Review

A Review on Pulsed Laser Preparation of Nanocomposites in Liquids and Their Applications in Photocatalysis

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Abstract: The purpose of photocatalysis is to realize the conversion between solar energy and chemical energy, and it is essential to develop a high-performance photocatalyst under visible-light irradiation. The conventional methods for photocatalyst preparation are mainly wet chemical routes, and abundant yields can be obtained. However, the products are not neat and accompanied by chemical groups and impurities, which are not beneficial for the enhancement of photocatalytic performance. In recent years, as a powerful tool for nanomaterial fabrication, pulsed laser heating in a liquid medium has been utilized to prepare a variety of nanocomposites. Products with synergistic effects and high crystallinity can be rapidly prepared under pulsed laser selective heating, which is beneficial for obtaining more effective photocatalytic performance. In this review, the typical characteristics of pulsed laser heating in liquids and their prepared nanocomposites for photocatalytic applications are summarized. This review not only highlights the innovative works of pulsed-laser-prepared nanocomposites in liquids for photocatalysis but also briefly introduces the specific challenges and prospects of this field.

Keywords: photocatalysis; nanocomposite; pulsed laser heating in liquid; challenges

1. Introduction

Just like the photosynthesis of plants in nature, photocatalysis is a technology to realize the conversion between solar energy and chemical energy; the crucial bridge is the photocatalyst. Since the pioneering work of water splitting under Xenon lamp irradiation on a titanium oxide (TiO2) electrode that was demonstrated by Fujishima and Honda in 1972 [1], semiconductor-based photocatalysts attracted remarkable attention [2–5] because the charge carriers from electron-hole pairs can be generated and migrated to the surface of semiconductors under the irradiation of light, which subsequently triggers a series of chemical reactions [6]. Nowadays, semiconductor-based photocatalysts have been utilized in many photocatalytic fields, such as the degradation of organic pollutants in the wastewater system, bacteria inactivation, and hydrogen/oxygen evolution from water splitting [3–5]. Particularly, the advanced oxidation process (AOP) plays a key role in photocatalytic reactions due to the high-activity hydroxyl radicals with powerful oxidation abilities that can be produced under the light-generated charge carriers and other reactive species. The contaminants can be removed by these intermediate radicals [7–9].

For many semiconductor materials, the high recombination rates of charge carriers also restrict their photocatalytic activities [2,10]. To overcome this issue, the raw semiconductor materials are modified to be nanocomposite photocatalysts by loading the assisted ligands on their surface [10–12]. According to this synergistic effect, the bandgap can be narrowed, and the visible-light response can be effectively enhanced to boost the photocatalytic activities. For instance, M. A. Ahmed et al. constructed a hybrid S-scheme BiVO4/g-C3N4 nanocomposite by dissolving bismuth nitrate in ethylene glycol and loading it on the surface
of g-C$_3$N$_4$ nanoparticles using the ultrasonic hydrothermal method. The photocatalytic hydrogen evolution rate of the prepared nanocomposite was enhanced ten times than that of bare g-C$_3$N$_4$ with a maximum value of 6.8 mmol g$^{-1}$ h$^{-1}$ (at 10 wt.% BiVO$_3$) [10]. F. A. Harraz et al. designed a ternary photocatalytic system of Au@Ppy-C/g-C$_3$N$_4$ by decorating the Au nanoparticles on the as-prepared mixture of Ppy-C/g-C$_3$N$_4$ via a highly effective hydrothermal method. The photocatalytic performance of the created ternary photocatalyst was enhanced 2.91 times more than the bare g-C$_3$N$_4$ towards the degradation of the target insecticide imidacloprid. A 96% removal rate can be obtained under the illumination of visible light within 25 min [11]. E. R. Nagarajan et al. prepared a dibasic CoWO$_4$/ZnO nanocomposite by using the straightforward hydrothermal method. The products exhibited excellent photocatalytic activities for the elimination of the ciprofloxacin drug and methylene blue dye under visible-light irradiation. Respectively, a 93.13% and 98.65% efficiency can be obtained, which are higher than those of pure components [12].

In general, the conventional methods for nanocomposite preparation are mainly wet chemical routes, such as the abovementioned hydrothermal. These approaches can be obtained in abundant yields, but the prepared products are not neat. Some chemical groups and impurities are unavoidable as surface obstacles for the fast migration of charge carriers [13–17]. Additionally, due to the uncontrolled nature of the reactions associated with the precursor materials, required thermal treatments of the products may lead to several defects, eventually providing a recombination center for charge carriers [18–20]. Hence, developing a simple and innovative approach to realizing the preparation of nanocomposites with neat and high crystallinity is significant. In recent years, as a powerful tool for nanomaterials fabrication, pulsed laser heating (PLH) in a liquid medium has been utilized to prepare a variety of colloids and nanocomposites [21,22]. Different from the conventional chemical approach based on the interaction between laser energy and nanoparticles, the photothermal effect can be rapidly transferred (typically in the magnitudes of ns, ps, or fs) so that the components of precursor materials can be well-composited without any chemical groups and impurities. This high-quality nanocomposite utilized as a photocatalyst is beneficial for the fast migration of charge carriers and enhancement of photocatalytic performance [19,22].

