, efficient, visible-light-driven, stable with fast kinetics, and highly efficient in photocatalytic H₂ evolution (Fig. 3). To achieve these goals, a great deal of effort has been devoted toward enhancing stability, reducing cost, improving charge separation and light absorption, as well as accelerating H₂ evolution kinetics. Herein, we mainly concentrate on the manipulation of the charge carriers of nanostructured CdS for efficient photocatalytic H₂ evolution (Fig. 4). This includes strategies for generating more charge carriers, promoting efficient charge separation, boosting interfacial charge transfer, accelerating charge utilization, and suppressing charge-induced self-photocorrosion. All these modification strategies are thoroughly discussed in this review. Exploring the important progress in this project may facilitate a new opportunity toward designing CdS-based nanostructured semiconductor photocatalysts for efficient photocatalytic H₂ evolution.

GENERATING MORE AVAILABLE CHARGE CARRIERS IN NANOSTRUCTURED CdS

Sensitization with dyes and plasmonic metals

As mentioned above, the application of nanostructured CdS for photocatalytic H₂ generation has been attracting considerable attention [182,183]. It is generally accepted that the ideal water-splitting bandgap for semiconductor photocatalysts is ~2.0 eV, wherein an adequate portion of visible light can be harvested to generate sufficient e⁻ and h⁺ with thermodynamic driving forces for photocatalytic

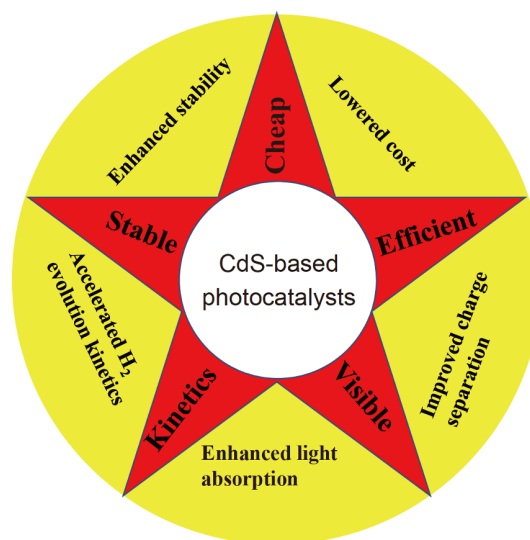


Figure 3 Factors governing the photocatalytic H₂ evolution efficiency of CdS-based photocatalysts and the corresponding engineering and modification strategies.

redox reactions. However, the bandgap of CdS is 2.4 eV, which ensures the sole utilization of solar light with a wavelength shorter than 520 nm. Various methods have been developed to increase the visible-light absorption of CdS-based photocatalysts to generate more available charge carriers. Sensitized semiconductor photocatalysts have been widely used in photoelectrochemistry. Dye sensitization is also a promising strategy to promote photocatalytic H₂ production over CdS-based semiconductor materials, whereby the dye molecules normally do not exhibit photocatalytic H₂ production activities [184,185]. The effectiveness of this step is determined by choosing a suitable dye, light source, and e⁻ donor. Dye photosensitization can extend the visible-light absorption range of CdS-based photocatalysts, thereby improving the photon-harvesting efficiency. With increased light absorption, an extra population of excited e⁻ from the dye molecules that may speed up charge transfer is presented, resulting in highly efficient photoelectric conversion [186,187]. Through this method, the photoexcited e⁻ from the dye molecules with a suitable lowest unoccupied molecular orbital level can pass into the CB of CdS-based semiconductors, thereby contributing additional charges for reactions. Compared with hybridized inorganic materials serving as cocatalysts, metal-free organic dyes display a stronger visible-light absorption capacity, which makes them suitable for enhancing photocatalytic H₂ production activities [185]. Based on the sensitization strategy, semiconductor photocatalysts with narrow

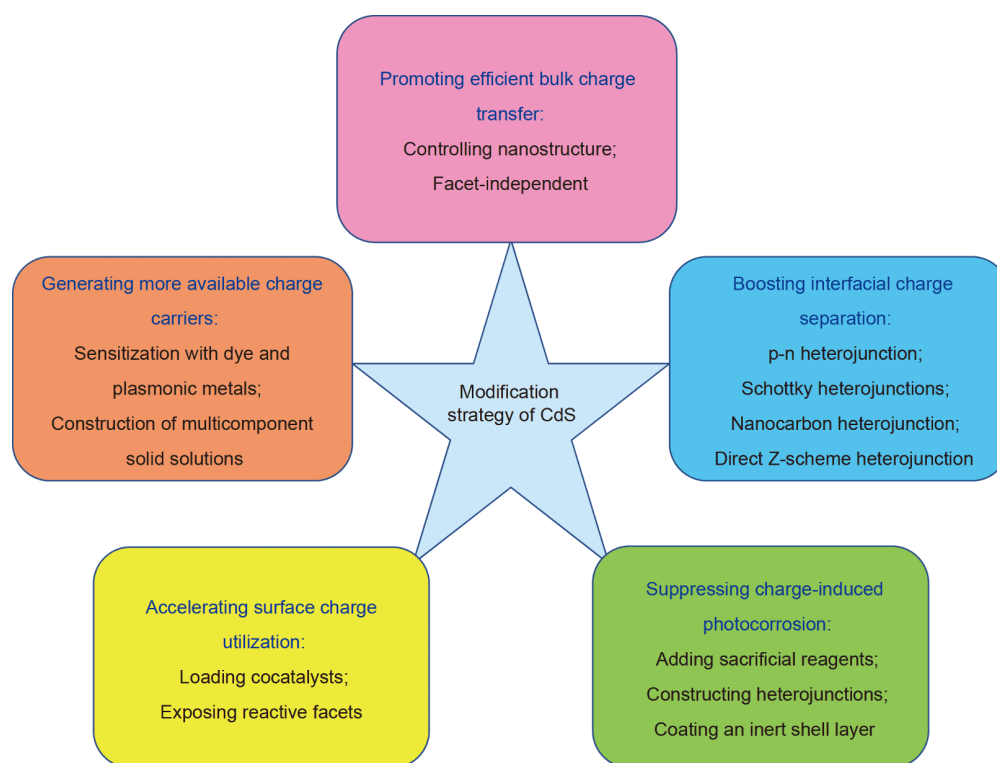


Figure 4 Engineering the charge carriers of nanostructured CdS for efficient photocatalytic H₂ evolution through different modification strategies.

bandgaps, such as graphene quantum dots and conjugated polymers [188,189], have been extensively studied and are considered as good photosensitizer candidates for enhancing the visible-light-driven photocatalysis of CdS-based semiconductors.

Furthermore, coupling CdS photo-semiconductors with other plasmonic metals has been extensively applied in visible-light-driven water splitting activities. The plasmonic energy transfer from the metal to the semiconductor can enhance photocatalytic performances *via* light scattering, near-field enhancement, hot-electron injection, and resonant energy transfer. For example, Ma *et al.* [190] reported multi-interfacial plasmon coupling with CdS for efficient photocatalytic H₂ evolution (Fig. 5a). The (Au/AgAu)@CdS core-shell photocatalysts exhibited excellent photocatalytic performances owing to the strong plasmonic light absorption and near-field enhancement induced by the multi-interfacial plasmon coupling, which could significantly enhance the light-harvesting efficiency from the ultraviolet to the near-infrared region (Fig. 5b and c). Meanwhile, as the number of Au/AgAu gaps increased, light harvesting gradually improved. The four-gap Au/AgAu hybrids displayed the highest extinction intensity and the broadest absorption

band, accompanied by the red-shifting of the localized surface plasmon resonance peaks to 576 nm (Fig. 5d–g). Consequently, the (Au/AgAu)@CdS core-shell photocatalysts exhibited an excellent H₂ evolution performance of 4.71 mmol g⁻¹ h⁻¹, which was 47.2 times higher than that of pure CdS photocatalyst [190]. It is envisaged that the combination of various sensitization strategies can be feasibly designed and applied to enhance the photocatalytic H₂ evolution of CdS-based semiconductors [191]. In the future, all these sensitization strategies are expected to be coupled with CdS-based cocatalysts and thoroughly studied with respect to the mechanisms.

Construction of multicomponent solid solutions

In addition to the extended visible-light absorption, adjusting the CB and VB redox potentials to achieve higher driving forces of the photoexcited charge carriers is another effective strategy. Notably, the fabrication of multicomponent solid-solution CdS-based photocatalysts provides a platform to fully maximize the advantages of the different components to generate more available charge carriers with considerable energetic redox potentials for photocatalytic reactions. Moreover, the bandgap energy (E_g) of multicomponent solid-solution photocatalysts can

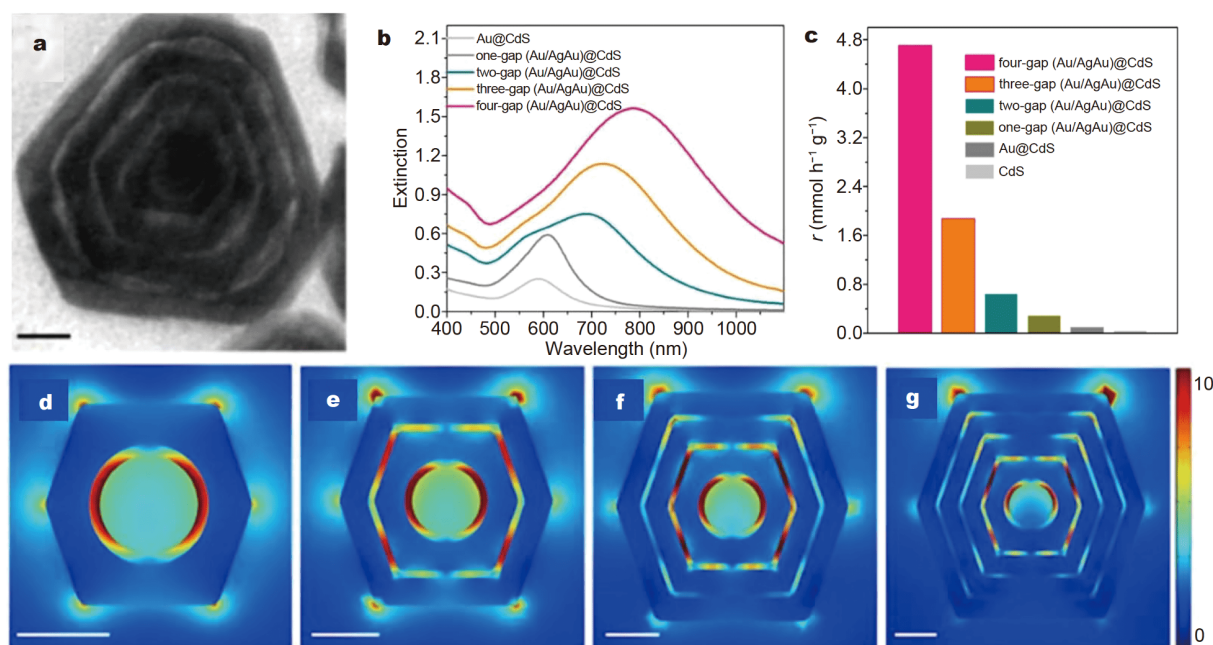


Figure 5 (a) Transmission electron microscopy (TEM) image of (Au/AgAu)@CdS core-shell hybrids. (b) Extinction spectra of Au@CdS and multigap (Au/AgAu)@CdS core-shell hybrids. (c) Photocatalytic H₂ evolution rates of different samples. (d–g) Calculated local electric field distributions of one-, two-, three-, and four-gap Au/AgAu hybrids. Reprinted with permission from Ref. [190]. Copyright 2020, Royal Society of Chemistry.

be continuously tuned by controlling their constituent stoichiometry, which will be rather helpful to achieve an optimized balance between visible-light absorption and redox potential. In recent years, doping transitional metal ions (e.g., Ni²⁺ [192], Cu²⁺ [193], Mn²⁺ [194, 195] and Zn²⁺ [196–200]) with CdS to construct CdS-based solid-solution photocatalysts has proven to be effective for improving photocatalytic water splitting for H₂ evolution. Compared with the original CdS, solid solutions, such as Zn_xCd_{1-x}S and Mn_xCd_{1-x}S, possess some specific advantages, such as tunable band edge positions and bandgap width, more active sites, and better electrical conductivity [201,202]. Additionally, in long-term photocatalytic reactions, the construction of solid-solution photocatalysts can inhibit photocorrosion during visible-light irradiation [195,203,204].

Among the solid solutions, Zn-doped CdS ternary alloys (Zn_xCd_{1-x}S) have received ample attention in the past few years owing to the crystal structure similarity shared by both ZnS and CdS. A well-matched coordination mode between ZnS and CdS can be established [205,206]. However, most reports on Zn_xCd_{1-x}S solid solutions reveal the morphological features of micro-/nano-spheres or irregular particles. In a recent example, Han *et al.* [207] fabricated 1D cubic Cd_{0.8}Zn_{0.2}S solid-solution NWs *via* a facile solvothermal method. Thioglycolic acid (TGA)

served as both the template agent and S source. The TGA revealed a “levelling effect” to nullify the disparity in the physicochemical properties of metal ions *via* its carboxyl and hydrosulfonyl groups, leading to the formation of a cubic Cd_{0.8}Zn_{0.2}S solid solution with a cylindrical morphology. Owing to the doping of Zn into the lattice of the CdS semiconductor, the absorption edges of the Cd_{0.8}Zn_{0.2}S semiconductor are extended to longer wavelengths than those of the CdS semiconductor [208]. In other words, with increasing Zn content in the Zn_xCd_{1-x}S semiconductor, its VB and CB levels shift to more positive and negative positions, respectively. Notably, a higher photo-semiconductor CB level induces a stronger reducing ability, which is pivotal for efficient H₂ generation. Hence, the Cd_{0.8}Zn_{0.2}S solid-solution photo-semiconductor displayed enhanced H₂ generation activity compared with pure CdS (52.3 mmol h⁻¹, with Na₂S–Na₂SO₃ as a sacrificial agent).

