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**Recent Advances in Visible-Light Driven Photocatalysis**

Yuhua Wang, Xinlong Ma, Hao Li, Bin Liu, Huihui Li, Shu Yin and Tsugio Sato

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**Abstract**

Semiconductor photocatalysis has been considered a potentially promising approach for renewable energy and environmental remediation with abundant solar light. However, the currently available semiconductor materials are generally limited by either the harvesting of solar energy or insufficient charge separation ability. To overcome the serious drawbacks of narrow light-response range and low efficiency in most photocatalysts, many strategies have been developed in the past decades. This article reviews the recent advancements of visible-light-driven photocatalysts and attempts to provide a comprehensive update of some strategies to improve the efficiency, such as doping, coupling with graphene, precipitating with metal particles, crystal growth design, and heterostructuring. A brief introduction to photocatalysts is given first, followed by an explanation of the basic rules and mechanisms of photocatalysts. This chapter focuses on recent progress in exploring new strategies to design TiO$_2$-based photocatalysts that aim to extend the light absorption of TiO$_2$ from UV wavelengths into the visible region. Subsequently, some strategies are also used to endow visible-light-driven Ag$_3$PO$_4$ with high activity in photocatalytic reactions. Next, a novel approach, using long afterglow phosphor, has been used to associate a fluorescence-emitting support to continue the photocatalytic reaction after turning off the light. The last section proposes some challenges to design high efficiency of photocatalytic systems.

**Keywords:** Photocatalysts, TiO$_2$, Ag$_3$PO$_4$, graphene, long afterglow phosphor

**1. Introduction**

It is well known that environmental pollution is affecting human survival and development. Photocatalytic technology is considered as an efficient, stable, and environmentally friendly method in the field of environmental pollution control [1]. In general, a photocatalytic reaction includes three steps [2] (Fig. 1). First, electrons (e$^-$) and holes (h$^+$) are generated through...
photoexcitation. Then, these electrons and holes migrate to the surface of photocatalysts and subsequently react with adsorbed electron acceptors and donors, respectively. Thus, an efficient photocatalyst requires a semiconductor with a suitable band gap for harvesting light, facile separation and transportation of charge carriers, and proper valence band (VB) and conduction band (CB) edge potential for redox reaction being feasible. To date, much effort is currently focused on how to improve the light absorption, charge separation, and surface reactivity in order to achieve outstanding photocatalytic performance, such as doping, coupled with graphene, precipitated with metal particles, crystal growth design, and heterostructuring.

In the past few decades, TiO$_2$ nanomaterials have attracted tremendous interest in environmental pollution removal and photocatalytic hydrogen generation. However, all of them have large electronic band gaps of 3.0–3.2 eV, which means only less than 5% of the entire solar energy can be utilized. Thus, much effort has been devoted to bring about the absorption of TiO$_2$ into the visible-light region and improved photocatalytic activity.

Figure 1. Schematic illustration of the reactions following light absorption by a photocatalyst. (a) Electron–hole pair formation; (b) donor oxidation by hole; (c) reduction by electron; (d) and (e) electron–hole recombination on the surface or in the bulk. Adapted with permission from ref. 2. © 1995 ACS.

Despite the fact that there are many research articles on modified TiO$_2$, the exploration of new active and efficient visible-light-driven photocatalysts attracts much attention [3–22]. In the effort to exploit novel photocatalyst systems working under visible light, it has been revealed that orbitals of some p-block metals with a d10 configuration [22], such as Ag 4d in Ag(I), could hybridize O 2p levels to form a new preferable hybridized VB, thus narrowing the band gap to harvest visible light.

Compared with single-phase semiconductor photocatalysts, hybrids of two or more semiconductor systems, i.e., heterostructures, seem to possess advantages in more efficiently utilizing solar light. Besides metal/semiconductor and carbon group materials/semiconductor-heterostructured photocatalysts [23–28], semiconductor/semiconductor-heterostructured photocatalysts with diverse models have been developed (Fig. 2), including type-I and type-II heterojunctions, Z-scheme, p–n heterojunctions, and homojunction band alignments [29–32].
2. TiO$_2$-based photocatalysts

In this section, strategies on the modulation of energy band structures or surface states of wide-band-gap photocatalysts, especially for TiO$_2$, are reviewed. Currently, many strategies have been developed to modulate the band structures of TiO$_2$, including doping, coupling with graphene, precipitating with metal particles, crystal growth design, and heterostructuring, which will be discussed.