In previous reports, there were several reviews concentrated on this field. For example, A. M. Müller et al. integrally summarized the state of the art, laser-made electrocatalysts for water/organic oxidation, oxygen/nitrogen/carbon dioxide reduction, and hydrogen evolution [22]. M. Y. Choi et al. briefly presented a research overview of the pulsed laser synthesis of metal nanoparticles, oxides, nonoxides, and carbon-based materials for photo/electrocatalytic applications [23]. E. Fazio et al. gave a brief overview of pulsed laser synthesis of nanocolloids for biomedicine, catalysis, and sensing [24]. Y. Chen et al. illustrated an overview of the high-power instant synthesis of carbon nanomaterials and nanocomposites for catalyst, energy storage, and conversion applications [25]. In this review, we mainly focus on the summary of pulsed laser synthesis of semiconductor-based nanocomposites for photocatalysis and discuss the specific challenges and prospects of this field. This review provides a summary of the dual investigations of photocatalysis and pulsed laser preparation of nanocomposites.

2. Pulsed Laser Heating in a Liquid Medium

The earliest study on pulsed laser heating in a liquid medium dates back to the V. G. Bhide group in 1987. They used a ruby-pulsed laser of 694 nm to ablate a pure iron target in water to form iron oxides with metastable phases [26]. This seminal work created a new field for bulk materials and laser interactions. In the early 1990s, this method was also utilized to synthesize colloidal nanoparticles in solutions by A. Fojtik and A. Henglein [27]. Since then, a vast number of relevant works, such as metal, alloys, oxide, hydroxide, ceramics, semiconductors, compounds, and carbon nanoparticles, were successfully prepared by this similar approach. To date, pulsed laser heating in a liquid medium has been devoted to being a scalable and versatile technique for nanomaterials fabrication [28,29].
2.1. Classification of Pulsed Laser Synthesis of Colloids

Colloidal nanoparticles, such as nanospheres and nanocrystals, are widely used as atomic clusters for the self-assembly and modification of functional materials [3]. Hence, the preparation of colloids with different particle sizes and morphological characteristics is vitally important, while the average particle size is mainly determined by the ways of pulsed laser irradiation. Figure 1 shows the laser synthesis of colloids is a mixture of photonics and nanotechnology (see Figure 1A). The specific patterns can be divided into three aspects, i.e., pulse laser ablation in liquids (PLAL, Figure 1B), pulsed laser fragmentation in liquids (PLFL, Figure 1C), and pulsed laser melting in liquids (PLML, Figure 1D).

![Figure 1. Classification scheme of pulsed laser heating in liquid medium. (A) Pulsed laser synthesis of colloids. (B) Pulsed laser ablation in liquids (PLAL). (C) Pulsed laser fragmentation in liquids (PLFL). (D) Pulsed laser melting in liquids (PLML). Reprinted with permission from Ref. [29].](image)

1. PLAL is a technique using a focused or high-power density laser beam (exceeding $1 \times 10^9$ W/cm$^2$) to generate the cavitation bubble of plasma from the surface of a bulk target; the plasma can be constrained and rapidly quenched by the liquid medium to form the desired nanomaterials [29]. During the formation of nanoparticles, because the laser fluence is a gradient form, it induces a wide size distribution, so this method is only suitable for the preliminary fabrication of nanoparticles.

2. PLFL is derived from PLAL, the laser energy directly interacting with the dispersed colloids in a liquid medium, and the initial particles with a bigger size can be fragmented into smaller nanoparticles under high-density laser irradiation. This improvement was first proposed by the P. V. Kamat group [30]. They used a high-density laser beam (355 nm, 6 ns) to irradiate the colloidal silver particles of bigger sizes (40–60 nm), and the colloids were transformed into smaller nanoparticles (5–20 nm) under the heat effect of the laser. In this way, particle size can be tailored by the tunability of laser fluence, but the morphology and phase may also be shifted under such a powerful laser density [29].

3. PLML is contrary to PLFL, the laser beam is unfocused and the laser fluence is relatively modest. The smaller colloids can be melted and composited into a bigger size. This method was first demonstrated by the N. Koshizaki group [31]. The raw CuO nanoparticles with an average size of 34 nm were transformed into 300 nm under
pulsed laser irradiation in acetone (355 nm, 66 mJ·pulse<sup>-1</sup>·cm<sup>-2</sup>, 30 min). Using this method, the irregularly shaped particles can also be spherically reshaped.

2.2. Characteristics of Pulsed Laser Synthesis of Colloids

For pulsed laser synthesis technology introduced above, one of the most unique features is their applications for the preparation of colloidal nanospheres [29]. Based on the rapid interaction between laser energy and colloids in liquids, the colloids can be well melted and even exploded into particle fragments because the pulsed laser heating process is instantaneous and discontinuous (mainly determined by the pulse width and repetitive rate). The colloids after one pulse of heating can be subsequently quenched (about 10<sup>-6</sup>–10<sup>-4</sup> s) until the next pulse arrives. This accumulation of numerous individual heating pulses greatly inhibits the anisotropic growth of the particles, which is beneficial for the formation of spherical geometry. Additionally, the laser absorbance of colloids is usually much higher than that of liquids. Colloids can be selectively heated under laser irradiation in liquids while the laser effect on liquids can be ignored. Hence, colloidal spheres can be effectively prepared [3, 31]. To date, there is a variety of microspheres and nanospheres successfully prepared by pulsed laser heating in liquids (Figure 2), such as noble metals (Au, Ag), semiconductor materials (Ge, Si), metal oxides (TiO<sub>2</sub>, ZnO, CuO), and some carbides (B<sub>4</sub>C, CQDs), demonstrating its diversity for potential applications in functional materials [32–40].