Li *et al.* [111] prepared a Mn_xCd_{1-x}S nanorod (NR) solid solution in a pure N₂ atmosphere through standard hot-injection synthesis. By adjusting the ratios of Cd and Mn, the *E_g* values of the Mn_xCd_{1-x}S (*x* = 0–1) solid-solution photocatalysts could be controlled over a wide range (*E_g* = 2.21–3.43 eV). The improved photocatalytic H₂ generation performance of the Mn_xCd_{1-x}S NR solid solution was attributed to the stronger oxidation and

reduction ability, charge separation efficiency, as well as light absorption. In particular, the $\text{Mn}_{0.5}\text{Cd}_{0.5}\text{S}$ NRs exhibited the highest photocatalytic H_2 generation rate of $26 \text{ mmol g}^{-1} \text{ h}^{-1}$ under visible-light irradiation, with an apparent quantum efficiency (AQE) of 30.3% at 400 nm. The photocatalytic performance of the solid-solution $\text{Mn}_x\text{Cd}_{1-x}\text{S}$ NRs was evidently superior to that of the $\text{Mn}_x\text{Cd}_{1-x}\text{S}$ solid solution and pristine CdS fabricated *via* the hydrothermal method [111]. The X-ray diffraction results proved that the sample formed a homogeneous $\text{Mn}_x\text{Cd}_{1-x}\text{S}$ rather than a simple mixture of h-CdS and γ -MnS. The corresponding geometry supercell models for $\text{Mn}_x\text{Cd}_{1-x}\text{S}$ are illustrated in Fig. 6a, while the CB and VB edges of the $\text{Mn}_x\text{Cd}_{1-x}\text{S}$ NR photocatalysts are presented in Fig. 6b. It was apparent that the CB became more negative with increasing Mn content, thereby resulting in a significantly extended bandgap. The improved H_2 generation activity of the $\text{Mn}_{0.5}\text{Cd}_{0.5}\text{S}$ NR solid-solution photocatalysts could be ascribed to a suitable bandgap and an adjusted CB position. In addition, the separation and transfer capacity of photoexcited e^- and h^+ play a key role in photocatalytic H_2 generation. Fig. 6c displays the transient photocurrent response of the $\text{Mn}_{0.5}\text{Cd}_{0.5}\text{S}$ NRs under visible-light irradiation. Under the same conditions, the photocurrent of the $\text{Mn}_{0.5}\text{Cd}_{0.5}\text{S}$ NR solid solution was evidently higher than that of bare CdS and MnS. Unfortunately, the photocatalytic H_2 generation performance of these multicomponent solid solutions

remains insufficient for satisfying the basic requirements for practical applications. Therefore, it is highly desirable to exploit other novel strategies to further enhance the photocatalytic H_2 generation performance of CdS-based solid-solution photocatalysts.

Although both the CB position and bandgap of CdS-based solid-solution semiconductor photocatalysts can be adjusted to a certain extent by tuning the value of x (stoichiometric ratio) to enhance the photocatalytic H_2 generation activity, the rapid recombination of photoexcited charges remains a crucial challenge that limits the further improvement of photocatalytic performances. The construction of CdS-based solid-solution photocatalysts results in a tunable bandgap, wherein these adjustable band potentials may lead to new photoredox reactions (beyond water splitting) and help in the chemical adsorption of specific reactants. Notably, the environmental concern of this type of composite semiconductor photocatalyst can also be assuaged by reducing the Cd content. In addition to constructing bimetallic solid solutions, the photocatalytic H_2 generation performance of CdS-based solid solutions can be further enhanced by intentionally introducing other transition metal ions.

PROMOTING EFFICIENT BULK CHARGE TRANSFER IN NANOSTRUCTURED CdS

In the past few years, efforts have been made to control the morphology, shape, and size of CdS with various

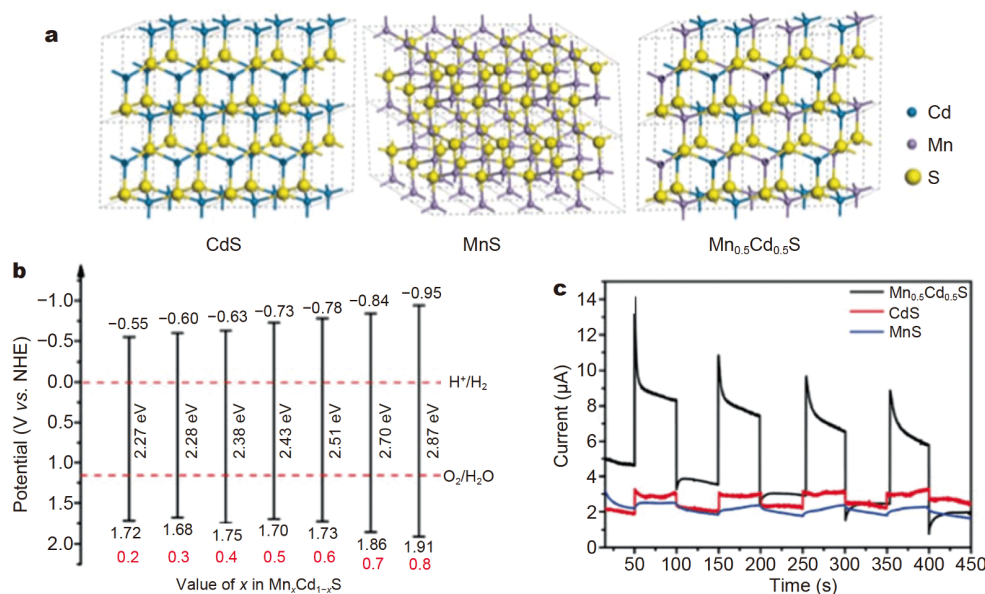


Figure 6 (a) Corresponding geometry supercell models for $\text{Mn}_x\text{Cd}_{1-x}\text{S}$. (b) CB and VB of the $\text{Mn}_x\text{Cd}_{1-x}\text{S}$ NRs solid solutions. (c) $I-t$ curves of $\text{Mn}_{0.5}\text{Cd}_{0.5}\text{S}$, MnS, and CdS. Reprinted with permission from Ref. [111]. Copyright 2018, Elsevier.

nanostructures (NSs). Many studies have reported the photocatalytic performance of CdS semiconductor photocatalysts with unique NSs, such as the 0D, 1D, 2D, and 3D structures. Different NSs impose varying effects on the photocatalytic H₂ generation performance of CdS photocatalysts (Fig. 7). Notably, the photocatalytic H₂ generation performance of photocatalysts with different morphologies, structures, and sizes can be affected by different synthetic methods. Nevertheless, with regard to CdS and CdS-based nanocomposite photocatalysts, researchers have focused on environmentally friendly and low-cost synthetic methods. These approaches include sonochemical, solvothermal, chemical bath deposition, impregnation, template, and template-free methods. Some fabrication methods for CdS photo-semiconductors and CdS-based nanocomposites are listed in Table 1.

Construction of 0D nanostructures

There are many synthetic approaches, such as solvothermal, combustion, sonochemical, biogenic synthesis, complex thermolysis, microwave-assisted polyol synthesis, and chemical precipitation methods, for constructing 0D CdS NSs [73,258–264]. Researchers suggest that although 0D CdS nanomaterials possess large surface area, they are susceptible to severe agglomeration, which reduces the photocatalytic efficiency.

Wang *et al.* [265] fabricated 0D CdS nanoparticles (NPs) modified with covalent triazine-based frameworks (CTF-1) in a controlled manner by means of a facile one-pot solvothermal synthesis. As the triazine unit of CTF-1 has an apparent Lewis property on its nitrogen sites, size-

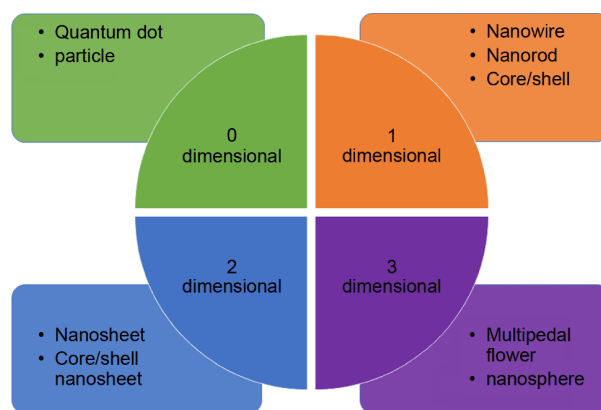


Figure 7 Descriptions of the morphologies of CdS photocatalysts.

controlled and highly dispersed CdS NPs can be prepared and stabilized on CTF-1 layers. The morphology of the as-fabricated 0D CdS NP nanocomposites was investigated *via* scanning electron microscopy (Fig. 8a). The formation processes of the porous CTF-1 frameworks and the subsequent decoration with 0D CdS NPs to form CdS/CTF-1 nanocomposite are presented in Fig. 8b. Compared with bare CdS, the electrochemical impedance spectrum of CdS/5%CTF-1 displays a smaller semicircle, implying that CdS/CTF-1 possesses a reduced charge transfer resistance for photoinduced e⁻ (Fig. 8c). This result was further supported by the photoluminescence (PL) spectra, which were used to study the recombination rate of the photoexcited charge carriers (Fig. 8d). The PL peak intensity of CdS/5%CTF-1 was evidently lower than

Table 1 Synthesis methods for CdS nanostructures

Photocatalyst	Synthesis method	Morphology	Precursor materials	HER (mmol g ⁻¹ h ⁻¹)	AQE (420 nm)	Ref.
CdS–TiO ₂	Hydrothermal	Nanodots	Cd(CH ₃ COO) ₂ ·2H ₂ O, dimethyl sulfoxide	1.5	11.9%	[209]
CdS	Hydrothermal synthesis	Nanoparticles	Graphdiyne, Cd(Ac) ₂ ·2H ₂ O	4.1	-	[210]
CdS	Hydrothermal	Nanoparticles	Cd(CH ₃ COO) ₂ ·2H ₂ O, DMSO	58.9	-	[211]
CdS	Solid-state	Nanoparticles	Cd(Ac) ₂ ·2H ₂ O, thioacetamide	1.15	16.5%	[212]
CdS	Microwave	Nanoparticles	C ₄ H ₁₀ CdO ₄ ·2H ₂ O, H ₂ NCSNH ₂	11.4	-	[213]
CdS	Hydrothermal	Nanoparticles	Cd(NO ₃) ₂ ·4H ₂ O, NH ₂ CH ₂ CH ₂ NH ₂	0.56	-	[214]
CdS	Hydrothermal	Nanoparticles	Cd(CH ₃ COO) ₂ , Na ₂ S	14.2	8.7%	[215]
CdS	Hydrothermal	Nanoparticles	Na ₂ S, Cd(CH ₃ COO) ₂	5.9	8.6%	[57]
CdS	Solvothermal	Nanoparticles	CdCl ₂ ·2.5H ₂ O, thiourea	1.89	-	[216]
CdS	Hydrothermal	Nanoparticles	Cd(NO ₃) ₂ ·4H ₂ O, dimethyl sulfoxide	0.61	-	[217]
CdS	Hydrothermal	Nanoparticles	Cd(NO ₃) ₂ ·4H ₂ O, thiourea	1.26	-	[218]
CdS/CdWO ₄	Hydrothermal	Nanoparticles	CH ₃ CSNH ₂ , Cd(CH ₃ COO) ₂ ·2H ₂ O	9.17	-	[219]

(Continued)

Photocatalyst	Synthesis method	Morphology	Precursor materials	HER (mmol g ⁻¹ h ⁻¹)	AQE (420 nm)	Ref.
CdS	Directly reacting	Nanoparticles	Na ₂ S, Cd(CH ₃ COO) ₂	5.89	19%	[220]
CdS	Hydrothermal	Nanoparticles	Na ₂ S·9H ₂ O, Cd(NO ₃) ₂ ·4H ₂ O	0.33	34.3%	[221]
CdS	Hydrothermal	Nanoparticles	Cd(Ac) ₂ ·2H ₂ O, CH ₄ N ₂ S	2.85	10%	[222]
CdS/Cu ₂ O/g-C ₃ N ₄	Hydrothermal	Nanoparticles	Na ₂ S, CdCl ₂	1.84	-	[223]
CdS-BCNNTs	Hydrothermal	Nanoparticles	Cd(NO ₃) ₂ ·4H ₂ O, Na ₂ S	0.526	4.01	[224]
CdS/ZnS	Solvothermal	Nanorod	CdCl ₂ ·2.5H ₂ O, thiourea	24.1	9.3%	[225]
CdS/TiO ₂	Solvothermal	Nanorod	Cd(NO ₃) ₂ , thiourea	1.118	-	[226]
CdS@MoO _x	Solvothermal photodeposition	Nanorod	Cd(NO ₃) ₂ , thiourea	5.42	-	[227]
CdS/g-C ₃ N ₄	Solvothermal method	Core/shell nanowires	Cd(NO ₃) ₂ ·4H ₂ O, thiourea	4.15	4.3%	[228]
CdS	Solvothermal	Nanowires	CdCl ₂ ·2.5H ₂ O, (C ₂ H ₅) ₂ NCSSNa	0.15	44.9%	[229]
CdS	Self-templated synthesis	Nanoporous structures	CdCl ₂ ·2.5H ₂ O, NaOH, Na ₂ S·9H ₂ O	27.33	60.34%	[169]
CdS	One-pot synthesis	Nanorods	Cd(NO ₃) ₂ ·4H ₂ O, CS(NH ₂) ₂ , H ₂ PtCl ₆	10.29	-	[230]
CdS	Hydrothermal	Nanorods	(CH ₃ CO) ₂ Cd·xH ₂ O, CH ₄ N ₂ S	15.56	-	[231]
CdS	Hydrothermal	Nanorods	Cd(NO ₃) ₂ ·4H ₂ O, thioacetamide	24.15	-	[232]
CdS	Hydrothermal	Nanorods	Cd(NO ₃) ₂ ·4H ₂ O, NH ₂ CSNH ₂	15.55	6.9%	[233]
CdS	Hydrothermal	Nanorods	Cd(NO ₃) ₂ ·4H ₂ O, thiourea	1.131	-	[234]
CdS	Hydrothermal	Nanorods	CdCl ₂ ·2.5H ₂ O, CH ₄ N ₂ S	106	29%	[235]
CdS	Hydrothermal	Nanorods	CdCl ₂ ·2.5H ₂ O, NH ₂ CSNH ₂	4.64	11.8%	[236]
CdS	Hydrothermal	Nanorods	CdO, Na ₂ S	11.58	16.3%	[237]
CdS	Solvothermal	Nanorods	CdCl ₂ ·2.5H ₂ O, thiourea	3.5	-	[155]
CdS/Ti ₃ C ₂	Hydrothermal	Nanorods	Cd(NO ₃) ₂ ·4H ₂ O	2.407	35.6%	[238]
Pt-CdS/g-C ₃ N ₄ -MnO _x	Hydrothermal	Nanorods	Cd(NO ₃) ₂ ·4H ₂ O	924.4	1.745%	[239]
CdS	Hydrothermal	Nanorods	Cd(NO ₃) ₂ , NH ₂ CSNH ₂	1.84	21.2%	[240]
CdS	Hydrothermal	Nanorods	Cd(NO ₃) ₂ , thiourea	167.1	1.5%	[241]
CdS/ZnS	Solvothermal, chemical bath deposition	Nanorod	CdCl ₂ , CH ₄ N ₂ S	239	16.8%	[18]
CdS	Solvothermal,	Nanorod	Cd(Ac) ₂ ·2H ₂ O, CH ₄ N ₂ S	20.2	-	[242]
CdS	Directly reacting	Nanorod	Cd(NO ₃) ₂ , thiourea	37.1	43%	[243]
CdS	Solvothermal	Nanorod	Cd(NO ₃) ₂ , thiourea	0.206	-	[244]
CdS/g-C ₃ N ₄	vapor deposition	Nanotubes	CdCl ₂ ·2.5H ₂ O	0.392	-	[245]
CdS	Hydrothermal	Nanosheets	Cd(Ac) ₂ ·2H ₂ O, sulfocarbamide, ethylenediamine	27.8	14.7%	[246]
CdS/GO	Hydrothermal	Nanosheets	CdCl ₂ ·2.5H ₂ O, DETA	10.5	29.5%	[247]
CdS	Hydrothermal	Nanosheet	CdSO ₄ , DL-dithiothreitol	1.293	-	[248]
CdS	Hydrothermal	Nanosheets	CdCl ₂ ·2.5H ₂ O, S powder	138.7	-	[249]
CdS-FeP	Hydrothermal	Nanosheets	CdCl ₂ ·2.5H ₂ O, S powder	18.63	11.2	[250]
CdS	Hydrothermal	Nanosheets	CdCl ₂ ·2.5H ₂ O, DETA	7.37	-	[251]
MoS ₂ /CdS	Two-step hydrothermal	Core/shell-like	Ammonium molybdate, thiourea	38.75	14.7%	[252]
CdS	Precipitation	Cubic-Phase	Cd(NO ₃) ₂ , Na ₂ S, Na ₂ SO ₃	18	-	[253]
CdS	One-pot wet-chemical	Nanocrystals	CdO	1.98	-	[254]
CdS	Hydrothermal	Nanospheres	Cd(CH ₃ COO) ₂ ·2H ₂ O, thiourea	4.65	7.31%	[255]
CdS	Hydrothermal	Nanospheres	Cd(CH ₃ COO) ₂ , thiourea	44.65	-	[256]
CdS	Hydrothermal	Nanospheres	Cd(Ac) ₂ ·2H ₂ O, thiourea	1.44	-	[257]