2.1. Doping

Doping is an available strategy to tune the absorption band of wide-band photocatalysts. Several studies have investigated nonmetal doping. Asahi et al. reported nitrogen-doped TiO$_2$ of nonmetals, such as N [33, 34], C [35–37], S [38, 39], B [40–42], F [43–45], Br [46], I [47–50], P [51], in 2001 [33]. In our study [34], nitrogen-doped titania nanoparticles (NPs) were successfully prepared by a microwave-assisted solvothermal process in a very short time, and the prepared samples showed visible-light absorption in the range of 400–550 nm, indicating its potential applications as visible-light-induced photocatalyst. The chemical states and locations of dopants are considered to be key factors in adjusting the spectral distribution of
the induced electronic states of those dopants and reconstructing favorable surface structure for photocatalysis.

Co-doping with two suitable heteroatoms can also achieve substantial synergistic effects [41, 52–55]. For example, B, N co-doped TiO$_2$ benefits from the B–N bonds formed, which can increase the amount of doped N on the TiO$_2$ surface and the promoted separation of photo-excited electron–hole pairs [41].

2.2. TiO$_2$-graphene composite

Graphene, a two-dimensional carbonaceous material, can be used in many applications due to its unique and remarkable properties such as high conductivity, large surface area, and good chemical stability [56–59]. Tremendous interest is devoted to fabricating numerous graphene–semiconductor composites to aid charge separation and migration and improve the performance of the photocatalysts [60–64]. Graphene works as an electron acceptor or transporter to induce electron transfer, leading to an efficient charge separation. Thus, an appropriate integration of graphene and TiO$_2$ could give rise to a nanocomposite that combines the desirable properties of graphene and TiO$_2$, e.g., the photocatalytic activity of TiO$_2$ can be improved. In the past few years, there were some reports about graphene–TiO$_2$ composites [13–17].

Recently, Liu et al. synthesized the graphene oxide–TiO$_2$ nanorod composites [65, 66]. Wu et al. reported the synthesis and application of graphene–TiO$_2$ nanorod hybrid nanostructures in microcapacitors [67]. As shown in Fig. 3, the assembling of TiO$_2$ nanocrystalline with exposed {001} facets on graphene sheets reported in our previous work showed a higher photocatalytic activity than the other normal TiO$_2$/graphene composites [68]. In another work (Fig. 4), graphene/rod-shaped TiO$_2$ nanocomposite was synthesized by the solvothermal method [69]. In a one-pot system, the rod-shaped TiO$_2$ can be homogeneously dispersed on the surface of graphene sheets by syngraphenization strategy. Owing to the combination of graphene and rod-shaped TiO$_2$, the graphene/rod-shaped TiO$_2$ nanocomposite shows a significant enhancement in the photocatalytic performance compared with that of the graphene/spherical TiO$_2$ nanocomposite, which can be attributed to the high electronic mobility of graphene, higher Brunauer-Emmett-Teller (BET) surface area, and rod-shaped structures of TiO$_2$. In our recent work [70], a series of B-doped graphene/rod-shaped TiO$_2$ nanocomposites were synthesized via one-step hydrothermal reaction. The photocatalytic activity of the obtained nanocomposites for the oxidative photodestruction of NO$_x$ gas showed better photocatalytic properties than pure TiO$_2$ and graphene/TiO$_2$ nanocomposites. This work provides new insight into the fabrication of TiO$_2$–carbon nanocomposites as high-performance photocatalysts and facilitates their application in addressing environmental protection issues.

2.3. TiO$_2$-based plasmonic photocatalysts

Plasmonic photocatalysis has offered a new opportunity to solve the problem of the limited efficiency of photocatalysts [71–73]. In these photocatalysts, the nanostructured plasmonic metals are often combined with a semiconductor-based material (e.g., TiO$_2$), and the photocatalytic activity is greatly enhanced due to the local surface plasmon resonance (LSPR) effect...
The LSPR effect endows the metal nanocrystals with very large absorption and scattering cross sections and local electromagnetic field enhancement in the near-field region near the surface of plasmonic metal nanocrystals, which is promising in manipulating light absorption in photocatalytic systems. The intensity of the local electromagnetic field is several orders of magnitude larger than that of the far-field incident light, and the highest charge carrier formation is observed at the semiconductor/liquid interface, which benefits the photocatalytic reactions [71–72].