![Figure 2](image-url)

Figure 2. Preparation of colloidal nanospheres by pulsed laser heating in liquid medium. (A) Au. (B) Ag. (C) Ge. (D) Si. (E) B<sub>4</sub>C. (F) TiO<sub>2</sub>. (G) Carbon Quantum Dots (CQDs). (H) ZnO. (I) CuO. Reprinted with permission from Refs. [32–40].

Another significant feature of pulse heating colloids in liquids is size regulation. The geometric size of colloidal particles is determined by the laser fluence due to the phase states of particles that can be affected by the powerful laser energy. Just like the ways of PLML and PLFL, size-tailored colloids are fabricated by the melting (at lower laser energy) or pneumatolysis (higher laser energy) of raw particles. Their melting point or boiling point can be exceeded by the varied heat effect of lasers. In particular, the phase transition can be
described by the thermodynamic “heating-melting-evaporation” model (HME), which was first proposed by the A. Takami group in 1999 [41]. After that, it was further developed by the A. Pyatenko group [42]. For example, as shown in Figure 3, the phase diagrams of Ag particles under pulsed laser heating at 355 nm and 532 nm were calculated using the HME model, respectively. For raw Ag particles, two possible phase-transition processes were successively performed. (1) From a solid state to the liquid phase if the initial solid particles (marked as symbol “S”) were heated by a relatively lower laser fluence. With the increase in laser energy, when the heat effect was over the melting point (about 961.78 °C for Ag), the surface of the Ag particles started melting and exhibiting a mixture of solid and liquid phases (S + L) before melting into the liquid phase (L) completely. This process usually occurs in the form of PLML. (2) From the liquid to the gaseous phase: With the laser energy continuously increasing, the boiling point (about 2212 °C for Ag) was reached, so the molten-state Ag gradually transferred into the gaseous phase (L + G) and quenched in the liquid medium. Hence, compared with the raw particles, the particle size of plasma sharply decreased, which is properly corresponding to the process of PLFL.

In the pulsed laser synthesis of colloids, several parameters, such as the laser conditions, targets/liquid chemistry, and the initial mass ratios of precursors, are crucial for the phase and crystallinity of the products [22,29].

Firstly, the colloids dispersed in the liquid medium can be affected by different laser parameters, including the pulsed repetition rate, pulsed width, output wavelength, laser fluence, irradiation time, and irradiation ways (focused or unfocused). For example, N. Koshizaki et al. used a 355 nm unfocused pulsed laser irradiation of colloidal nanoparticles of raw CuO in acetone which performed a phase-controlled CuO nanosphere under different laser fluences. When laser fluence is below 33 mJ pulse\(^{-1}\) cm\(^{-2}\), the phases of products are always CuO. Once it increases to 50 mJ pulse\(^{-1}\) cm\(^{-2}\), the phases of products appear in the form of a mixture of CuO/Cu\(_2\)O. Subsequently, when laser fluence is exceeds 132 mJ pulse\(^{-1}\) cm\(^{-2}\), the phase of products is metal copper. This indicates that the phase of colloids can be controlled by the tunability of laser fluence because the phase-transformation of colloids can be achieved under different temperatures, which is directly related to the laser fluence [31]. X. Li et al. presented a phase-controlled titanium dioxide (TiO\(_2\)) microsphere under different laser wavelengths. They used an unfocused pulsed laser irradiation of anatase TiO\(_2\) in ethanol solvent. The single-crystalline rutile TiO\(_2\) spheres amorphous and polycrystalline anatase TiO\(_2\) spheres can be selectively prepared by outputting the laser wavelengths of 355 nm and 1064 nm, respectively [37]. Because the laser absorbance of anatase TiO\(_2\) was different between 355 nm and 1064 nm, the 355 nm laser can be absorbed more effectively, and thus the different products were formed.
Additionally, colloids under laser irradiation may also be reacted with a liquid medium and produce special compounds. This process is usually called “reactive laser ablation in liquids (RLAL)”. Due to the laser ablated species with high temperatures can be quenched and induced decomposition in the liquid medium [29]. For instance, Y. Ishikawa et al. prepared a boron carbide (B4C) submicron-particle with pulsed laser irradiation of boron particles in ethyl acetate under atmospheric pressure and room temperature. The boron particles were melted by laser energy and compounded with carbon species from ethyl acetate to form B4C particles [36]. During the pulsed laser-focused ablation of colloids (355 nm, 10 Hz, 7 ns), the ethyl acetate was decomposed and encapsulated in a graphite surface of B4C submicron spheres.

3. Pulsed Laser Preparation of Nanocomposites for Photocatalysis

3.1. Pulsed Laser Preparation of Nanocomposites

The diversity of pulsed laser synthesis of colloids provides a facile approach for the preparation of nanocomposites with multivariate components; the different elements can be rapidly composited with the heat effect of laser energy. The synthesis of a nanocomposite is determined by the precursor materials; these multivariate precursors can be welded or modified by the laser effect. Figure 4 shows a typical schematic diagram of the whole procedure for pulsed laser synthesis of nanocomposites, and four steps are included. (1) Sample preparation: The multivariate precursors (at least two components) are dispersed into the liquid solvent to form the colloidal suspension. (2) Homogeneous dispersion: The colloidal suspension with nanoparticles is uniformly dispersed by an ultrasonic oscillator to overcome the aggregation effect of nanoparticles. (3) Pulsed laser irradiation: The homogeneously dispersed colloidal nanoparticles are selectively heated under a laser beam of unfocused irradiation, and the suspension is stirred by a magnetic rotor to prevent the gravitational settlement of nanoparticles. (4) Products separation: The laser-prepared nanocomposites can be obtained by evaporation of the solvent and used for photocatalysis or other significant fields [43].