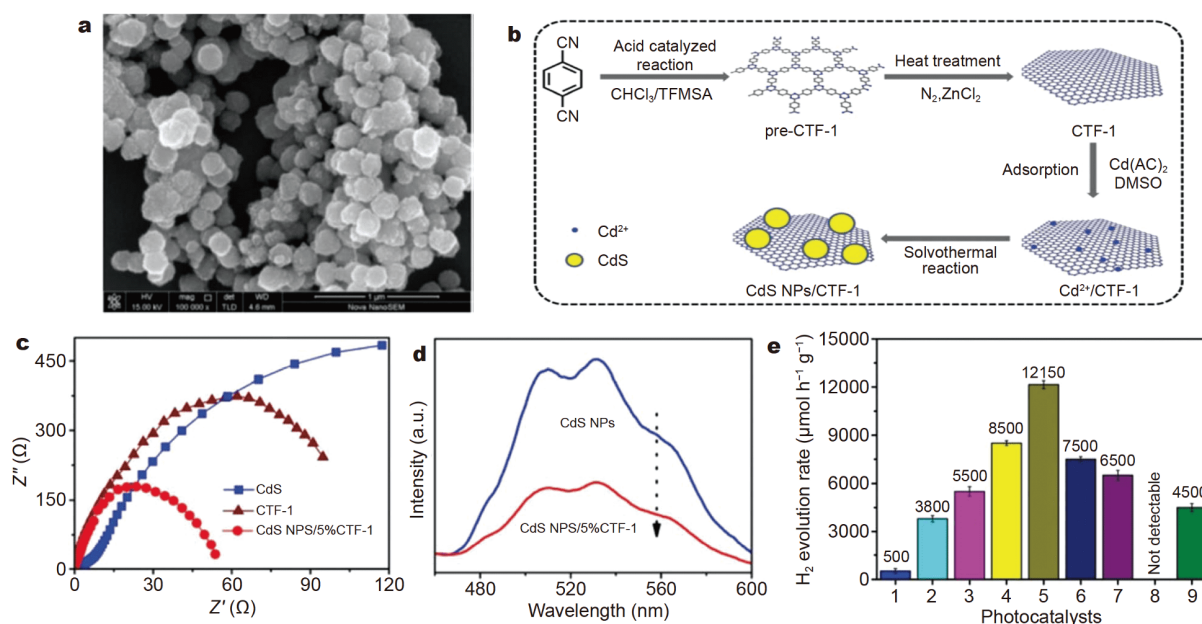


Figure 8 (a) SEM image of CdS, (b) schematic of the formation of 0D CdS NPs/CTF-1, (c) Nyquist plots, (d) PL spectra, and (e) amount of H₂ evolved over CTF-1 (1), CdS (2), CdS NPs/0.5%CTF (3), CdS NPs/1%CTF (4), CdS NPs/3%CTF (5), CdS NPs/5%CTF (6), CdS NPs/10%CTF (7), without catalyst or visible-light irradiation (8), and physically mixed CdS and CTF-1 (9). Reprinted with permission from Ref. [265]. Copyright 2018, Royal Society of Chemistry.

that of the bare 0D CdS NPs, suggesting that the charge recombination rate decreased in the nanocomposites. The photocatalytic H₂ generation performance of the as-fabricated CdS/CTF-1 was investigated using Pt as a cocatalyst in a lactic acid medium under visible-light irradiation. Fig. 8e shows that the H₂ generation rate of CdS/5%CTF-1 was better than those of bare CdS NPs and CTF-1. An insignificant amount of H₂ was released when bare CTF-1 was used as the catalyst. Hydrogen could not be detected without photocatalysts or visible-light irradiation, which suggested that H₂ generation was induced by irradiating CdS/5%CTF-1.

Construction of 1D nanostructures

Generally, 1D CdS NRs with improved charge transport properties compared with the 0D CdS quantum dots displayed quantum constraint effects and directional charge carrier transport. This reduced the recombination losses at the grain boundaries and enhanced the light harvesting capability [266–268]. The visible-light absorption occurred along the longer dimensions (microns or more) of the 1D NSs, while carrier separation through diffusion took place over a short radial distance [269]. The enhanced light absorption, combined with the merits of long distance as well as fast charge transfer, makes the 1D NS a promising candidate for solar energy conversion.

There is increasing attention on the fabrication of 1D photocatalytic NSs, such as NWs, NRs, and their core-shell structures [270–273].

In 1D CdS NRs, the transfer velocity of the carrier is different in the axial and radial directions. Generally, the carrier moves much faster in the radial direction than in the axial one [274]. Thus, it is particularly vital to construct the morphology of CdS nano-semiconductors for 1D NR semiconductors with ideal diameters and high aspect ratios. Several methods, including ion-exchange [163], ligand-assisted growth [275,276], and metal particle-seeded growth [277], have been developed to fabricate 1D rod-shaped semiconductor materials. For example, Wu *et al.* [278] prepared 1D CdS NRs using the seeded growth method and studied the effects of 1D CdS and CdS–Pt NRs with different diameters on the dissociation kinetics and exciton localization. They studied the excitation dynamics and electronic structure of the photocatalytic 1D CdS–Pt NRs. In the presence of Pt, e⁻ were efficiently transferred from CdS to Pt, and photoexcited e⁻-h⁺ pair recombination was suppressed. The tips of 1D CdS NRs can offer selective deposition sides for Pt, wherein the selectively tip-deposited Pt can promote the charge separation of 1D CdS NRs and CdS-based NSs. These results demonstrate that 1D CdS NRs with larger diameters possess more specific properties than tradi-

tional CdS NR structures. Many factors, including reaction time and temperature, as well as reactant and surfactant amounts, influence the synthesis of 1D CdS NSs.

Construction of 2D nanostructures

The photochemical, optical, and photoelectrical properties of the photo-semiconductors can be indirectly influenced by their morphology, size, and structure [279–281]. Since the discovery of 2D photocatalytic nanomaterials, 2D CdS NSs have attracted much attention because of their unique phase characteristics, and excellent photocatalytic properties, larger surface area and lower recombination rate of e^-h^+ pairs [171,246,282,283]. To synthesize 2D CdS photo-semiconductors, it is important to seek a low-cost and environment-friendly process that can control the formation of reasonable sizes and predictable shapes.

Apparently, the decreased thickness of 2D semiconductor nanosheets can effectively reduce the migration distance of photogenerated e^- from the bulk phase to the semiconductor photocatalyst surface, thereby preventing the recombination of photogenerated e^-h^+ pairs inside the semiconductor. For instance, in 2013, Xu *et al.* [170] synthesized ultrathin CdS nanosheets with thickness of ~ 4 nm *via* ultrasonic-induced aqueous exfoliation of lamellar CdS–diethylenetriamine hybrid nanosheets using *L*-cysteine as a stabilizing agent. The as-obtained CdS ultrathin nanosheets achieved H_2 production rates of $41.1 \text{ mmol g}^{-1} \text{ h}^{-1}$ with an AQE of 1.38% at 420 nm. In 2018, Bie *et al.* [171] synthesized the thinnest CdS nanosheets (1.5-nm-thick) through a simple and low-cost oil-bath method using sodium citrate as the structure-directing agent. They exhibited a photocatalytic H_2 production rate of $2.155 \text{ mmol g}^{-1} \text{ h}^{-1}$, which was approximately 3.7 times higher than that of CdS NPs ($0.582 \text{ mmol g}^{-1} \text{ h}^{-1}$). More recently, Xie *et al.* [172] constructed five-layer ultrathin Cd_4S_5 nanosheets with unsaturated surface S anions *via* chemical vapor deposition. The thickness of the CdS nanosheet was approximately 1 nm. The average H_2 evolution rate of the ultrathin nanosheets was $29.44 \text{ mmol g}^{-1} \text{ h}^{-1}$. Evidently, this unique structure induces a higher CB position than that of their thicker counterparts, which can fundamentally improve electrical conductivity, reduce recombination rate, and eventually enhance the driving force for the H_2 evolution reaction (HER). Consequently, ultrathin CdS nanosheets achieve a high AQE of 4.15% under 420-nm light irradiation.

Meanwhile, exposing the reactive facets on the surface of 2D CdS nanosheets is another strategy to boost the

photocatalytic H_2 evolution. For instance, Li *et al.* [284] synthesized flake-like hexagonal phase CdS micro-/nanoleaves with exposed (0001) facets to promote the photocatalytic activity toward H_2 generation (Fig. 9a). The growth process of the dendrites is shown in Fig. 9b. Compared with CdS NPs, the photocatalytic H_2 generation activity of CdS micro-/nanoleaves with NR-branched NSs was significantly enhanced ($468.4 \text{ mmol h}^{-1}$) (Fig. 9c). The average H_2 evolution rate of photocatalytic reactions reached $740.9 \text{ mmol h}^{-1}$, which was more than 6 times that of spheroidal particles under visible-light irradiation. The catalysts also evinced good stability (Fig. 9d). The enhanced activity of the CdS micro-/nanoleaves was partly due to the unique 2D micro-/NS and the exposure of high surface energy facets (0001) [284].

Additionally, constructing a 2D/2D NS coupled with layered composite photocatalysts could further boost charge separation between the CdS nanosheets and the other 2D cocatalysts owing to strongly coupled contact interfaces [179,285,286]. For example, Ma *et al.* [66] successfully synthesized 2D/2D layered hybrid CdS/MoS₂ nanocomposites *via* a one-step hydrothermal method. The coupling of 2D MoS₂ NSs as cocatalysts boosted the photocatalytic H_2 evolution performance of CdS NSs (Fig. 10). It was confirmed that the 2D CdS/1%MoS₂ nanocomposites exhibited the highest photocatalytic H_2 generation activity ($1.75 \text{ mmol g}^{-1} \text{ h}^{-1}$) in an aqueous solution containing sulfite and sulfide under visible light. The loading of ultrathin 2D MoS₂ NSs and the tight 2D/2D coupling interfaces led to excellent H_2 generation performances owing to the effectively boosted separation and migration of charge carriers as well as improved surface H_2 evolution kinetics. In our previous study, we demonstrated that the 2D/2D CdS/Cu₇S₄ nanocomposite could serve as advanced photocatalysts for photocatalytic water splitting toward H_2 generation [246]. The highest achieved H_2 generation rate of the 2D/2D CdS/2%Cu₇S₄ nanocomposites was $27.8 \text{ mmol g}^{-1} \text{ h}^{-1}$, which was approximately 11 times higher than that of pure CdS NSs ($2.6 \text{ mmol g}^{-1} \text{ h}^{-1}$). The construction of the 2D/2D CdS/Cu₇S₄ heterojunction not only promoted photoexcited e^-h^+ pair separation, boosted photoexcited e^- transfer, and prolonged the lifetime of the photoexcited e^- , but also increased the visible-light absorption and H_2 generation kinetics.

Compared with the interfaces of 0D/1D, 0D/2D, 1D/1D, and 1D/2D, the 2D/2D coupled nanocomposites display a larger contact surface, which is conducive to more efficient interfacial charge migration [285]. Therefore, the construction of unique interfaces with tight 2D/

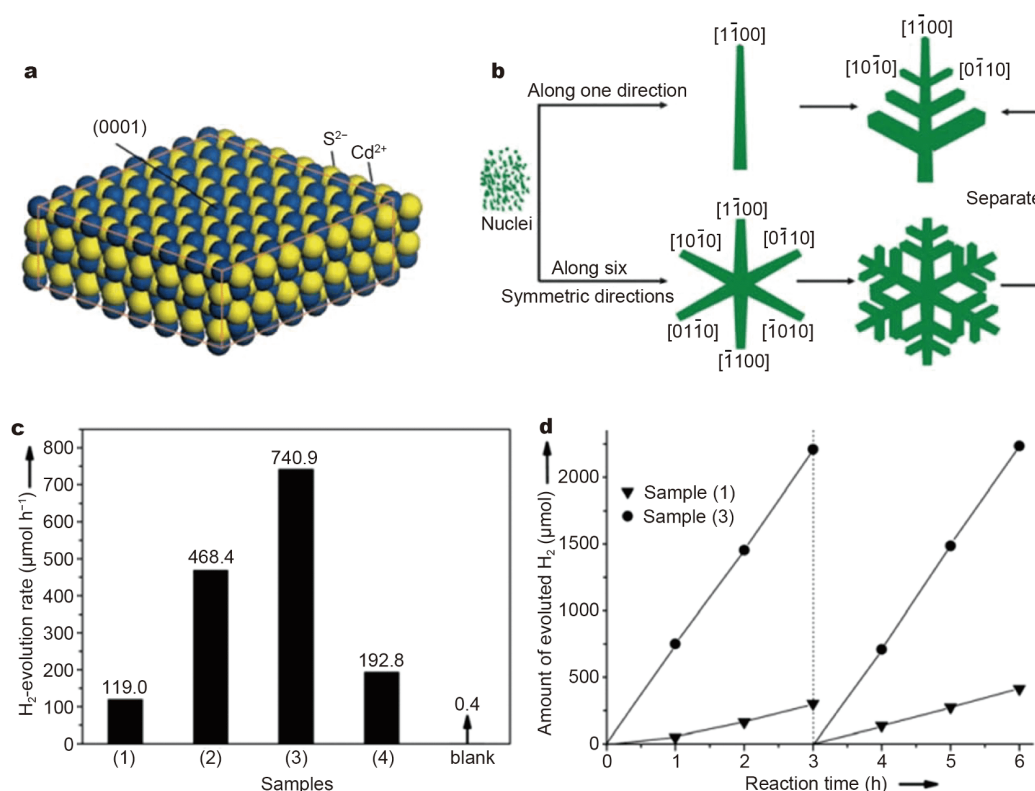


Figure 9 (a) Crystal structure of the hexagonal CdS. (b) Schematic of the growth process of CdS. (c) Average photocatalytic H₂ generation rate of CdS. (d) Time courses of photocatalytic H₂ generation on CdS, (1) without HF, and with (2) 0.125 mol L⁻¹ HF, (3) 0.200 mol L⁻¹ HF, and (4) 0.500 mol L⁻¹ HF. Reprinted with permission from Ref. [284]. Copyright 2012, Royal Society of Chemistry.