Recently, noble metal nanoparticle-deposited TiO$_2$ has attracted significant attention. The main advantages of these noble metal NPs can be attributed to their chemically inert properties [74]. The LSPR effect endows the metal nanocrystals with very large absorption and scattering cross sections and local electromagnetic field enhancement in the near-field region near the surface of plasmonic metal nanocrystals, which is promising in manipulating light absorption in photocatalytic systems. The intensity of the local electromagnetic field is several orders of magnitude larger than that of the far-field incident light, and the highest charge carrier formation is observed at the semiconductor/liquid interface, which benefits the photocatalytic reactions [71–72].

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toward (photo) oxidation and LSPR effect on the surface [75, 76]. Meanwhile, our previous work found the plasmonic enhancement of the photocatalytic activity of semiconductor–metal nanocomposite materials [77–79]. Thus, noble metal NP-deposited TiO\(_2\) could be an appropriate approach to improve the photocatalytic performance of TiO\(_2\).

A variety of nanostructured Au/TiO\(_2\) with different morphologies have been synthesized, such as highly stable mesoporous Au/TiO\(_2\) spheres (~500 nm) [80] and mesoporous Au–TiO\(_2\) nanocomposites using a simple spray hydrolytic method [81]. In our study, Au NPs are precipitated on the highly porous one-dimensional (1D) TiO\(_2\) nanotubes (NTs), and the plasmonic photocatalytic properties of the material are investigated [82]. Compared with nanoparticles, there are some advantages of 1D NT structures, such as favorable recycling characteristics and the vectorial transport of photogenerated charge carriers [83, 84], which have great potential for superior photocatalytic performance. The Au NPs/TiO\(_2\) NTs were synthesized by emulsion electrospinning followed by deposition–precipitation (DP) method. The results in Fig. 5 show that the modified porous TiO\(_2\) NTs with the presence of Au NPs increased photocatalytic destruction of methylene blue (MB) solution under visible-light irradiation. Furthermore, the migration of Au NPs from the rutile phase to the interface of

Figure 4. TEM images of (a1) graphene oxide, (a2) graphene, (a3) 0.48% GR/RT, and (a4) HRTEM image of a single rod-shaped TiO\(_2\) nanocrystal in the composite; (b) photocatalytic degradation of MO solution over graphene/rod-shaped TiO\(_2\) nanocomposites with various graphene contents compared with that of GR/ST; (c1) SEM image, (c2) TEM image, (c3) EDX, and (c4) HRTEM image of 7.5% BG/RT; (d) photocatalytic activity for the destruction of NO\(_x\) gas under UV irradiation using different photocatalysts. Reproduced with permission from ref. 69 & ref. 70 © 2012 & 2014 RSC.
rutile/anatase was found when the calcination temperature changed from 250 °C to 350 °C. The optimal photocatalytic activity was obtained in the sample $\text{Au}_3(\text{DP}_{350})/\text{TiO}_2$, due to the plasmon activation of the Au NPs followed by consecutive electron transfer that induced efficient charge separation. Therefore, such a highly porous Au/TiO$_2$ heterojunction structure provides a new pathway for the design and fabrication of other energy- and environment-related applications.

Figure 5. (a) SEM and (b) TEM images of as-prepared TiO$_2$ NTs and (c) SEM and (d) TEM images of the representative sample $\text{Au}_3(\text{DP}_{350})/\text{TiO}_2$ NTs; TEM images of Au$_3$(DP$_y$/TiO$_2$ ($y = 150$ (e11), 250 (e12), 350 (e13), and 450 °C (e14), respectively) and the dispersion of Au co-catalysts (e21), (e22), (e23), and (e24), respectively; (f1) effect of calcination temperature on the location and size of the Au NPs on TiO$_2$ NTs; (f2 and f3) TEM and HRTEM images of $\text{Au}_3(\text{DP}_{350})/\text{TiO}_2$. Variation of normalized $C/C_0$ of MB concentration as a function of visible-light irradiation time for (g1) Au$_x$(DP$_{y}$/TiO$_2$ and (g2) $\text{Au}_3(\text{DP}_{y}$/TiO$_2$ ($y = 150$, 250, 350, 450 °C, respectively); (g3) schematic diagram for the possible mechanism for photocatalytic degradation of MB over $\text{Au}_3(\text{DP}_{350})/\text{TiO}_2$ under visible-light irradiation. Reproduced with permission from ref. 82 © 2013 RSC.
2.4. TiO$_2$-based heterostructure photocatalysts