Figure 4. A schematic diagram of the whole procedure for pulsed laser synthesis of the nanocomposite. (A) Sample preparation. (B) Homogeneous dispersion. (C) Pulsed laser irradiation. (D) Products separation. Reprinted with permission from Ref. [43].

Using this innovative method, various nanocomposites can be rapidly prepared. For instance, Figure 5 illustrates the energy-dispersive X-ray spectroscopy (EDS) mapping image of Ag/TiO2 nanocomposites prepared by pulsed laser irradiation of raw Ag and TiO2 nanomaterials mixed in ethanol solvent (532 nm, 478 mJ-pulse−1 · cm−2, 15 min) [44]. The Ag element was sandwiched in between the components of TiO2 during the laser heating process because the particle size of Ag was much less than the scalebar (50 nm), according to the HME model described by the A. Pyatenko group (see Figure 3B). Their phases under this laser fluence are a mixture of liquid and gaseous states (L + G) so that Ag nanoparticles could be prepared and welded with TiO2 nanomaterials under liquid
quenching. Herein, only Ag nanoparticles were melted and loaded on the surface of TiO₂ because the laser wavelength of 532 nm could be selectively absorbed by Ag nanoparticles, and the laser effect on TiO₂ was weak due to its wide bandgap restriction (<420 nm). In addition, another way to the compound is the melting of all precursors. When the laser energy was effectively absorbed by all precursors, their lattice units were compatible on a nanoscale and formed multivariate nanocomposites. Moreover, this method can also be utilized to prepare other nanocomposites, such as hybrid liquid metals and hollow or core-shell spheres [19,45], demonstrating the effectiveness of this technology.

Figure 5. The energy-dispersive X-ray spectroscopy mapping image of laser-prepared Ag/TiO₂ nanocomposites. (A) High-angle annular dark field (HAADF). (B) Mixed. (C) O element. (D) Ti element. (E) Ag element. Reprinted with permission from Ref. [44].

3.2. Laser-Prepared Nanocomposites for Photocatalysis

Contrary to the wet chemical routes, for the pulsed laser preparation of nanocomposites, various toxic chemical reagents and complicated reaction conditions were not necessary. During sample preparation, the liquids only worked as dispersant and quenching mediums for precursors, and the nanocomposite could be rapidly prepared by pulsed laser heating in a short timescale. Hence, the chemical groups or impurities were hardly involved in the products; these nanocomposites with neat and high crystallinity utilized as photocatalysts were beneficial for the fast migration of light-generated charge carriers, and the related photocatalytic performances can be effectively enhanced [22]. To date, a vast number of nanocomposites prepared by pulsed laser heating in liquids were applied in different photocatalytic fields and illustrated desirable performances [46–49].

3.2.1. Degradation of Organic Pollutants

Organic pollutants, such as wasted dyes and drugs dissolved in the water system, are extremely harmful to the environment. Photocatalytic removal of them via the AOP technique is considered a promising approach, and laser-prepared nanocomposites with high...
quality are widely utilized as photocatalysts. For instance, X. Li and K. Liu et al. prepared a core-shell Ag/ZnO nanocomposite by pulsed laser selective irradiation (532 nm, 10 Hz, 8 ns) in ethanol solvent. The raw Ag nanoparticles after laser heating were transformed into nanospheres and anchored on the surface of ZnO. For the degradation of methylene blue dyes under a 250 W ultraviolet lamp illumination (the main maximum wavelength is 365 nm), the photocatalytic performance of the prepared nanocomposites was enhanced two times than neat ZnO with a higher rate constant of 0.04 min$^{-1}$ [43]. T. T. Kim et al. presented a mixed CeO$_2$/Ce$_2$O$_3$ nanohybrid using a 355 nm pulsed laser irradiation of CeO$_2$ nanoparticles in deionized water. The ultraviolet laser irradiation induced the formation of oxygen vacancy defects in CeO$_2$, which promoted its redox ability. The photocatalytic activities of pure CeO$_2$ and the prepared CeO$_2$/Ce$_2$O$_3$ were evaluated by the degradation of the methylene blue aqueous solutions, respectively. The calculated reaction rate constant of CeO$_2$/Ce$_2$O$_3$ was $2.16 \times 10^{-3}$ min$^{-1}$, which was 1.5 times higher than that of pure CeO$_2$ ($1.46 \times 10^{-3}$ min$^{-1}$). The enhanced activity of mixed-phase CeO$_2$/Ce$_2$O$_3$ originated from the effective separation of photogenerated electron-hole pairs within the composite through a Z-scheme pattern. The electrons in the conduction band of Ce$_2$O$_3$ and holes in the valence band of CeO$_2$ could react with methylene blue in solution and degraded pollutants [46].