2D coupling and large surface areas is critical for achieving efficient charge separation and photocatalytic H₂ generation activity.

Construction of 3D nanostructures

CdS photocatalysts with 3D NSs, such as hierarchical dendritic CdS, porous CdS flowers, and hollow spheres, have been widely studied [284,287–291]. Because of their unique structural characteristics, such as low material density and high surface/volume ratio, constructing 3D CdS NSs has been considered as a useful strategy to boost the photocatalytic activity. For example, to overcome the shortcomings of traditional non-porous CdS/TiO₂ spherical nanocomposites (rapid recombination of e⁻-h⁺ pairs and inferior light absorption), Wu *et al.* [292] designed non-noble metal cocatalysts covered in hollow core-shell NSs for photocatalytic H₂ evolution. The unique hollow structure enhanced the absorption in the visible spectral range, while the non-noble metal cocatalysts provided active sites for H₂ evolution. The coupling of these three components reduced photogenerated e⁻-h⁺ recombination. The synergistic effects of photocatalysts and e⁻-h⁺

separation were preliminarily studied. Chen *et al.* [293] synthesized hollow core-shell CdS/TiO₂/Ni₂P photocatalysts for H₂ production with SiO₂ spheres as sacrificial templates (Fig. 11a and b). The formation of CdS/TiO₂ nanocomposites extended the absorption range of TiO₂ into the visible-light region, while the outer TiO₂ layer protected the CdS core from photocorrosion. The hollow structure improved the transmission capacity of light and reduced the reflectance of visible light. The H₂ generation rate of CdS@TiO₂ (4.65 mmol g⁻¹ h⁻¹) showed significant improvement compared with that of CdS/TiO₂/Ni₂P nanocomposites (13.91 mmol g⁻¹ h⁻¹). The H₂ generation rate for the CdS/TiO₂/Pt nanocomposite was 16.81 mmol g⁻¹ h⁻¹, which was slightly higher than that of CdS/TiO₂/Ni₂P (Fig. 11c and d). The activity of CdS/Ni₂P and CdS/TiO₂/Ni₂P remained rather unchanged after 5 cycles. However, CdS/Pt and CdS/TiO₂/Pt exhibited a diminishing photocatalytic activity after each recovery cycle due to photocorrosion and poor stability of the Pt deposit (Fig. 11e) [293]. In the future, it is expected that more 3D CdS-based NS photocatalysts can be constructed through the self-assembly of promising 2D ultrathin CdS

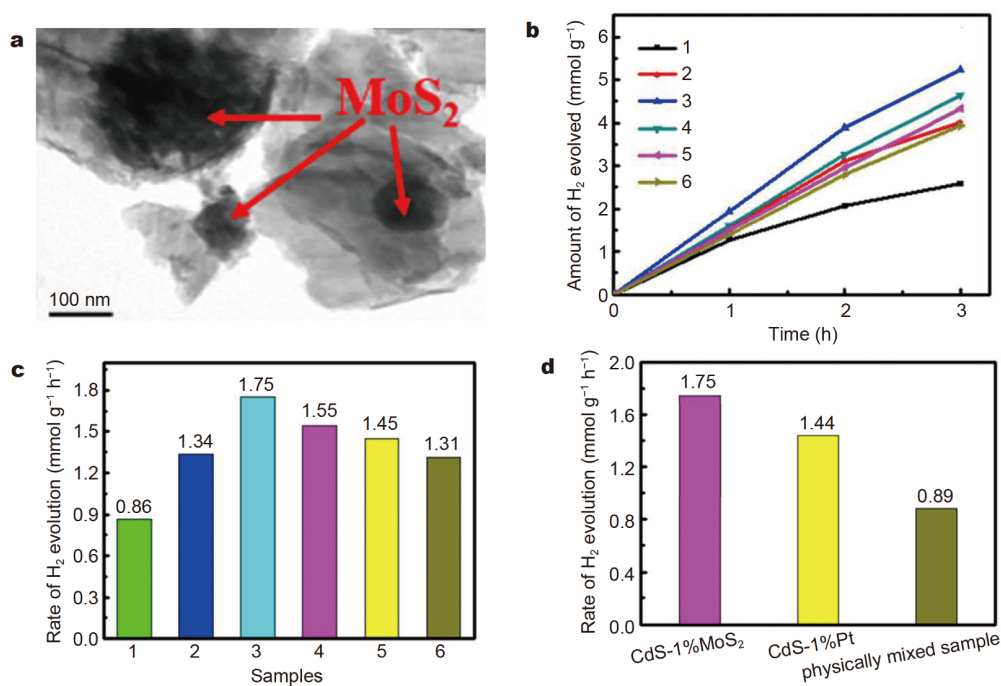


Figure 10 (a) TEM image of CdS/1%MoS₂, (b) time-dependent of H₂ generation (1: CdS NSs, 2: CdS-0.5% MoS₂, 3: CdS-1% MoS₂, 4: CdS-2% MoS₂, 5: CdS-3% MoS₂, 6: CdS-5% MoS₂), (c) the average rate of photocatalytic H₂ generation over the photocatalysts, and (d) comparison of H₂ generation activities of CdS/1%MoS₂, CdS/1%Pt, and physically mixed sample of CdS/1% MoS₂. Reprinted with permission from Ref. [66]. Copyright 2017, Elsevier.

nanosheets, which could maximize all the advantages of 2D nanosheets by circumventing their unfavorable stacking.

BOOSTING INTERFACIAL CHARGE SEPARATION IN NANOSTRUCTURED CdS

Boosting the interfacial charge transfer of CdS-based photocatalysts can also be achieved by constructing semiconductor heterojunctions, which induce an internal electrical field for interfacial charge transfer [10]. Usually, semiconductor heterojunctions are categorized into the Schottky junction, as well as carbon-based, direct Z-scheme, Type I, Type II, and Type III heterojunctions. The Schottky and Type II junctions are beneficial for constructing efficient CdS-based photocatalysts. For instance, heterojunctions between CdS and other semiconductors (such as Fe₂O₃ [179], TiO₂ [63,226,294,295], g-C₃N₄ [296,297], NiWO₄ [298,299], BiOBr [300], WO₃ [165,178,222,301], MoO₂ [302], ZnO [150,303–306], LaFeO₃ [307], SiC [308], Bi₂WO₆ [309], and Co₃O₄ [310]) have been widely used in photocatalytic H₂ evolution.

However, there are still debatable aspects associated with the separation mechanism of photogenerated e⁻-h⁺ pairs in these heterojunctions [311]. In addition, the reduction and oxidation potentials will be compromised

after forming the Type II heterojunction, restricting their activity enhancement. Therefore, various favorable heterojunctions, such as p-n, Z-scheme, Schottky-based, carbon-based, and multicomponent heterojunctions, have also been studied to improve the photocatalytic performance of CdS-based photocatalysts.

p-n heterojunction

The p-n heterojunction has been widely applied toward enhancing the photocatalytic performance of CdS-based photocatalysts. The formation mechanism of the p-n heterojunction is shown in Fig. 12 [311]. Under visible-light illumination, the e⁻ from the n-type semiconductor are transferred to the p-type semiconductor owing to the larger work function of the latter. This results in the formation of a built-in electric field, which is beneficial for the transfer of photogenerated e⁻ between p-type and n-type semiconductors.

Ai *et al.* [224] designed novel CdS/boron carbon nitride nanotubes (BCNNTs) using a new band-matching transformation strategy. In this system, they realized the heterojunction transformation from Type I to Type II by tuning the carbon content of BCNNTs (Fig. 13a). The rational design of the band-matching process played an important role in constructing efficient photocatalysts *via*

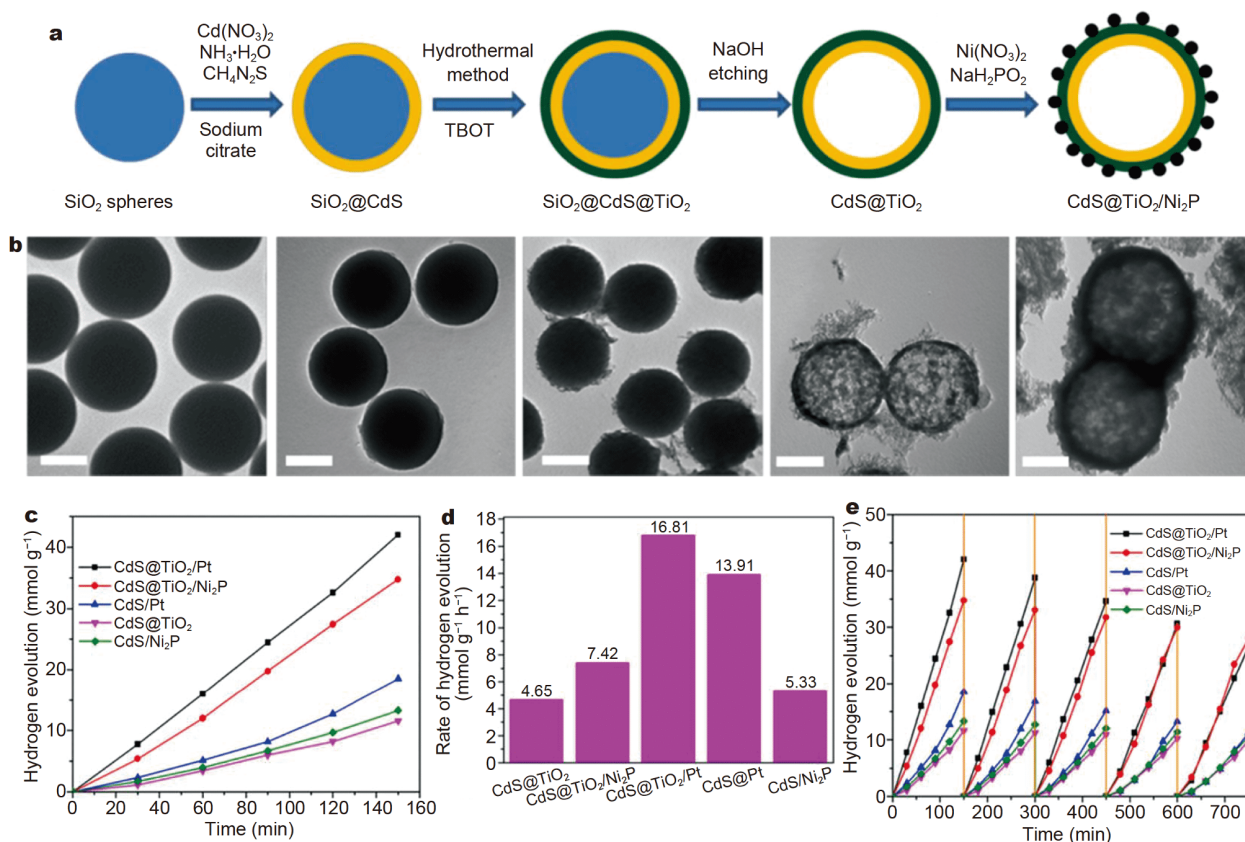


Figure 11 (a) Schematics of CdS@TiO₂/Ni₂P preparation, (b) TEM images of SiO₂, SiO₂/CdS, SiO₂/CdS/TiO₂, CdS/TiO₂, and CdS@TiO₂/Ni₂P, (c) photocatalytic H₂ evolution, (d) rate of H₂ evolution and (e) stability of H₂ evolution. Reprinted with permission from Ref. [292]. Copyright 2019, Elsevier.

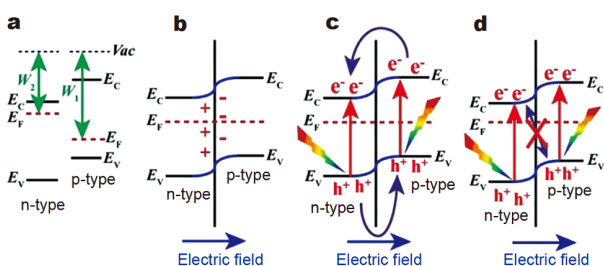


Figure 12 Formation mechanism of the p-n heterojunction: (a) before contact, (b) in contact, (c) photogenerated charge transfer, and (d) unsuccessful direct Z-scheme transfer of photogenerated charge carriers. Reprinted with permission from Ref. [311]. Copyright 2018, Elsevier.

the formation of a strong internal electric field to guarantee rapid charge separation and transfer. As the C content increased, the bandgap of BCNNTs decreased from 2.79 to 2.05 eV. At low C content, the CdS/BCNNTs belong to the Type I heterojunction, and all of the photogenerated e⁻ and h⁺ were transferred to the CB and VB of CdS, respectively. With a high C content, the CB potential of BCNNTs was more positive than that of CdS,

and VB was negative relative to CdS. This composite also belongs to the Type I heterojunction, wherein the CdS photogenerated e⁻ and h⁺ were all transferred to BCNNTs. However, with moderate C-doping, both the CB and VB of BCNNTs were more positive than those of pure CdS, thereby fulfilling the requirement for the formation of Type II heterojunctions (Fig. 13c). The internal electric field between the CdS and BCNNTs could boost the separation of photogenerated e⁻-h⁺ pairs. Hence, the photocatalytic performance of the Type II heterojunctions exceeded that of the Type I heterojunction between CdS and BCNNTs (Fig. 13b) [224]. Zhang *et al.* [310] constructed a ZIF-67(Co)-derived Co₃O₄ framework modified with a CdS p-n heterojunction photocatalyst (Fig. 14a-c). The formation of p-n heterojunctions could effectively reduce the bandgap of the composite, enhance the light absorption intensity, shorten the fluorescence lifetime (2.61 ns), accelerate the electron injection rate ($K_{ET} = 1.17 \times 10^8 \text{ s}^{-1}$), and improve electron injection efficiency ($\eta_{inj} = 30.6\%$) (Fig. 14d-f). The hollow structure of the Co₃O₄ framework served not only as a h⁺ collector