The heterojunctions provide a facile way to enable the effective separation of photoexcited electron–hole pairs, thus to enhance the photocatalytic performance. Table 1 lists recent prominent heterostructured photocatalysts as well as their photocatalytic application.

<table>
<thead>
<tr>
<th>Type</th>
<th>Heterostructured photocatalysts</th>
</tr>
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<tr>
<td></td>
<td>ZnO/CdS,[93] AgInS5S8/TiO$_2$[94]</td>
</tr>
<tr>
<td></td>
<td>Ag$_2$VO$_4$/TiO$_2$[95] ZnFe$_2$O$_5$/TiO$_2$[96]</td>
</tr>
<tr>
<td></td>
<td>CuInSe$_2$/TiO$_2$[102] TiO$_2$/ZnO,[103] ZnFe$_2$O$_5$/TiO$_2$[104]</td>
</tr>
<tr>
<td></td>
<td>NiO/ZnO[105]</td>
</tr>
<tr>
<td></td>
<td>Pt/n-Si/n+–Si/Ag[111]</td>
</tr>
<tr>
<td>Z-scheme</td>
<td>CdS/Au/TiO1.96C0.04,[112] CdS/Au/TiO$_2$,[113] ZnO/CdS,[114] CdS/Au/ZnO$_2$[115]</td>
</tr>
<tr>
<td></td>
<td>CuO/TiO$_2$[116] CaFe$_2$O$_4$/WO$_3$[117]</td>
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Table 1. Recent reports of diverse heterostructured photocatalysts.

According to different electronic energy levels and band gaps of photocatalysts, five kinds of semiconductor heterojunctions have been reported: straddling alignment (type I), staggered alignment (type II), Z-scheme system, homojunctions, and p–n heterojunctions [118, 119]. In those work, the dynamics of electron and hole were studied, including the band gap, the electron affinity, and the work function of different semiconductor heterojunctions.

3. Visible-light-driven Ag$_3$PO$_4$ photocatalysts

A breakthrough was made in finding a novel semiconductor material, Ag$_3$PO$_4$, as an active visible-light-induced photocatalyst [4]. Ag$_3$PO$_4$ demonstrates extremely high capability for O$_2$ evolution from H$_2$O and organic dye decomposition under visible-light irradiation [120]. More importantly, this novel photocatalyst can achieve a quantum efficiency up to 90% at wavelengths longer than 420 nm, which is clearly higher than that reported previously by using semiconductor photocatalysis.

So far, various methods have been proposed to further enhance the photocatalytic activity of Ag$_3$PO$_4$ under visible-light irradiation. One approach is the synthesis of Ag$_3$PO$_4$ with various morphologies. This is because photocatalytic reactions are typically surface-based processes; thus, the photocatalytic efficiency is closely related to the morphology and microstructure of a photocatalyst [83]. Recently, some new morphologies of Ag$_3$PO$_4$ have been developed [120–126]. For example, Bi et al. fabricated the single-crystalline Ag$_3$PO$_4$ rhombic dodecahedrons and cubes, and they found that both of these samples exhibited higher photocatalytic activity.
than the microsized spherical $\text{Ag}_3\text{PO}_4$ particles [120]. Liang et al. synthesized hierarchical $\text{Ag}_3\text{PO}_4$ porous microcubes with enhanced photocatalytic property [123]. Wang et al. reported the synthesis of $\text{Ag}_3\text{PO}_4$ tetrapod microcrystals, and they demonstrated that $\text{Ag}_3\text{PO}_4$ tetrapod showed higher photocatalytic activity than the microsized spherical $\text{Ag}_3\text{PO}_4$ particles [126].