M. A. Gondal et al. synthesized a series of TiO$_2$/CdS nanocomposites by using a 532 nm pulsed laser ablation in an aqueous solution of hydrogen peroxide. The nanocomposite with a 10% percentage weight of CdS in anatase TiO$_2$ showed the best performance for the photocatalytic degradation of methyl orange (MO) dyes under ultraviolet-visible-light irradiation. Compared with the pure TiO$_2$, the percentage of removed MO that was degraded by this nanocomposite was enhanced from 58% to 82% within 36 min with a higher reaction rate constant of 0.0587 min$^{-1}$ [47]. Furthermore, Y. H. Chu et al. fabricated a complex oxide-noble metal hybrid heterodimer of Au-SrTiO$_3$ nanocomposite by using a 248 nm KrF excimer laser ablation of SrTiO$_3$ ceramic targets in an aqueous gold nanoparticle colloid. For photocatalytic degradation of rhodamine B (RhB) and MO under the illumination of visible light ($\lambda > 420$ nm), this heterodimer exhibited a more active degradation ability for both RhB and MO than that of pure SrTiO$_3$ and pure TiO$_2$ aerosol, respectively [48]. M. Y. Choi et al. synthesized a series of ZnO/Au/Pd nanocomposites by pulsed laser ablation assisted with subsequent photo deposition technology. For the photocatalytic degradation of methylene blue dyes under visible light illumination ($\lambda > 420$ nm), the photocatalytic activity of the optimized nanocomposite was enhanced 5.4 times than that of pure ZnO. The reaction rate constant of 0.0145 min$^{-1}$ was obtained. The enhanced mechanism was attributed to the coupling effect of the noble metals. The surface plasmon resonance (SPR) effect of Au for enhanced visible light absorption was not only displayed, but also the Pd nanocomposite served as an electron storage chamber enabling the separation of charge carriers [49].

For the multivariate nanocomposites fabricated by pulsed laser heating in liquid, H. J. Shipley et al. prepared a multilayered TiO$_2$-ZSM5 (Zeolite)-MoS$_2$ nanocomposite by pulsed laser ablation in an aqueous solution of MoS$_2$ (1064 nm, 20 Hz, 3.6 ns) assisted with a microwave treatment at high temperature and pressure. The products utilized as photocatalysts displayed excellent performance towards arsenite photooxidation under ultraviolet lamp irradiation (365 nm, 115 V). An approximate 100% photo-conversion rate was obtained from arsenite to arsenate within 180 min, which was stronger than either pure TiO$_2$ nanoparticles (75%) or TiO$_2$-ZSM5 (90%). The enhanced mechanism was attributed to the heterojunction formation of a closed-lattice neighborhood among TiO$_2$, ZSM5, and MoS$_2$; the insertion of a vertically aligned MoS$_2$ layer promoted the in-plane electrical conductivity so that the electrons could migrate more efficiently. Meanwhile, the vertically aligned MoS$_2$ layers with a higher in-plane electrical conductivity were also beneficial for the efficient separation of photo-induced electron-hole pairs via an inner electric field between interfaces of p-type MoS$_2$ and n-type TiO$_2$ [50]. In addition, based on this two-step strategy, the same group also synthesized a hybrid nanocomposite of HBITiO$_2$/RBIHM-MoS$_2$ by decorating the vertically aligned MoS$_2$ nanosheets on the mixture of rutile-based inorganic hollow
microspheres (RBIHM) and hydrogenated black TiO$_2$ (HBTiO$_2$) nanoparticles via a similar approach. Particularly, this nanocomposite also displayed higher photocatalytic activity for arsenite oxidation under visible light illumination ($\lambda > 420$ nm). A 96.6% photooxidation efficiency was obtained, which was enhanced 1.37 times compared with the mixture of HBTiO$_2$/RBIHM (70.3%). The superiority of the nanocomposite was attributed to the introduction of MoS$_2$ nanosheets, which extended the visible light absorption range along with the efficient charge carriers’ accumulation and separation in the heterojunctions between the vertically aligned MoS$_2$ nanosheets and HBTiO$_2$/RBIHM [51].

For a nanocomposite that can be used for photocatalytic removal of organic hazardous materials, many innovative works were also presented. For instance, T. K. Kim et al. fabricated a CeO$_2$/Pd nanocomposite by pulsed laser ablation of a Pd metal plate in an aqueous solution containing CeO$_2$ nanosheets (1064 nm, 10 Hz, 3–6 ns). Towards the catalytic reduction of 4-nitrophenol to 4-aminophenol in the presence of sodium borohydride (NaBH$_4$) at room temperature, the nanocomposite showed much higher photocatalytic activity than bare CeO$_2$ nanostructures. The content of 4-nitrophenol completely vanished by CeO$_2$/Pd nanocomposite within 60 s while the reduction rate of pure CeO$_2$ was only 68% within 80 min. The improved catalytic performance of this nanocomposite originated from the chemical interaction between the CeO$_2$ and Pd nanomaterials, which increased the rate of electron transfer across the interface of the CeO$_2$-Pd hybrid and thus improved the local electron concentration for catalytic conversion. Meanwhile, this fast electron transfer, in turn, can be efficiently achieved from BH$_4^-$ (donor) to 4-nitrophenol (acceptor) via hybrid nanocatalysts [32]. The same group also produced core-shell Pd/C composited nanospheres by pulsed laser ablation of a target Pd foil in acetonitrile. Towards the reduction of nitrobenzene to aniline upon the addition of NaBH$_4$, the catalytic performance of the Pd/C nanocomposites and Pd counterparts was tested, respectively. The core-shell nanocomposite also displayed a higher conversion rate than that of bare Pd, about 3.03 times enhanced. Specifically for the Pd nanostructures, 70% nitrobenzene was reduced within 11 min, and the reaction rate constant was 0.079 min$^{-1}$. While for Pd/C nanocomposites, 72% nitrobenzene was reduced within 6 min, and the corresponding rate constant was 0.24 min$^{-1}$. The enhanced catalytic mechanism of the core-shell Pd/C nanocomposites was attributed to the encapsulating of graphitized C layers on the surface of the Pd nanostructures. The graphitized C layers along with a high specific surface area induced the high adsorption rate of nitrobenzene into the Pd nanostructures via the stacking interaction between the graphitized C layers. The efficient electron transfer from the graphitized C layers to the Pd nanostructures increased the electron concentration and, in turn, facilitated the electron transfer from the BH$_4^-$ to nitrobenzene through the Pd nanostructures, resulting in a highly efficient catalytic reduction activity [53].