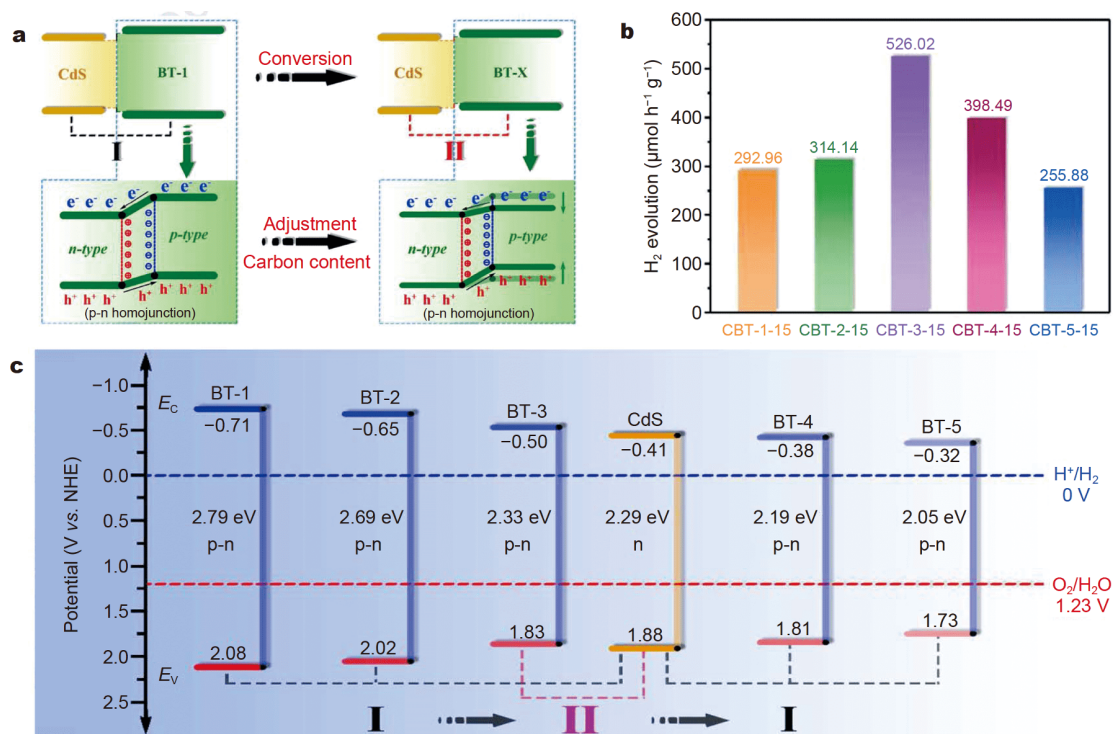


Figure 13 (a) Schematic for band-matching transformation of the CdS/BCNNTs photocatalytic system, (b) average H₂ evolution rate of CdS/BCNNTs, (c) schematic of the band-matching transformation process between BCNNTs ($X=1, 2, 3, 4, 5$) and CdS. Reprinted with permission from Ref. [224]. Copyright 2020, Elsevier.

but also as a supporting material for CdS, thus increasing the specific surface area of the catalyst [310]. The results further confirmed the key roles of the p–n heterojunction toward achieving efficient charge separation and transfer across the heterojunction interface as well as the prolonged lifetime of the charge carriers.

Schottky junctions

Enhancing the photocatalytic activity of CdS-based semiconductors could also be realized by constructing Schottky junction-based internal electrical fields [4,87]. The Schottky junction can boost the spatial separation of e⁻ and h⁺, thus delaying their recombination and lengthening their lifetime. Usually, for metal photo-semiconductor systems, because of the higher work function (Φ_m) of the metal than that of the semiconductor (Φ_s), the photoexcited e⁻ in the photo-semiconductor migrate to the metal and realign their Fermi levels; this leads to the formation of a Schottky barrier with an upward band bending and depletion layer (Fig. 15). As an effective method to trap photogenerated e⁻, Schottky-based heterojunctions can promote e⁻-h⁺ separation in photocatalysis [312].

When photo-semiconductors are coupled with metal, the e⁻ in the CB of the photo-semiconductor approach the metal until the two Fermi energy levels are in equilibrium. A space-charge layer is formed on the surface of the metal, resulting in efficient separation of photoexcited e⁻-h⁺ pairs and migration of the charge carriers. For example, the work functions of metallic Cd clusters and CdS are 4.08 and 5.18 eV, respectively (Fig. 16a and b) [313]. After contact, the e⁻ in Cd migrate to the CdS until the Fermi levels are realigned, triggered by the higher Fermi level in metallic Cd than CdS. When CdS is excited by visible-light irradiation, the built-in field drives the migration of the photoexcited e⁻ on the CdS to the Cd sites. Thus, the construction of a Schottky junction can enhance visible-light-driven water splitting for H₂ generation. Similarly, Shang *et al.* [314] decorated CdS NPs on Cd nanosheets to form Schottky junctions by the polyol reduction method and oxidation-sulfurization process (Fig. 16c and d). The Cd nanosheets that served as a support were evidently better than those of carbon nanotubes and graphene in that study because the work function of Cd is much higher than that of the nano-carbon materials (Fig. 16e). More importantly, the black

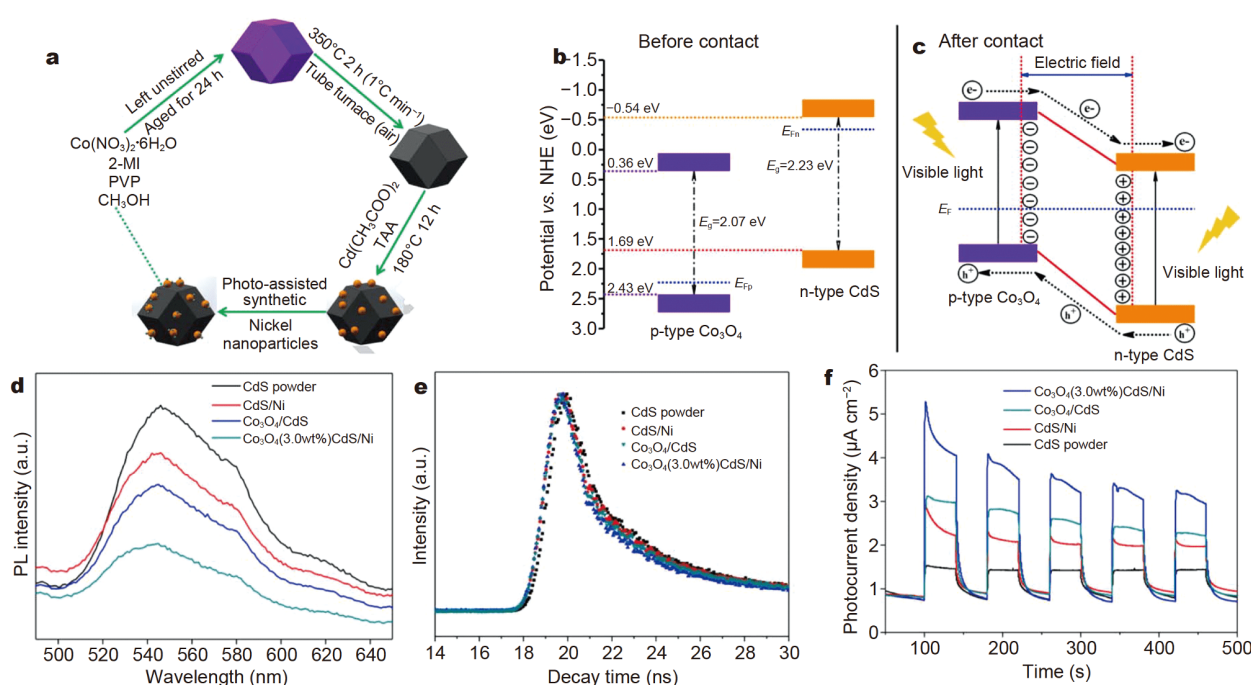


Figure 14 (a) Schematic of the fabrication of $[\text{Co}_3\text{O}_4/\text{CdS}/\text{Ni}]$. Schematics for energy bands of p-CdS and n- Co_3O_4 (b) before contact and (c) after the formation of the p-n heterojunction. (d) PL spectra, (e) time-resolved PL spectra, and (f) photocurrent of the as-prepared photocatalysts. Reprinted with permission from Ref. [310]. Copyright 2018, Elsevier.

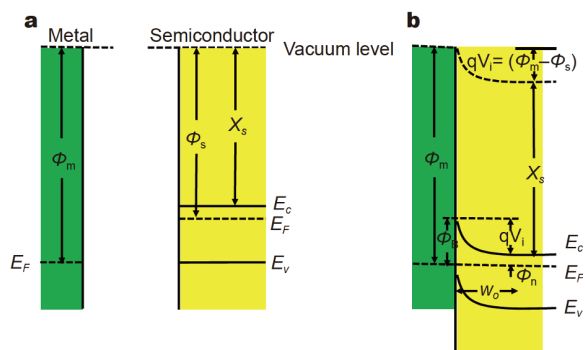


Figure 15 Schematic for the formation of the Schottky barrier ($\Phi_m > \Phi_s$): (a) before contact and (b) thermal equilibrium after contact (E_g , Φ , E_F , and χ represent the bandgap energy, work function, Fermi level, and electron affinity, respectively).

color of the nanocarbon shields a fraction of the visible light from reaching CdS [314]. Xiao *et al.* [238] demonstrated that a 1D CdS NR/2D MXene nanosheet with Schottky junctions reached an excellent photocatalytic H_2 evolution rate of $2407 \mu\text{mol g}^{-1} \text{h}^{-1}$, which was attributed to the high charge carrier mobility of highly conducting metallic Ti_3C_2 and strong interfacial coupling between CdS and Ti_3C_2 [238].

Additionally, it is well known that nanocarbon mate-

rials (e.g., graphene [291,315–319], carbon quantum dots [211,236,257,320,321], carbon black [322], carbon fiber [323–325] and carbon nanotubes [78,326–328]) have been considered as outstanding promoters for photocatalytic performances. The attraction is attributed to their superior adsorption capacity, unique electronic conductivity, nontoxicity, low cost, high stability, and large surface area. The formation of carbon-based Schottky junctions and the narrowing bandgap by carbon doping are both beneficial to photocatalytic reactions.

Direct Z-scheme heterojunction

Biomimetic artificial photosynthesis by constructing direct Z-scheme photocatalysts represents a feasible strategy for improving the photocatalytic performance [46,204,306,329–332]. Specifically, the direct Z-scheme photocatalytic system has a charge carrier transfer pathway similar to the letter “Z” (Fig. 17) [333]. During the photocatalytic reaction, the photoexcited e^- (with lower reduction capacity) in semiconductor A recombine with the photoexcited h^+ (with lower oxidation capacity) in semiconductor B. Therefore, the photoexcited e^- (with high reduction capacity) in semiconductor B and photoexcited h^+ (with high oxidation capacity) in semiconductor A can be utilized. The direct Z-scheme

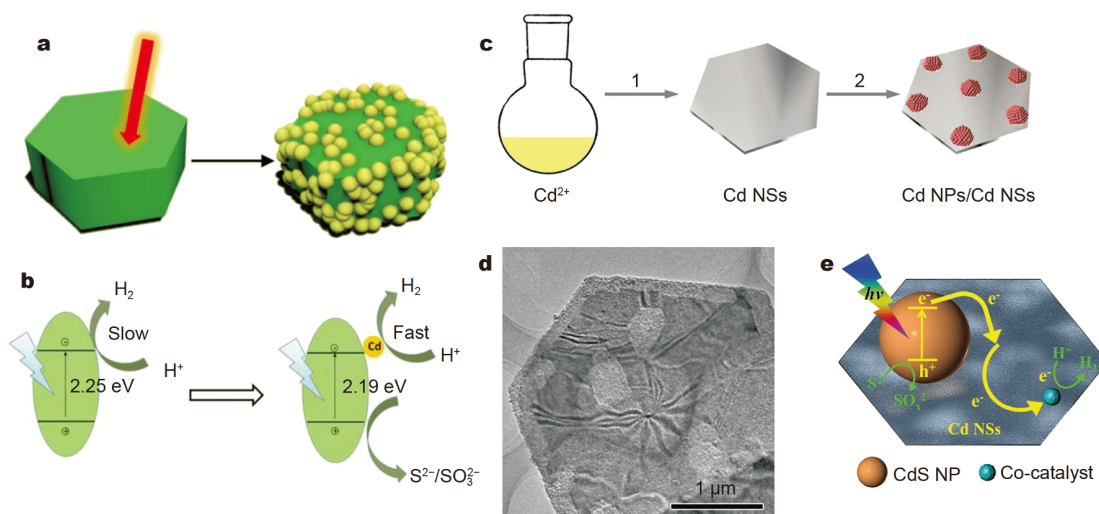


Figure 16 (a) Schematic of the laser irradiation-induced formation of a CdS/Cd Schottky junction, (b) synthesis process [313], (c) schematic of photocatalytic H₂ generation performance, (d) TEM image and (e) charge transfer mechanisms in CdS/Cd photocatalysts. Reprinted with permission from Ref. [314]. Copyright 2016, Wiley-VCH.

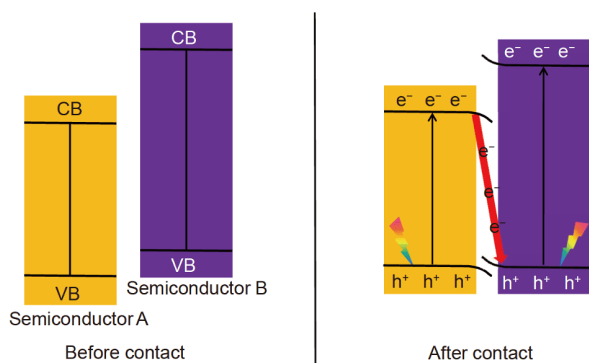


Figure 17 Mechanisms of charge carrier separation in the Z-scheme system.

photocatalyst possesses an enlarged and stronger redox capacity. Furthermore, the charge carrier transport for direct Z-scheme nanocomposites is more physically feasible because the transport of photoexcited e^- from the CB of photo-semiconductor A to the photoexcited h^+ -rich VB of photo-semiconductor B is favorable due to the electrostatic attraction between h^+ and e^- .

In 2009, Cheng's group [177] reported the direct Z-scheme of CdS/ZnO heterostructure photocatalysts. They found that the formation of Z-scheme heterostructures between ZnO and CdS could effectively prolong the lifetime of photogenerated e^- , reaching a 14-fold improvement in H₂ evolution performance compared with that of pure CdS. Since then, numerous direct Z-scheme CdS-based photocatalysts have been applied in photo-

catalytic H₂ generation applications [334], including CdS/WO_{3-x} [222], FeC₂O₄·2H₂O/CdS [335], CdS/WO₃ [165,301], CdS/MoO_{3-x} [336], CdS/g-C₃N₄ [337,338], CoWO₄/CdS [44], CdS/Fe₂O₃ [179], CdS/BiVO₄ [339], TiO₂/CdS [329,340,341], CdS/CdWO₄ [219,342], ZnO/CdS [306] and CdS/PI [217]. In our previous study, we reported the fabrication of 2D/2D CdS/g-C₃N₄ direct Z-scheme heterojunction nanocomposites through the *in-situ* growth of 2D CdS NSs on 2D g-C₃N₄ NSs [338]. As shown in Fig. 18a and b, the direct Z-scheme CdS/g-C₃N₄ nanocomposites displayed improved photocatalytic H₂ generation activity compared with pure g-C₃N₄ and CdS. The highest H₂ generation rate is realizable with the CdS/0.7 g-C₃N₄ direct Z-scheme heterostructures at 15.3 mmol g⁻¹ h⁻¹, which is 3000 and 4 times higher than those of bare g-C₃N₄ and CdS, respectively. After 21 h of continuous visible-light irradiation, the 2D/2D CdS/0.7 g-C₃N₄ maintained excellent photocatalytic H₂ generation activities without noticeable decay (Fig. 18c). The apparent efficiency of the direct Z-scheme CdS/0.7 g-C₃N₄ was 6.86% at 420 nm (Fig. 18d). Based on the calculations and experimental results, a schematic for photocatalytic H₂ generation with CdS/g-C₃N₄ nanocomposite photocatalysts was suggested (Fig. 18e). This work highlights the synergistic effect between the direct Z-scheme and the 2D/2D NS in promoting photocatalytic H₂ generation reactions.