Another approach is to couple $\text{Ag}_3\text{PO}_4$ with other semiconductors, carbon materials, or noble metals to improve the photocatalytic activity, such as $\text{Ag}_3\text{PO}_4/\text{TiO}_2$ [127, 128], $\text{Ag}_3\text{PO}_4/\text{AgX}$ ($X = \text{Cl, Br, I}$) [129], $\text{Ag}_3\text{PO}_4/\text{BiOCl}$ [130], $\text{Ag}_3\text{PO}_4/\text{Fe}_2\text{O}_4$ [131], $\text{Ag}_3\text{PO}_4/\text{SnO}_2$ [132], $\text{Ag}_3\text{PO}_4/$ carbon quantum dots [133], $\text{Ag}_3\text{PO}_4$/reduced graphite oxide sheets [134], and $\text{Ag}_3\text{PO}_4$/Ag composites [135–137].

3.1. Crystal growth design

Manipulating the crystal structure will result in controlling the percentage of exposed facets on crystal surfaces and thus can lead to a dramatic change in reactivity, which has been widely investigated in sensing [138], electronics [139], magnetic memory devices [140], and catalysis [141]. It is widely accepted in catalysis that a higher surface energy leads to a more reactive surface. Therefore, the control of the exposed facets is one of the most available and efficient methods to obtain more active surface [142], which has been investigated recently to promote photocatalytic activity [83].

Herein, we controllably prepared $\text{Ag}_3\text{PO}_4$ crystals with various new morphologies (including branched, tetrapod, nanorod-shaped, and triangular-prism-shaped $\text{Ag}_3\text{PO}_4$ crystals) via a facile and efficient synthesis process in the solvent mixture of N,N dimethylformamide (DMF) and H$_2$O at room temperature (Fig. 6). The results indicate that the branched $\text{Ag}_3\text{PO}_4$ sample shows highly enhanced photocatalytic activity compared with other as-prepared $\text{Ag}_3\text{PO}_4$ samples, and the BET-specific surface area makes a greater contribution to the enhanced photocatalytic activity of as-prepared $\text{Ag}_3\text{PO}_4$ crystals [143].

3.2. $\text{Ag}_3\text{PO}_4$–graphene composite

The photocorrosion phenomenon of $\text{Ag}_3\text{PO}_4$ reduces the photocatalytic activity during the process. Up to now, some solutions have been put forward to improve its stability and photocatalytic properties under visible-light irradiation. Apart from its unique electronic properties [58], graphene has several other excellent attributes, such as the large theoretical specific surface area [57] and good chemical stability [59]. The large specific surface area of graphene facilitates the attachment of inorganic nanomaterials [144, 145]. Thus, the combination of graphene and $\text{Ag}_3\text{PO}_4$ could be a good choice to construct a stable and efficient photocatalyst composite.

In one of our works, $\text{Ag}_3\text{PO}_4$/reduced graphite oxide (RGO) nanocomposites were synthesized to enhance the visible-light photocatalytic activity and the stability of $\text{Ag}_3\text{PO}_4$. The results show that the graphene content obviously affects the photocatalytic activity of $\text{Ag}_3\text{PO}_4$/RGO nanocomposites [134]. As shown in Fig. 7, among a series of $\text{Ag}_3\text{PO}_4$/RGO, the $\text{Ag}_3\text{PO}_4$/2.1 wt % RGO shows the best photocatalytic activity despite the degradation of MB or Methyl Orange (MO) solution. In addition, the $\text{Ag}_3\text{PO}_4$/RGO is more stable than pure $\text{Ag}_3\text{PO}_4$ since the RGO
can be used as protective coatings that inhibit the photocorrosion of Ag$_3$PO$_4$. Thus, the Ag$_3$PO$_4$/RGO nanocomposites with excellent photocatalytic performance and enhanced stability can find promising applications in addressing environmental protection issues.

3.3. Ag$_3$PO$_4$-based heterostructure photocatalysts

Many narrow-band-gap metal oxides or chalcogenides have been coupled with Ag$_3$PO$_4$ photocatalyst to enhance its photocatalytic activity and/or improve its stability. In Yao’s work, Ag$_3$PO$_4$/TiO$_2$ has been synthesized via the in situ deposition of Ag$_3$PO$_4$ nanoparticles onto the TiO$_2$ surface, which facilitates electron-hole separation, thereby leading to enhanced photocatalytic activity [128].