C. Wang and R. Yang et al. rapidly prepared a ligand-free Rh/GO nanocomposite by ultrafine rhodium (Rh, size: 1.8 ± 0.4 nm) nanoparticles decorated on the surface of graphene oxide (GO) nanosheets. As illustrated in Figure 6, the Rh nanoparticles were generated by the pulsed laser ablation of a target in ethanol (532 nm, 10 Hz, 10 ns) and composited with the GO sheets via electrostatic interaction. Towards the reduction of 4-nitrophenol (4-NP) in presence of NaBH$_4$, the catalytic performance of the prepared Rh/GO nanocomposite was evaluated. A remarkable conversion rate of 100% was obtained. The kinetic rate constant was $7.62 \times 10^{-3}$ s$^{-1}$, which was 27 times higher than commercial Rh/C catalysts. This prepared Rh/GO nanocomposite also showed excellent stability of catalytic activity after five circulations due to the pulsed-laser-ablated small size of Rh nanoparticles that were uniformly dispersed on the GO nanosheets. Particularly, the remarkable high catalytic activity of the Rh/GO nanocomposite was also ascribed to the $\pi-\pi$ stacking interaction between 4-NP and GO [54].
M. Ghiaci et al. prepared a composite Au-Pd/g-C$_3$N$_4$ via the pulsed laser ablation of Au and Pd nanocomposites in polyvinylpyrrolidone (PVP) aqueous solution and decorating on the surface of g-C$_3$N$_4$ by using the assisted chemical approach. After optimization, the hybrid catalyst showed the capability of activating hydrogen peroxide and superiority for oxidization of benzene to phenol under visible-light irradiation at 50 °C. The benzene conversion rate was as high as 26% at a phenol selectivity of 100%, giving no dehydroxylated byproducts [55]. The same group also synthesized a named Cu-time/a-C$_3$N$_4$ nanocomposite by pulsed laser ablation in liquids. Under different laser irradiation times, such as 20 min, 40 min, and 60 min, the nanocomposites were denoted as Cu-20 min/a-C$_3$N$_4$, Cu-40 min/a-C$_3$N$_4$, and Cu-60 min/a-C$_3$N$_4$, respectively. Thereinto, the copper metallic nanoparticles were successfully immobilized on the surface of amorphous carbon nitride (a-C$_3$N$_4$) at different times of laser irradiation. In particular, the Cu-40 min/a-C$_3$N$_4$ catalyst exhibited significant photocatalytic activity for the oxidation of cyclohexane to the ketone-alcohol mixture (KA oil) under visible-light irradiation (λ > 420 nm). An 88% conversion rate was reached at a selectivity of 95%. The superiority of this Cu-time/a-C$_3$N$_4$ nanocomposite was attributed to the construction of amorphous carbon nitride and amorphous carbon heterojunction, which increased the lifetime of the photoexcited state and thus boosted the light absorption ability. Meanwhile, the narrowing of the band gap was also beneficial for the excitation of charge carriers under visible light irradiation. The electrons located on the valence band of a-C$_3$N$_4$ can be easily transferred to the conduction band and then trigger the subsequent redox reactions [56]. Moreover, the pulsed laser ablated copper nanoparticles dispersed on the matrix of the composite during carbonization also promoted visible light absorption via the SPR effect [3,43].

3.2.2. Photocatalytic Bacteria Inactivation

The bacteria breeding in humans and animals has caused a variety of diseases, such as pneumonia, endocarditis, diarrhea, and skin and soft tissue infections (SSTIs) [57,58]. In recent years, with the long-term abuse of commercial antibiotics (such as methicillin, vancomycin, and daptomycin), drug-resistance has appeared. Hence, developing a substitute
for these conventional antibacterial agents is eagerly required. Functional nanomaterials with enzyme-like characteristics have been taken into consideration and gradually prepared in many unique ways, including pulsed laser heating in liquids. For instance, X. Li et al. prepared a hybrid Ag/CeO$_2$ nanocomposite by pulsed laser unfocused irradiation in liquid (532 nm, 10 Hz, 8 ns), and the photocatalytic activity for inactivation of Staphylococcus aureus (S. aureus) under visible light illumination ($\lambda > 420$ nm, 150 W) was evaluated. An 82.4% sterilization rate was obtained which was 2.93 and 2.99 times higher than those of pure Ag and pure CeO$_2$ counterparts. A possible enhanced mechanism was proposed from the production of superoxide dismutase (SOD) mimetic activity. As shown in Figure 7, for a pure CeO$_2$ nanoparticle, its crystal structure was a face-centered-cubic fluorite type; the Ce$^{4+}$ and O$^{2-}$ ions were located on the octahedral interstitial and tetrahedral interstitial sites, respectively. With the formation and migration of inner oxygen vacancies, the oxygen atom accompanied by two valence electrons was released; these electrons were captured by Ce$^{4+}$ ions and oxygen molecules to produce Ce$^{3+}$ ions and superoxide anions. With the increased ratio of Ce$^{3+}$/Ce$^{4+}$, the SOD-like activity was displayed via a unique route [59]. For the prepared Ag/CeO$_2$ nanocomposite, the introduction of the Ag component not only boosted the concentration of Ag$^+$ ions but also provided sufficient electrons to produce more Ce$^{3+}$ ions, and thus a generally enhanced SOD-like activity was displayed [60].