Shen *et al.* [179] reported 2D/2D Z-scheme nanocomposites fabricated *via* the *in-situ* growth of CdS NSs on α -Fe₂O₃ NSs (Fig. 19a). In addition, the modification

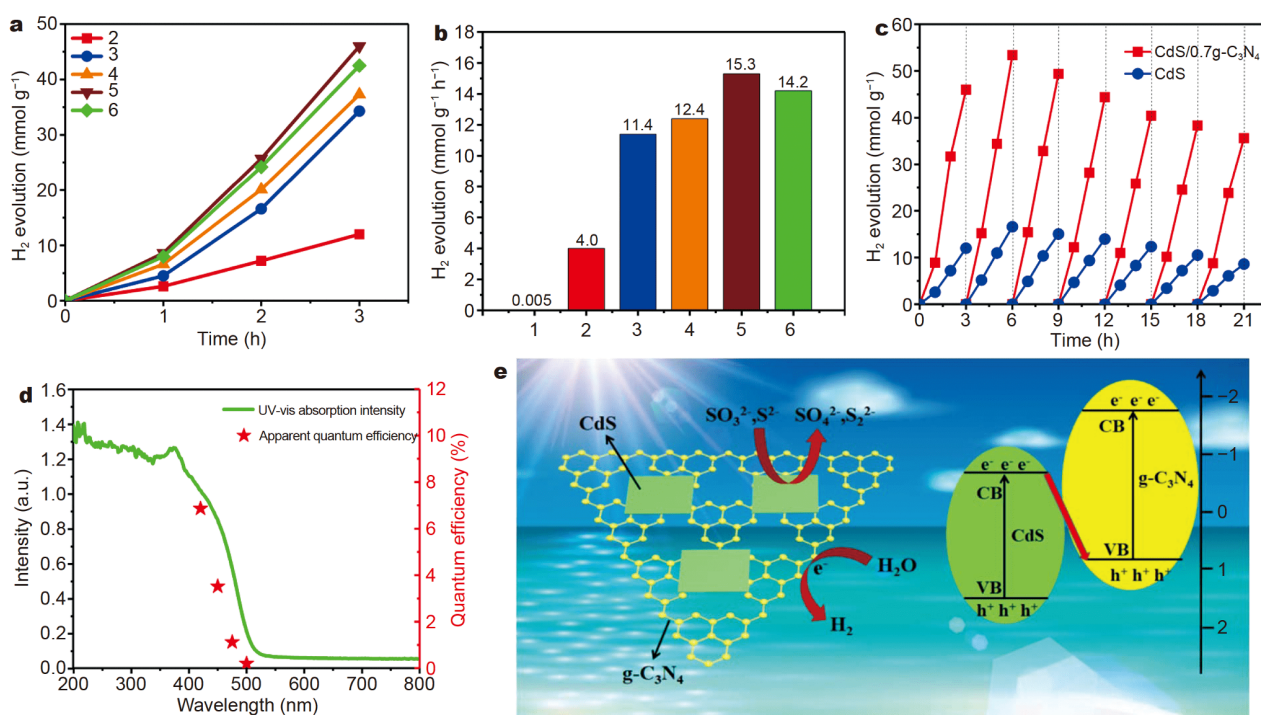


Figure 18 (a) Time-dependent photocatalytic H₂ evolution, (b) average H₂ evolution rates for different samples (1: g-C₃N₄, 2: CdS, 3: CdS/0.5g-C₃N₄, 4: CdS/0.6g-C₃N₄, 5: CdS/0.7g-C₃N₄, 6: CdS/0.8g-C₃N₄), (c) repeated time courses of photocatalytic H₂ evolution, (d) apparent quantum efficiency for CdS/0.7g-C₃N₄, and (e) proposed schematic for 2D/2D CdS/g-C₃N₄ S-scheme heterostructures. Reprinted with permission from Ref. [338]. Copyright 2020, Wiley-VCH.

of CdS/ α -Fe₂O₃ Z-scheme nanocomposites could be performed with metallic β -NiS, which continued to construct an ohmic junction as H₂-generation active sites (Fig. 19b). The CdS/ α -Fe₂O₃/NiS ultrathin 2D/2D heterojunction exhibited an outstanding H₂ generation rate (45 mmol g⁻¹ h⁻¹) with high AQEs at 420 nm (46.9%). The outstanding photocatalytic performance was ascribed to: (1) ohmic-based heterojunction that offered a large number of H₂-evolution sites; (2) large and intimate interfaces, which facilitated charge transfer; and (3) boosted charge migration in the direct Z-scheme heterojunction (Fig. 19c-e).

Besides 2D/2D Z-scheme nanocomposites, those with core-shell structures also offer advantages in photocatalytic water splitting. Compared to 2D/2D structures, core-shell structures can better protect CdS from photocorrosion. Ma *et al.* [304] synthesized CdS@ZnO core-shell structured photocatalysts *via* atomic layer deposition (ALD) (Fig. 20a). The growth of ZnO along certain facets was effectively controlled by the number of ALD cycles (Fig. 20b and c). The formation of core-shell structures in the Z-scheme composite could suppress the recombination of photogenerated e⁻-h⁺ pairs and induce strong and intimate heterojunction interfacial contact between CdS

and ZnO. Upon loading Pt and PdS as cocatalysts on the CdS@ZnO surface, the photocatalytic H₂ evolution rate reached 98.82 mmol g⁻¹ h⁻¹ with an AQE of 69.59% at 420 nm (Fig. 20d-f) [304].

These results highlight the promising performance of the 2D/2D layered Z-scheme heterojunctions with larger contact area for fast separation of photoexcited charge carriers across their interfaces with respect to the 0D/2D and 1D/2D coupling systems. Notably, coupling semiconductors at their active facets are apparently more promising for efficient photogenerated e⁻ transfer. It is expected that CdS-based composites with 2D/2D layered Z-scheme heterojunctions coupled with their active facets can be rationally fabricated for photocatalytic H₂ evolution [179,338].

ACCELERATING SURFACE CHARGE UTILIZATION OF NANOSTRUCTURED CdS

Structural defects in CdS may result in fast recombination of photo-excited e⁻-h⁺ pairs [257,343]. As photocatalytic H₂ generation from water is a thermodynamically unfavorable reaction, the sluggish kinetics on the CdS surface need to be addressed. It is common to use suitable

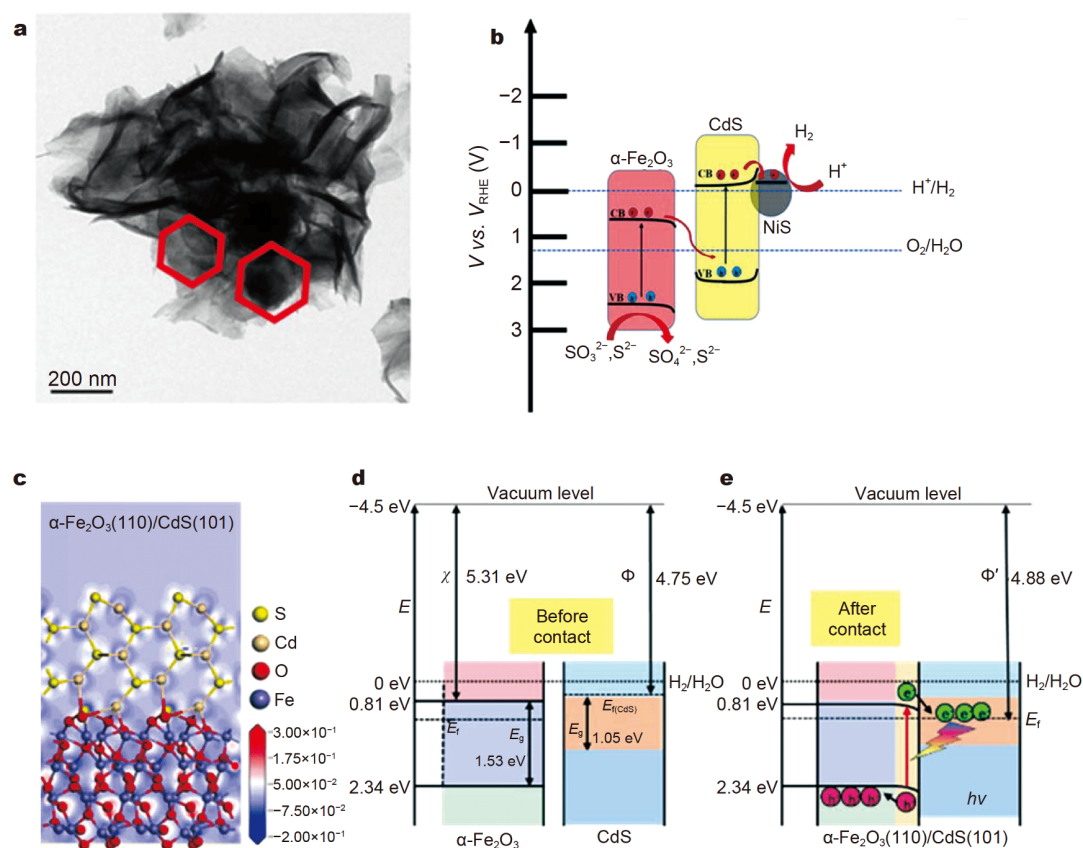


Figure 19 (a) TEM image of CdS/ α -Fe₂O₃, (b) photocatalytic mechanism of CdS/ α -Fe₂O₃/NiS, (c) simulated charge density difference distribution at the CdS/ α -Fe₂O₃ heterojunction interface, (d) and (e) the band bending in the space charge region at the interface before and after coupling α -Fe₂O₃ and CdS. Reprinted with permission from Ref. [179]. Copyright 2020, Elsevier.

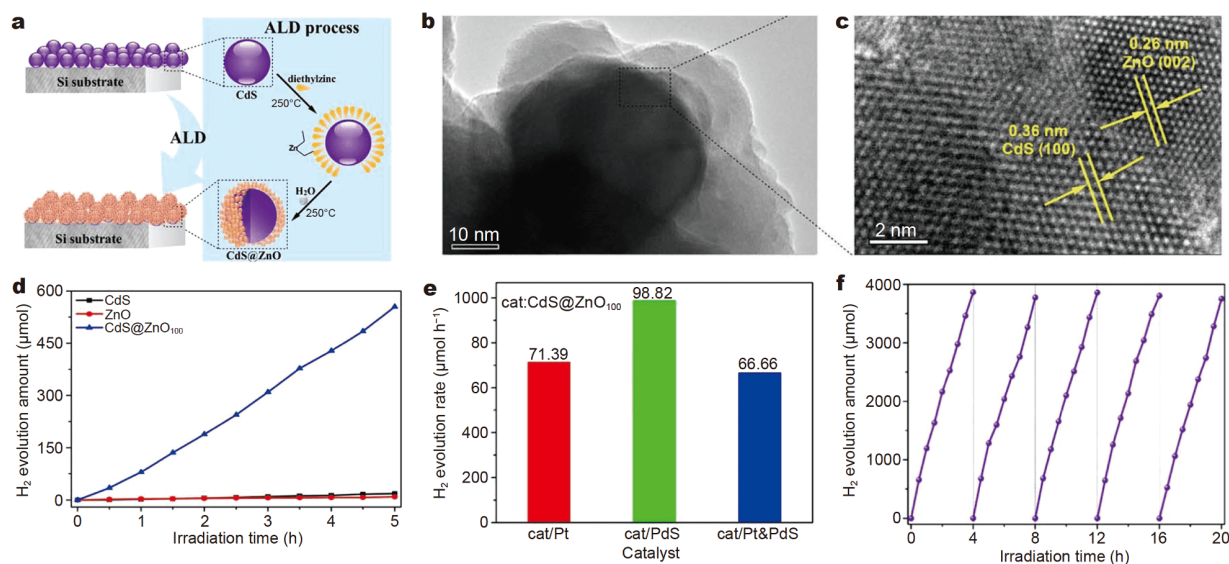


Figure 20 (a) Synthesis process, (b) TEM image, and (c) HRTEM image of CdS@ZnO core-shell photocatalysts. (d) H₂ evolution over CdS, ZnO, and CdS@ZnO; (e) average H₂ evolution rate; (f) cyclic runs for photocatalytic H₂ evolution over the CdS@ZnO-PdS. Reprinted with permission from Ref. [304]. Copyright 2017, Elsevier.

cocatalysts to promote the kinetics of photocatalytic H₂ generation. To date, various cocatalysts, such as Au [344,345], Pt [321,346,347], Ag₂S [348], MXenes [238,349–352], CoMoS_x [353], MoO_xS_y [354], MoS₂ [222,355–358], Ni₃C [176], Ni₂P [359,360], NiS_x [63,322,361], NiCoP [362], CoP_x [199,363], CuS_x [212,246], and WS₂ [364,365], have been coupled with CdS-based semiconductors for photocatalytic H₂ evolution. Typically, noble metals (Pt, Ag, and Au) are known as excellent cocatalysts for this application. For example, Liu *et al.* [232] demonstrated a facile and rapid ultrasonic-chemistry-based approach to load Pt onto CdS NRs, and the resulting Pt/CdS showed efficient H₂ evolution performances. The maximum H₂ evolution rate could reach 24.15 mmol g⁻¹ h⁻¹ over 0.5 wt.% Pt/CdS. Bao *et al.* [169] reported a facile aqueous solution process for the large-scale synthesis of nanoporous CdS NSs and CdS hollow NRs by the air-insensitive inorganic reactants of Na₂S·9H₂O and CdCl₂·2.5H₂O at room temperature. The obtained CdS/Pt exhibited the highest AQEs (60.34%) at 420 nm for photocatalytic H₂ generation.

However, the scarcity and high cost of noble metals restrict their large-scale applications in photocatalytic H₂ generation. Therefore, earth-abundant-metal-based and noble-metal-free cocatalysts have received increasing attention [4,18,366,367]. For example, Li *et al.* [229] found that a 0.2 molar ratio of NiS improved the average charge carrier lifetime of CdS by 97 times, potentially leading to more efficient charge separation and transfer. They synthesized CdS NWs and NiS with tight connections using a two-pot solvothermal synthesis. Tight interfaces between CdS and NiS with a smaller intrinsic bandgap led to significantly enhanced photocatalytic H₂ evolution activities. As the content increased, NiS aggregated on the surface of CdS and unfavorably caused fast recombination of photogenerated e⁻-h⁺ pairs. An evident decay was observed in the broad transient bleach signal after loading NiS, which indicated that the NiS improved the separation of CdS photogenerated e⁻ and h⁺ pairs. CdS with a NiS molar ratio of 20 reached an optimal photocatalytic H₂ evolution rate of 1512.4 μmol g⁻¹ h⁻¹ in lignin and lactic acid aqueous solution, which was 5041 times higher than that of pristine CdS [229]. The resulting CdS/NiS also exhibited excellent stability for 900 min of experiments in the lignin and lactic acid solution [229].