In our work, the phenomenon of “self-corrosion” was first observed in the simple physical mixed Ag$_3$PO$_4$/TiO$_2$ compounds (Fig. 8). It is found that both self-corrosion and photocorrosion in Ag$_3$PO$_4$/TiO$_2$ compounds alter the chemical environment of Ag. The corrosion degree, however, is different due to a slight difference in the chemical environment of Ag. Furthermore, it is the strong adsorption capacity that determines the photocatalytic activities of...
Ag₃PO₄/TiO₂ compounds under UV light irradiation, which is almost independent of self-corrosion. In contrast, it is the amount of visible-light response of Ag₃PO₄ in Ag₃PO₄/TiO₂ compounds that mainly determines the photocatalytic activities under visible-light irradiation, which is highly relevant to self-corrosion [146].

Besides Ag₃PO₄/TiO₂ composite heterostructures, AgX/Ag₃PO₄ (X = Cl, Br, I) heterocrystals have also attracted much attention due to their excellent photocatalytic activity [129]. Bi and coworkers have reported that the AgX/Ag₃PO₄ (X = Cl, Br, I) heterocrystals embodied some advantages compared to the single Ag₃PO₄ and it is a more promising and fascinating visible-
light-driven photocatalyst than pure Ag₃PO₄ [129]. In their work, the AgBr/Ag₃PO₄ hybrid displayed much higher photocatalytic activity than single AgBr or Ag₃PO₄, as well as high stability under visible-light irradiation.

4. Long afterglow phosphor-assisted photocatalysts

Long afterglow phosphor-assisted novel photocatalysts become promising functional materials due to their effective utilization of solar light in practical applications of environmental purification. In this section, the recent development on TiO₂-based and Ag₃PO₄-based fluorescence photocatalyst composites with full-time active photocatalytic properties is reviewed.

The long afterglow phosphor (CaAl₂O₄:Eu, Nd) has a high luminescent brightness around 440 nm of wavelength, long afterglow time, good chemical stability, and low toxicity. The luminescent brightness around 440 nm can excite the visible-light-responsive nitrogen-doped titania (TiO₂–xNₓ). Therefore, TiO₂–xNₓ photocatalyst was expected to possess a novel photocatalytic property after coupling with CaAl₂O₄:Eu, Nd.)
In our research [147–149], the degradation of continuous NO gas was achieved by TiO$_2$–xN$_y$ surface-immobilized CaAl$_2$O$_4$:(Eu, Nd) microparticles (Fig. 9). The results show a persistent deNOx ability of the CaAl$_2$O$_4$:(Eu, Nd)/TiO$_2$–xN$_y$ composite. In another work [149], the persistent fluorescence-assisted photocatalysts of Ag$_3$PO$_4$ composites have been synthesized to expand the application of Ag$_3$PO$_4$ on dye decomposition day and night. Sr$_4$Al$_{14}$O$_{25}$:(Eu,Dy)/Ag$_3$PO$_4$ composites exhibited excellent photocatalytic activities for the Rhodamine B (RhB) decomposition reaction in the dark without additional light sources (Fig. 9c).

5. Challenges and perspectives

Photocatalysis appears to be a promising avenue to solve environmental and energy issues in the future. A variety of strategies, such as doping, coupling with graphene, precipitating with metal particles, crystal growth design, heterostructuring, were explored to enhance the efficiencies of photocatalytic activities. Besides modified TiO$_2$ and Ag$_3$PO$_4$, other visible-light-driven photocatalysts, including CdS, BiVO$_4$, Bi$_2$WO$_6$, and g-C$_3$N$_4$, even fluorescence-assisted photocatalyst composites, have attracted increased attention [147–156].
Although great progresses have been achieved, some challenges still exist to design high efficiency of photocatalytic systems. First, fundamental studies are essential to tackle the bottleneck problems in the field, including improved charge separation and transfer, promoted optical absorption, optimized band-gap position, lowered cost, and toxicity. Second, faceted photocatalysts remain a challenge and the development of surfactant-free synthesis routes is highly desirable, since most synthesis strategies involve the use of morphology-controlling agents that must be eventually removed in order to obtain clean facets. Finally, photostability of photocatalyst is and will continue to be a main challenge for practical applications. Therefore, new material design and innovative strategies for improving the efficiency and increasing the visible-light absorption of photocatalysts will be the key challenge and opportunity in this field.

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