Figure 7. The schematic diagram of crystal structures of pure CeO$_2$ (A) and the prepared Ag/CeO$_2$ nanocomposite (B), respectively. Reprinted with permission from Ref. [60].

M. A. Gondal et al. synthesized a CdS@g-C$_3$N$_4$ nanocomposite using a 532 nm pulsed laser ablation of a mixture of as-prepared g-C$_3$N$_4$ powders and commercially procured CdS powders in deionized water. The CdS nanoparticles were anchored on the surface of g-C$_3$N$_4$ nanosheets. Experimental investigation showed that the CdS@g-C$_3$N$_4$ nanocomposite exhibited a higher antibacterial activity than pure g-C$_3$N$_4$ towards the photocatalytic deactivation of Escherichia coli (E. coli) under visible light illumination. A remarkable 129% increase rate of the photocatalytic degradation constant was obtained, increased from 0.07 min$^{-1}$ to 0.16 min$^{-1}$, and the total depletion time of E. coli was decreased from 156 min to 67 min. The enhanced mechanism originated from the formation of heterojunction between CdS and g-C$_3$N$_4$; the authoring of CdS shifted the relative energy positions of g-C$_3$N$_4$ so that the electrons on the conduction band of g-C$_3$N$_4$ could be transferred to CdS, and the holes on the valence band of CdS could be migrated to g-C$_3$N$_4$, which boosted
the visible light absorption and reduced the recombination rate of photogenerated charge carriers [61]. In addition, J. Li et al. prepared a series of Ag/TiO\textsubscript{2} nanocomposites by a pulsed-laser-irradiated mixture of raw Ag/TiO\textsubscript{2} nanoparticles in liquids under different initial mass ratios; Ag nanospheres of 20–30 nm size were generated and precisely loaded on the surface of TiO\textsubscript{2} nanocrystalline. In particular, the prepared nanocomposites with an initial mass ratio of Ag and TiO\textsubscript{2} at 1:4 performed the best for the photocatalytic sterilization of \textit{S. aureus} under visible light illumination (\(\lambda > 420\) nm, 150 W). Figure 8 shows the antibacterial effect of this laser-prepared Ag/TiO\textsubscript{2} nanocomposite and comparison with different antibacterial agents, including counterparts of pure Ag, pure TiO\textsubscript{2}, and their mixture under laser or not. A photocatalytic sterilization rate as high as 85.7\% was achieved, which was 2.13 and 2.05 times higher than pure Ag and pure TiO\textsubscript{2}, respectively. This improvement was attributed to the coupled SPR effect of Ag nanoparticles, which boosted visible light harvesting and generated more charge carriers to produce abundant reactive oxygen species for the advanced oxidation process of bacterial inactivation [44].

![Figure 8](image_url)

\textbf{Figure 8.} The antibacterial effects of different antibacterial agents under visible light or not. (A) The liquid broth plates supported the growth of bacteria. (B) The comparison of bacterial colony counts under visible light or not. (C) The corresponding sterilization rate over different antibacterial agents. Reprinted with permission from Ref. [44].

3.2.3. Photocatalysts for Water Splitting

Photocatalytic overall water splitting under solar light stimulation was a promising strategy to produce hydrogen and oxygen because hydrogen and oxygen can be generated on the surfaces of photoanodes and photocathodes, respectively. To date, due to the reduction and oxidation reactions that hardly occur in a single material, pulsed laser synthesis technology in liquids has been utilized to prepare a variety of nanocomposites for the photocatalytic evolution of gaseous hydrogen and oxygen fuels. For instance, A. M. Müller et al. developed a mixed-metal of nickel–iron (Ni/Fe) layerd with double hydroxide nanoparticles by pulsed laser ablation in liquids. For water oxidation, higher activity was observed as the amount of Fe decreased to 22\% after 30 min [62]. On this basis, J. K. Song and T. T. Kim et al. designed a double hydroxide CdS/NiFe layered nanocomposite by liquid-phase pulsed laser ablation and a hydrothermal method. First, they used a 1064 nm, 10 Hz, 3–6 ns Nd: YAG laser to irradiate the Ni foil fixed in an FeCl\textsubscript{3}
aqueous solution to prepare NiFe nanosheets. After that, CdS nanorods were synthesized by a hydrothermal method using cadmium acetate and thiourea as the precursor materials and composites with the as-prepared NiFe nanosheets via ultrasonication. The prepared CdS/NiFe nanocomposites were obtained through ultrasonication. For the photocatalytic hydrogen production test, the resultant CdS/Co-MoS$_2$ nanocomposite consisted of 1 wt.% Co nanocrystals dispersed by hydrothermal treatments, respectively. The overall CdS/Co-MoS$_2$ nanocomposites exhibited a higher performance for photocatalytic hydrogen evolution rate of 275 mmol h$^{-1}$ g$^{-1}$ at 42 wt.% of NiFe was obtained, which was 30 times higher than pure CdS nanorods [63].