Cocatalysts supported on semiconductors can facilitate the prompt separation and migration of photoexcited charge carriers. For example, we reported a novel strategy to form 2D/2D nanocomposites by coupling Ni₂SP NSs with CdS NSs [286]. Fig. 21a depicts the ultrathin 2D NS

of CdS/Ni₂SP. The as-fabricated 2D/2D CdS/Ni₂SP photocatalysts displayed an outstanding H₂ generation performance under visible light (λ ≥ 420 nm). The CdS/2%Ni₂SP yielded the highest H₂ generation rate of 18.96 mmol g⁻¹ h⁻¹, which was approximately threefold higher than that of bare 2D CdS without the cocatalysts (Fig. 21b). As shown in Fig. 21c, the highest AQE was 4.8% at 420 nm. The *I*-*t* curve of CdS/2%Ni₂SP composites is obviously higher than that of nanostructured CdS, indicating that the 2D Ni₂SP cocatalysts promoted interfacial charge migration and accelerated charge utilization of nanostructured CdS (Fig. 21d). The interfacial coupling effects between the 2D Ni₂SP cocatalyst and 2D CdS enhanced photocatalytic H₂ generation by increasing the number of active sites, thereby facilitating rapid charge separation and transfer, while enhancing H₂ generation kinetics (Fig. 21e).

Recently, single metal atoms (serving as active sites) have shown excellent performances in photocatalytic and electrocatalytic reactions. The atomically dispersed single metal atoms on the semiconductor surface exhibit unique catalytic performances. For example, Zhang *et al.* [368] designed atomically dispersed Ni single atoms anchored on the surface of CdS NRs. Under visible light, the photocatalytic H₂ rate reached 630.1 mmol g⁻¹ h⁻¹. The single Ni atoms were stabilized by the Ni-O bonds. Density functional theory calculations revealed that the Ni atoms could optimize H₂ binding and electronic properties, thus improving the separation of photo-generated e⁻-h⁺ pairs. Meanwhile, Ni-CdS composites showed a stabilized photocatalytic performance during 16 h of irradiation. Some examples of CdS photocatalysts modified with cocatalysts are presented in Table 2.

In future, dual cocatalysts that combine different types of cocatalysts [369,370], including metal, metal oxide/sulfide/carbide/phosphide, and nanocarbons, might attract increasing attention in photocatalytic H₂ evolution over CdS-based photocatalysts. Additionally, apart from loading H₂ evolution cocatalysts, it should be noted that the oxidation cocatalysts are also expected to be loaded on CdS-based photocatalysts to boost charge separation and suppress photocorrosion, thereby fundamentally enhancing photocatalytic H₂ evolution [371].

SUPPRESSING CHARGE-INDUCED PHOTOCORROSION IN NANOSTRUCTURED CdS

Presently, research on the reaction mechanism and the fundamental carrier dynamics in CdS-based nanocomposite half-reaction systems has seen meaningful

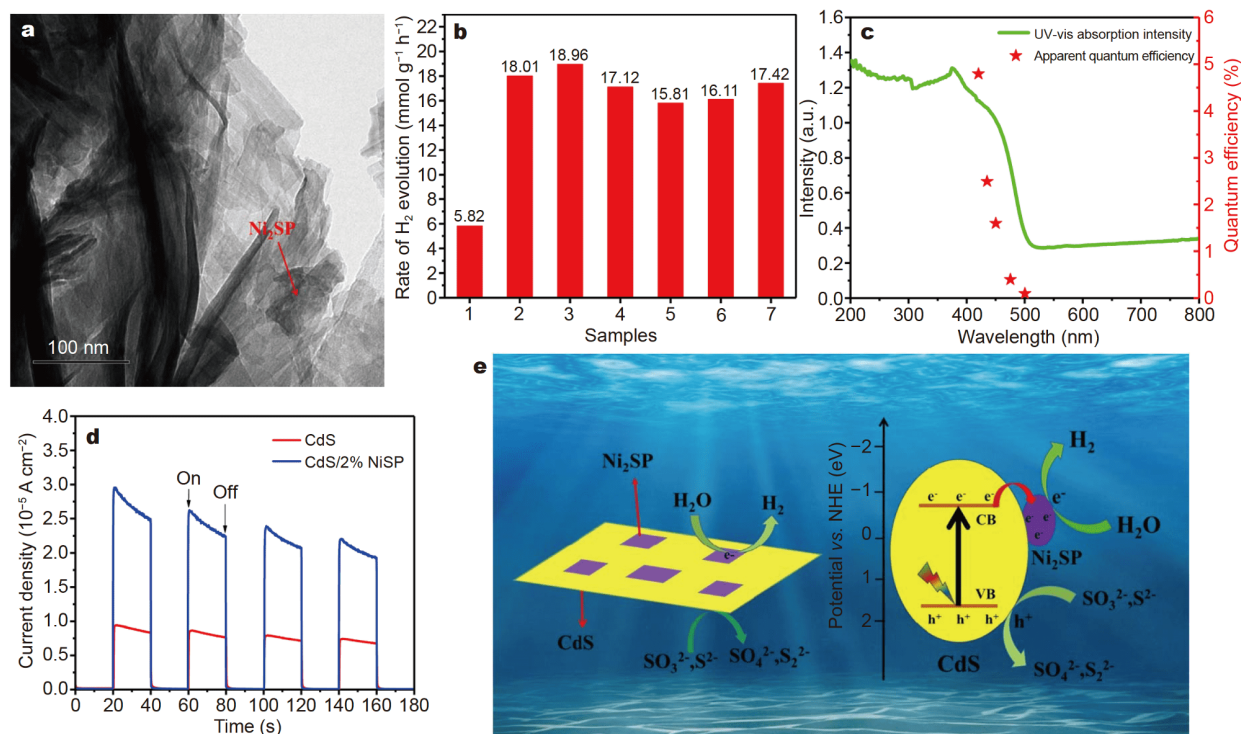


Figure 21 (a) HRTEM image of CdS/Ni₂SP, (b) average rate of H₂ evolution (1: CdS, 2: CdS/1%Ni₂SP, 3: CdS/2%Ni₂SP, 4: CdS/3%Ni₂SP, 5: CdS/4%Ni₂SP, 6: Ni₂SP, 7: CdS/2% NiS, 8: CdS/2%Ni₂SP nanoparticles), (c) AQEs of CdS/2%Ni₂SP, (d) photocurrent responses, and (e) proposed mechanistic scheme: (a) CdS; (b) CdS/1%Ni₂SP; (c) CdS/2%Ni₂SP; (d) CdS/3%Ni₂SP; (e) CdS/4%Ni₂SP; (f) Ni₂SP; (g) CdS/2%NiS; (h) CdS/2%Ni₂SP nanoparticles. Reprinted with permission from Ref. [286]. Copyright 2020, Elsevier.

progress. However, due to the photocorrosion phenomenon of CdS-based nanocomposites, the stability and durability of most reported CdS-based photocatalysts need further improvement. The accumulation of photoexcited h^+ in its VB can induce the self-oxidation of S^{2-} in CdS, which results in the dissolution of Cd^{2+} . Apparently, the effective removal of photoexcited h^+ from the VB of CdS impedes the self-oxidation of S^{2-} , enhancing the durability and photostability of CdS. It is well known that the use of sacrificial reagents (such as $Na_2S-Na_2SO_3$, ethanol, and lactic acid) as e^- donors for CdS photocatalysts in photocatalytic H₂ generation systems is an effective strategy to suppress photocorrosion of CdS.

Furthermore, Z-scheme heterojunctions could reduce the accumulation of h^+ in the VB of CdS. Li *et al.* [381] found that the formation of CdS–MnS Z-scheme photocatalysts through a cation exchange process could maintain good stability over 42 h of measurement (Fig. 22a and b). The chopped photocurrent density–voltage measurements show that the pure CdS was photocorroded rapidly within 800 min of illumination (Fig. 22c). After the formation of the Z-scheme heterojunction between

CdS and MnS, the photocorrosion phenomenon was suppressed. Moreover, the formation of Z-scheme heterojunctions also increased the light-absorption range from 520 to 800 nm (Fig. 22d). Consequently, the obtained CdS–MnS showed an optimal H₂ evolution rate of 1595 $\mu\text{mol g}^{-1} \text{h}^{-1}$ with an AQE of 22.6% at 420 nm [381].

Constructing a p–n heterojunction is another strategy to transfer h^+ from the VB of CdS. After the formation of the p–n heterojunction, the h^+ in the VB of CdS could be transferred to the relatively positive VB of a p-type semiconductor, which effectively reduces h^+ accumulation in the VB of CdS. Ai *et al.* [382] designed CdS@Ti₃C₂@CoO with a hierarchical tandem p–n heterojunction for photocatalytic HER. The insertion of Ti₃C₂ on the surface of CdS served as a platform for the growth of CoO NPs, which were introduced as a bridge to consolidate CdS and CoO into a special tandem p–n heterojunction. Meanwhile, Ti₃C₂ also served as a bridge with a powerful unidirectional internal electric field wherein another internal electric field was generated between Ti₃C₂ and CoO, causing photogenerated carriers to transfer to Ti₃C₂ and restricting h^+ migration (Fig. 23a). Thus, CdS@

Table 2 Cocatalyst loading on CdS for photocatalytic H₂ production

Nanocomposites	Content of cocatalysts	Light source	Sacrificial reagent	HER (mmol g ⁻¹ h ⁻¹)	AQE (420 nm)	Ref.
Pt-PdS/CdS	0.3 wt.%	300 W Xe lamp	Na ₂ S-Na ₂ SO ₃	29.23	93	[173]
MoS ₂ /CdS	0.2 wt.%	300 W Xe lamp	Lactic acid	5.4	-	[166]
WS ₂ /CdS	1 wt.%	300 W Xe lamp	Lactic acid	4.2	-	[364]
NiS/CDs/CdS	10 wt.%	350 W Xe lamp	Na ₂ S-Na ₂ SO ₃	1.44	-	[257]
CdS/Co-MoS _x	2 mol%	300 W Xe lamp	Lactic acid	0.54	23.5	[263]
NiS/CdS	1.2 mol%	300 W Xe lamp	Lactic acid	2.18	51.3	[174]
CdS/WS ₂ /graphene	4.2 wt.%	300 W Xe lamp	Na ₂ S-Na ₂ SO ₃	1.84	21.2	[240]
PdNi/CdS	2 wt.%	300 W Xe lamp	(NH ₄) ₂ SO ₃	32.4	63.97	[372]
PtNi _x /CdS	2.0 wt.%	300 W Xe lamp	(NH ₄) ₂ SO ₃	11.4	51.24	[373]
CdS/g-C ₃ N ₄ /CuS	10 wt.%	350 W Xe lamp	Na ₂ S-Na ₂ SO ₃	1.15	16.5	[212]
CuS/CdS	25 wt.%	300 W Xe lamp	Lactic acid	5.62	19.7	[374]
Pt/CdS	0.5 wt.%	300 W Xe lamp	Lactic acid	24.15	-	[232]
MoS ₂ /CdS	3 wt.%	300 W Xe lamp	Na ₂ S-Na ₂ SO ₃	11.4	-	[213]
Ni@NiO/CdS/g-C ₃ N ₄	1 wt.%	300 W Xe lamp	Triethanolamine	1.26	-	[218]
Ni ₂ P/CdS	3.51 wt.%	300 W Xe lamp	Na ₂ S-Na ₂ SO ₃	44.65	-	[256]
Co(OH) ₂ /CdS	6.5 mol%	350 W Xe lamp	Lactic acid	14.43	-	[375]
NiSe ₂ /CdS	5 wt.%	300 W Xe lamp	Na ₂ S-Na ₂ SO ₃	167.1	1.5	[241]
VC/CdS	15 wt.%	300 W Xe lamp	TEOA	14.2	8.7	[215]
Cobalt-salen/CdS	0.015 mmol L ⁻¹	300 W Xe lamp	Na ₂ S-Na ₂ SO ₃	106	29	[235]
NiO _x /CdS	1 mol%	300 W Xe lamp	Methanol and Ni(CH ₃ COO) ₂	5.9	8.6	[57]
NiS/CdS	20 mol%	300 W Xe lamp	Lignin and lactic acid	0.15	44.9	[229]
CDs/CdS-S	1 wt.%	300 W Xe lamp	Lactic acid	4.64	11.8	[236]
MoS ₂ /CdS	5 wt.%	300 W Xe lamp	Na ₂ S-Na ₂ SO ₃	4.65	7.31	[255]
CdS/MnS	5 wt.%	300 W Xe lamp	Lactic acid	15.55	6.9	[233]
Cu ₂ MoS ₄ /CdS	5 wt.%	150 W Xe lamp	Lactic acid	15.56	-	[231]
Ni ₂ P/MCdS-DETA	0.4 wt.%	300 W Xe lamp	Na ₂ S-Na ₂ SO ₃	6.84	26.4	[359]
Ni ₃ B/CdS	0.8 wt.%	300 W Xe lamp	Lactic acid	4.8	21	[376]
RhP/CdS	20 wt.%	5 W LED	Lactic acid	0.33	34.3	[221]
P-MoS ₂ /CdS	20 wt.%	300 W Xe lamp	Na ₂ S-Na ₂ SO ₃	5.89	19	[220]
Mn ₁₃ -cluster/CdS	7 wt.%	White-light LED	Lactic acid	3.6	-	[377]
Ni _{2-x} Co _x P/CdS	40 wt.%	300 W Xe lamp	Ethanol-water	218	76.3	[378]
CdS/Ti ₃ C ₂	10%	300 W Xe lamp	Lactic acid	2.407	35.6	[238]
CdS/MoN ₂	-	300 W Xe lamp	Lactic acid	9.2	-	[379]
MXene@CdS	-	300 W Xe lamp	Na ₂ S-Na ₂ SO ₃	12.34	-	[349]
CdS/SnO ₂	5%	150 W Xe lamp	Lactic acid	20.2	-	[242]
CdS/Mo ₂ C	3%	300 W Xe lamp	Na ₂ S-Na ₂ SO ₃	1.843	-	[380]
CdS-FeP	5%	300 W Xe lamp	Lactic acid	18.63	11.2	[250]

Ti₃C₂@CoO (CTC-5-5) maintained good stability over 10 cycles, which was significantly better than that of CdS@Ti₃C₂ (CT-5) and CdS@CoO (CC-5) (Fig. 23b and c). These results indicate that its combination with a p-type semiconductor to form a p-n heterojunction can suppress the extent of photocorrosion [382].