Meanwhile, M. Lim and T. K. Kim et al. also prepared a series of CdS/Co-MoS$_2$ nanocomposites by pulsed laser ablation in liquids assisted with hydrothermal treatment. As shown in Figure 9, the cobalt (Co) nanocrystals with different sizes were fabricated by the pulsed laser ablation of Co metal plates in deionized water (532 nm, 10 Hz, 3–6 ns) under different laser fluences. The layered MoS$_2$ nanostructures and CdS nanorods were synthesized by hydrothermal treatments, respectively. The overall CdS/Co-MoS$_2$ nanocomposites were obtained through ultrasonication. For the photocatalytic hydrogen production test, the resultant CdS/Co-MoS$_2$ nanocomposite consisted of 1 wt.% Co nanocrystals displayed a maximum hydrogen evolution rate of 275 mmol h$^{-1}$ g$^{-1}$ and outstanding stability under a 150 W Xe lamp irradiation, which was 114.5 and 2.23 times higher than those of bare CdS and CdS/MoS$_2$ nanostructures, respectively [64].

Figures 9. Schematic diagram of the synthesis of CdS/Co-MoS$_2$ nanocomposites. (1) The size-controlled Co nanocrystal was prepared by pulsed laser ablation of target metal plate in liquid under different laser fluences. (2) Formation of bulk MoS$_2$ nanosheets through hydrothermal synthesis method. (3) Preparation of few-layer Co-MoS$_2$ nanocomposites by ultrasonication. (4) Integration of ultrathin Co-MoS$_2$ nanosheets on as-prepared CdS nanorods by ultrasonication and longtime magnetic stirring to generate interfacial contact between CdS and Co-MoS$_2$ nanostructures. Reprinted with permission from Ref. [64].

M. A. Gondal et al. fabricated a series of polymeric-inorganic nanocomposites by pulsed laser ablation of SiC and g-C$_3$N$_4$ mixtures in deionized water (532 nm, 10 Hz, 9 ns). Therein, the SiC nanoparticle was anchored on the surface of the as-prepared g-C$_3$N$_4$ polymer network. The prepared SiC/g-C$_3$N$_4$-10% nanocomposites exhibited a higher photocatalytic hydrogen production rate than bare g-C$_3$N$_4$ for the water oxidation reaction under visible light illumination ($\lambda > 420$ nm, 300 W) because this composite formation enhanced visible light absorption and diminished photogenerated charge recombination [65]. In addition, Q. A. Damosh et al. also constructed a cauliflower-like heterojunction of tungsten oxide/black phosphorous/graphitic nitride (WO$_3$/BP/g-C$_3$N$_4$) using pulsed laser
ablation of an as-prepared mixture of WO$_3$/g-C$_3$N$_4$/BP in dimethylformamide (DMF) under sonication (355 nm, 10 Hz, 10 ns). Towards the overall photocatalytic production of gaseous hydrogen, the production rate of WO$_3$/g-C$_3$N$_4$/BP nanocomposites was around 400 µmol g$^{-1}$ h$^{-1}$, approximately five times higher than WO$_3$/BP (75 µmol g$^{-1}$ h$^{-1}$). The enhancement in photocatalytic performance was attributed to the formation of a Z-scheme heterojunction, which impeded the recombination of beneficial electrons and holes [66].

4. Summary and Outlook

In summary, photocatalytic reactions provide an effective approach for energy conversion between light and chemistry; the bridge medium is the photocatalyst. To date, a variety of photocatalysts have been developed, including many semiconductors, single-atoms, metal-organic frameworks (MOFs), and even transition metal carbides (2D Mxenes), and the corresponding photocatalytic applications are also diverse. Particularly, as the most classic photocatalyst with a long history, semiconductor-based nanocomposites have attracted remarkable attention because the light-generated charge carriers on the energy band can be effectively separated via the coupling effect or constructing heterojunction. As a powerful tool for nanomaterials fabrication, pulsed laser heating in liquids has been utilized to synthesize nanocomposites; the different components can be well-composited by facilely laser deposition, and the related photocatalytic performances can be enhanced.

Meanwhile, a few technical challenges also need to be investigated. On one hand, the way to improve the yield of products is worth considering. Despite the interaction between laser energy and colloids in the liquid being very fast (mainly determined by the pulse width and repetition rates), the production of laser-synthesized nanocomposites for each interval remains at a lower level (typically in milligram magnitude) because sample circulation can be hardly achieved by automation at the present time. Assuming that the colloidal suspension can be uniformly moved in the form of a stream or heated on an automatic platform, the yield of products can be easily expanded because the manual work for sample intervals can be canceled. This automatic approach may also be assisted by using advanced artificial intelligence. Moreover, most of the previous reports were investigated in the laboratory; there still existed a long period to realize industrialization because it is also restricted by the yield of production. For real industry, massive product quantities are needed. Despite the technique of pulsed laser synthesis of colloids for photocatalysis being innovative and smart, the lower production is still difficult to meet industry needs. Fortunately, with the development of high-power lasers, these issues are expected to be solved in the future because the volume of samples can probably be expanded under this laser source. On the other hand, the precise control of crystallinity and crystal phase of nanomaterials is also significant because the phase or crystallinity transformation of nanomaterials can easily occur during pulsed laser heating on the interface of each nanocomposite, which is not beneficial for the optimization of photocatalytic performance. Thus, more attention should be paid to these crafts and functional materials. Overall, this review provides a brief introduction to pulsed laser liquid preparation of nanocomposites for photocatalysis. It not only highlights the innovative works of this field but also outlines the related issues, giving great potential for dual investigations of high-performance photocatalysts and pulsed laser preparation of nanocomposites.

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