Besides constructing a heterojunction to avoid the ac-

cumulation of h⁺ in the VB of CdS, coating a chemically inert shell could also prevent the undesirable photocorrosion. Ning *et al.* [383] constructed a thin shell layer of Al₂O₃ on CdS NPs, which could remove the dissolved O₂ to inhibit photocorrosion (Fig. 24a). During the photocatalytic reaction, the photogenerated h⁺ are trapped by the Al³⁺ cationic vacancy network in amorphous

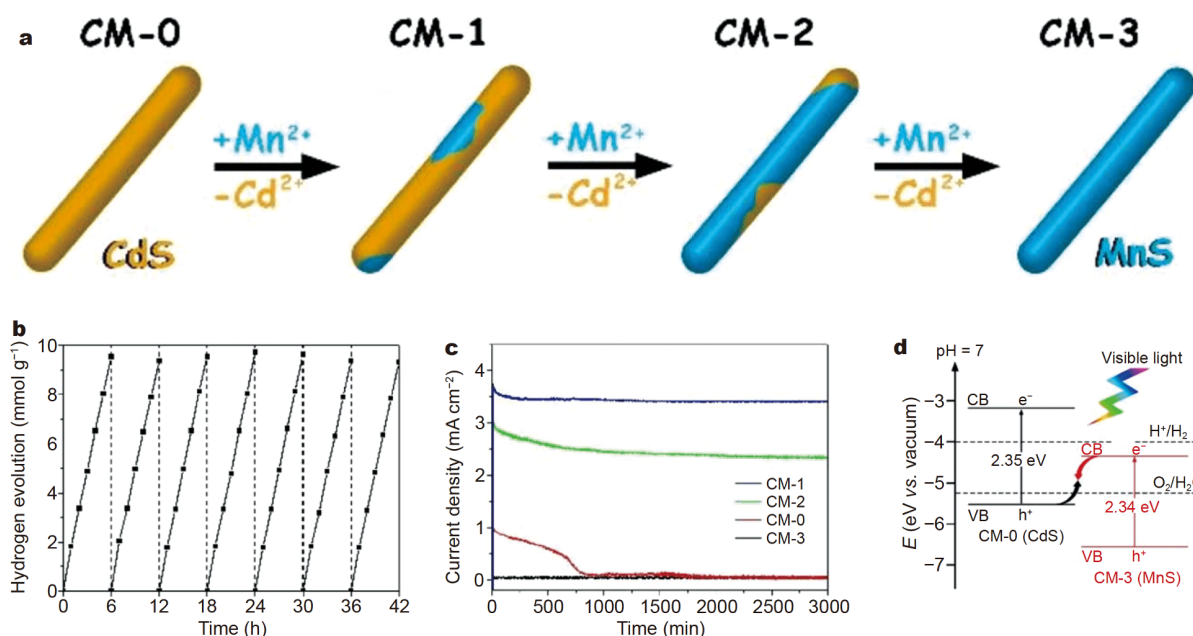


Figure 22 (a) Schematic of the synthesis process, (b) cyclic experiment of H₂ generation using CdS–MnS, (c) amperometric photocurrent density measurements (0.5 mol L⁻¹ Na₂SO₄, 100 mW cm⁻², visible light), (d) Z-scheme electronic band structures between CdS and MnS heterojunction photocatalysts. Reprinted with permission from Ref. [381]. Copyright 2020, Wiley-VCH.

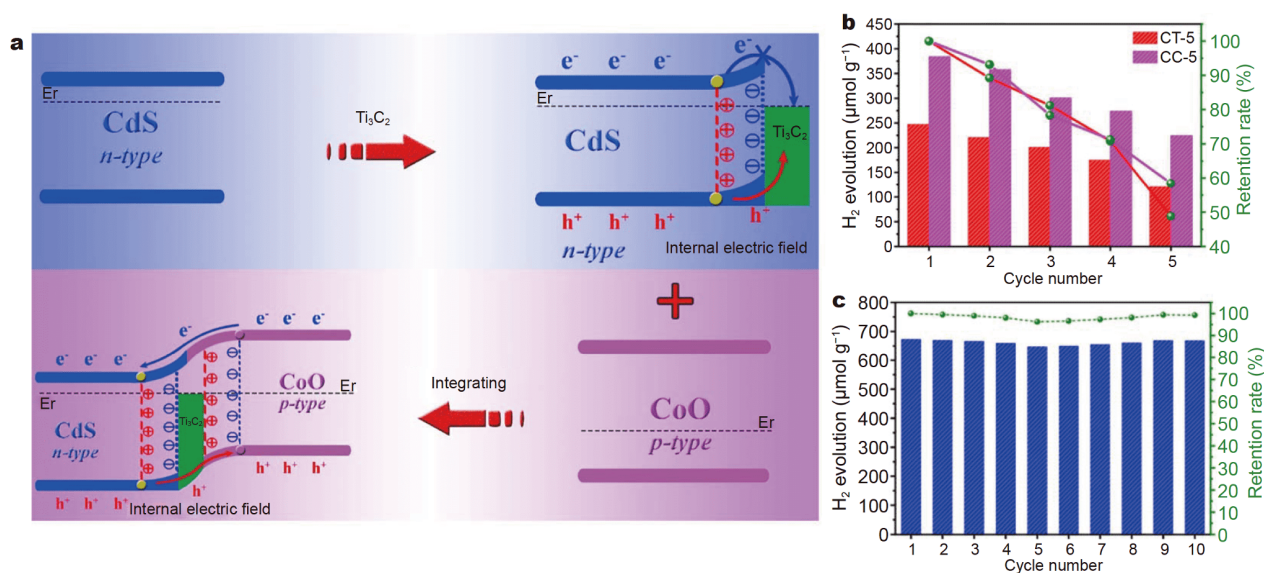


Figure 23 (a) Schematic of CdS@Ti₃C₂@CoO hierarchical tandem p–n heterojunction; recycling tests of photocatalytic H₂ evolution of (b) CT-5 and CC-5, (c) CTC-5-5. Reprinted with permission from Ref. [382]. Copyright 2020, Elsevier.

Al₂O₃. Compared with pure CdS, only trace amounts of Cd²⁺ were dissolved in the reaction solutions. Thus, CdS/Al₂O₃–Pt exhibited excellent stability during the photocatalytic HER (Fig. 24b and c). Moreover, CdS/Al₂O₃–Pt could induce overall water splitting by avoiding photo-

corrosion (Fig. 24d) [383].

CONCLUSIONS

In recent years, CdS-based nanostructured photocatalysts have been widely investigated. Significant progress has

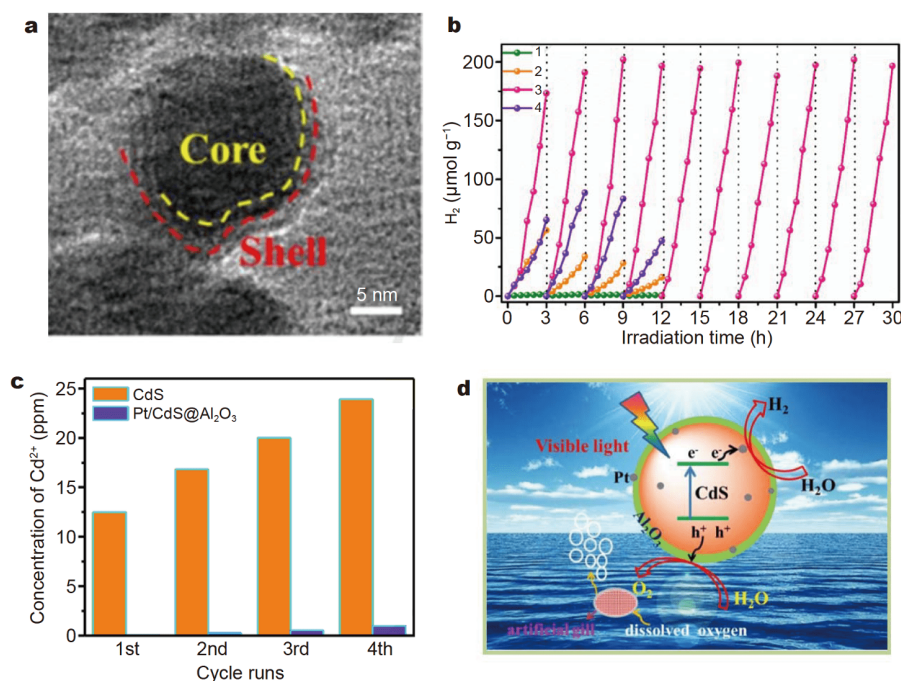


Figure 24 (a) TEM image of CdS@Al₂O₃, (b) cyclic runs of the photocatalytic H₂ evolution activity under visible light over various catalyst samples: (1) CdS NPs, (2) Pt/CdS, (3) Pt/CdS@Al₂O₃ with artificial gill and (4) Pt/CdS@Al₂O₃ without artificial gill, (c) the changes in cadmium ion concentration in the CdS NPs and Pt/CdS@Al₂O₃ composite solution with extended irradiation time, and (d) mechanism of overall water splitting over Pt/CdS@Al₂O₃ composite under visible illumination. Reprinted with permission from Ref. [383]. Copyright 2018, Elsevier.

been made in the field of solar-fuel conversion and environmental purification using CdS [15,213,364,384]. This review first systematically discusses the recent progress of CdS-based nanostructured photocatalysts with a detailed summary of the design strategies adopted to enhance H₂ generation efficiency. Although great advances have been made in recent years, many challenges remain, entailing the need for an in-depth understanding of the underlying mechanism for the observed H₂ evolution enhancement. Their intrinsic relations with modification routes should be unambiguously established [361,370,385,386].

Various strategies have been devised to increase photocatalytic H₂ generation using photocatalytic CdS-based nanomaterials: constructing multicomponent solid solutions, loading cocatalysts, constructing Z-scheme heterojunctions, designing 2D NSs, and reducing photocorrosion. These aspects are thoroughly highlighted and discussed. Notably, 2D nanomaterials possess many advantages, such as good chemical and physical stability, high interlayer adhesion, large surface area, high electron mobility, and rich surface-active sites, which facilitate photocatalytic H₂ evolution. In addition, bandgap tuning and heterojunction construction can be conveniently

engineered by combining the strengths of different 2D nanomaterials in the form of 2D/2D layered nanocomposites [72,387]. Therefore, it is possible to obtain CdS photocatalysts with extended visible-light absorption and enhanced redox capacities, as well as better charge separation efficiencies. The durability and stability of photocatalysts remain an urgent problem that needs to be solved in the future.

A knowledge gap remains in the fundamental revelation of the actual active sites in CdS photocatalysts. Specifically, a far-reaching investigation into the detailed mechanisms of how CdS photocatalysts function through the advancement *in situ/operando* characterizations is highly sought. The e⁻ transfer mechanisms between the heterojunctions of CdS and other components are still debatable. Upon clarifying the transfer mechanism, targeted e⁻ transfer to the selected active sites can significantly improve the photocatalytic HER performance of CdS photocatalysts. In addition, shortening the migration distance is effective in increasing the consumption rate of the photogenerated e⁻-h⁺ pairs. The construction of single-layer CdS nanosheets could minimize the charge migration distance from the bulk to the CdS surface. Therefore, developing methodologies to

synthesize ultrathin CdS is expected to continue to receive widespread attention.

A theoretical mechanistic study using computational methods is another important aspect to assist in the rational design of CdS photocatalysts with ideal electronic structures, species adsorption, and other properties. In addition, it could provide insights into the cocatalyst/semiconductor loading sites and interfacial interactions to guide the preparation of functional CdS. In summary, the exploration of new strategies to strengthen the overall performance of CdS-based nanocomposites, including their stability, can offer new opportunities toward the efficient utilization of solar energy for chemical fuel conversion.

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Conflict of interest The authors declare that they have no conflict of interest.



Rongchen Shen received his Bachelor degree from Huaibei Normal University in 2016 and Master degree from South China Agricultural University in 2019. He is a PhD candidate at South China Agricultural University. His research interests mainly focus on two-dimensional materials for photocatalytic application.



Yun Hau Ng is an associate professor at the School of Energy and Environment, City University of Hong Kong. He received his PhD from Osaka University in 2009. Before joining the City University of Hong Kong, he was a lecturer (2014) and senior lecturer (2016) in the School of Chemical Engineering, University of New South Wales. His research is focused on the development of novel photoactive semiconductors for solar energy conversion. He received the APEC ASPIRE Prize in 2019, Distinguished Lectureship

Award from the Chemical Society of Japan in 2018, and Honda-Fujishima Prize by the Electrochemical Society of Japan in 2013.



Peng Zhang received his PhD in materials physics and chemistry from Northeast Normal University, China (2014). After postdoctoral research at Helmholtz-Zentrum Berlin, Germany, he joined the School of Materials Science and engineering at Zhengzhou University. His current research interests focus on micro- and nanotechnology including low-dimensional carbon-based materials (graphene, CNF, carbon layer, etc.) and their applications in the fields of environment remediation and energy storage. He has published more than 60 SCI papers in the above fields such as *Adv. Mater.*, *Adv. Func. Mater.*, *Energy Storage Mater.* His work has been cited more than 4,000 times, and his H-index is 33.



Xin Li received his BS and PhD degrees in chemical engineering from Zhengzhou University in 2002 and South China University of Technology in 2007, respectively. Then, he joined South China Agricultural University as a faculty staff member, and became an associate professor of applied chemistry in 2011. In 2017, he became a Professor at South China Agricultural University. During 2012–2013, he worked as a visiting scholar at the Electrochemistry Center, the University of Texas at Austin, USA. His research interests include photocatalysis, photoelectrochemistry, adsorption, and the development of nanomaterials and devices (see <http://www.researcherid.com/rid/A-2698-2011>).

纳米结构硫化镉光催化分解水产氢综述

沈荣晨^{1†}, 任豆豆^{1†}, 丁英娜², 关雅彤², 吴永豪^{3*}, 张鹏^{4*}, 李鑫^{1*}

摘要 太阳能驱动光催化分解水产氢是实现可持续制氢气的一种有效策略。硫化镉半导体光催化剂基于其较强的可见光响应、适宜的氧化还原反应带边位置以及优异的电荷传输性能而备受关注。本文综述了近年来国内外在提高硫化镉基光催化剂制氢性能的设计、改性和制备等方面的研究进展。首先简要介绍了光催化制氢的基本概念和机理，阐述了硫化镉光催化制氢的基本性质、重要进展和瓶颈，综述了该材料的发展前景。随后，重点讨论了硫化镉基光催化剂光催化分解水产氢的各种改性的策略，其中有效的策略是产生更多的载流子，促进电荷的有效分离，促进界面电荷转移，加速电荷利用，以及抑制电荷诱导的自光腐蚀。针对每一种改性策略，都详细讨论了影响光催化剂性能的重要因素和未来潜在的研究方向。最后介绍了纳米结构硫化镉和硫化镉基纳米复合材料在光催化分解水产氢中的发展前景和面临的挑战。本综述将为开发镉基半导体光催化剂提供重要和及时的理论指导，并促进其在太阳能氢气生产中的应用。