


REVIEW

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Advancements in visible-light-driven double perovskite nanoparticles for photodegradation

Runia Jana¹, P. Mary Rajaitha¹, Sugato Hajra¹ and Hoe Joon Kim^{1,2*} 

Abstract

Perovskites are of significant interest in the field of photocatalysis. To date, many perovskite nanostructures have been developed, and their applications in photocatalysis have been studied. There has been considerable improvement in the research on metal doping in the perovskite structure to improve their optical and structural properties. This mini-review examines the recent progress in the synthesis of lead-free double perovskite nanoparticles and their application in visible-light photocatalysis. Lead-free perovskites are emerging as an eco-friendly solution in energy, electrochemistry, and sensing. Double perovskites are known for their flexible structural, optical, and morphological properties due to their lattice framework having a general form $AA'BB'O_6$. They are more useful for hydrogen evolution due to their higher conduction band potential than simple perovskites. Here, we summarize the current progress and provide insights for the future development of double perovskites toward efficient photodegradation.

Keywords Double perovskite, Visible-light photocatalysis, Photodegradation, Lead-free perovskite

Introduction

The steadily increasing waste generation levels and the need to combat them using renewable energy sources have enabled the search for materials that can be produced easily and have no side effects harming the environment [1–3]. As solar energy is one of the abundantly available renewable energy sources, it makes sense to utilize it to counter waste degradation by synthesizing materials that can utilize directly [4, 5]. As photocatalysis is one of the most widely researched methods for eliminating effluents from factories and the agricultural sector for their various advantages (Fig. 1), semiconductor metal oxides have always been the materials of choice for helping to generate non-toxic by-products by degrading

wastes [1, 6–8]. But semiconductors have drawbacks: fast electron–hole recombination rate, high bandgap energy, and lower adsorption rates deter the catalytic progress [9, 10]. As one of the most crucial factors of photocatalysis is charge separation, knowledge of the crystal structure and constituent elements of the material is particularly important [11].

Photocatalysis for waste degradation is a promising technology that utilizes the photo-induced redox reaction of a photocatalyst material to convert harmful pollutants into less toxic or non-toxic substances. When a photocatalyst is irradiated with light, it generates electron–hole pairs, which act as powerful oxidizing or reducing agents. These electron–hole pairs can then react with water or oxygen molecules to produce highly reactive oxygen species (ROS), such as hydroxyl radicals ($\bullet OH$), superoxide radicals ($\bullet O_2^-$), and hydrogen peroxide (H_2O_2) [12]. These ROS can attack and break down organic and inorganic pollutants in waste materials, converting them into simpler and less harmful compounds. One of the key advantages of photocatalysis is that it is a green and sustainable approach that does not require

*Correspondence:

Hoe Joon Kim
joonkim@dgist.ac.kr

¹ Department of Robotics and Mechatronics Engineering, Daegu Gyeongbuk Institute of Science and Technology (DGIST), Daegu 42988, Republic of Korea

² Robotics and Mechatronics Research Center, Daegu Gyeongbuk Institute of Science and Technology (DGIST), Daegu 42988, Republic of Korea

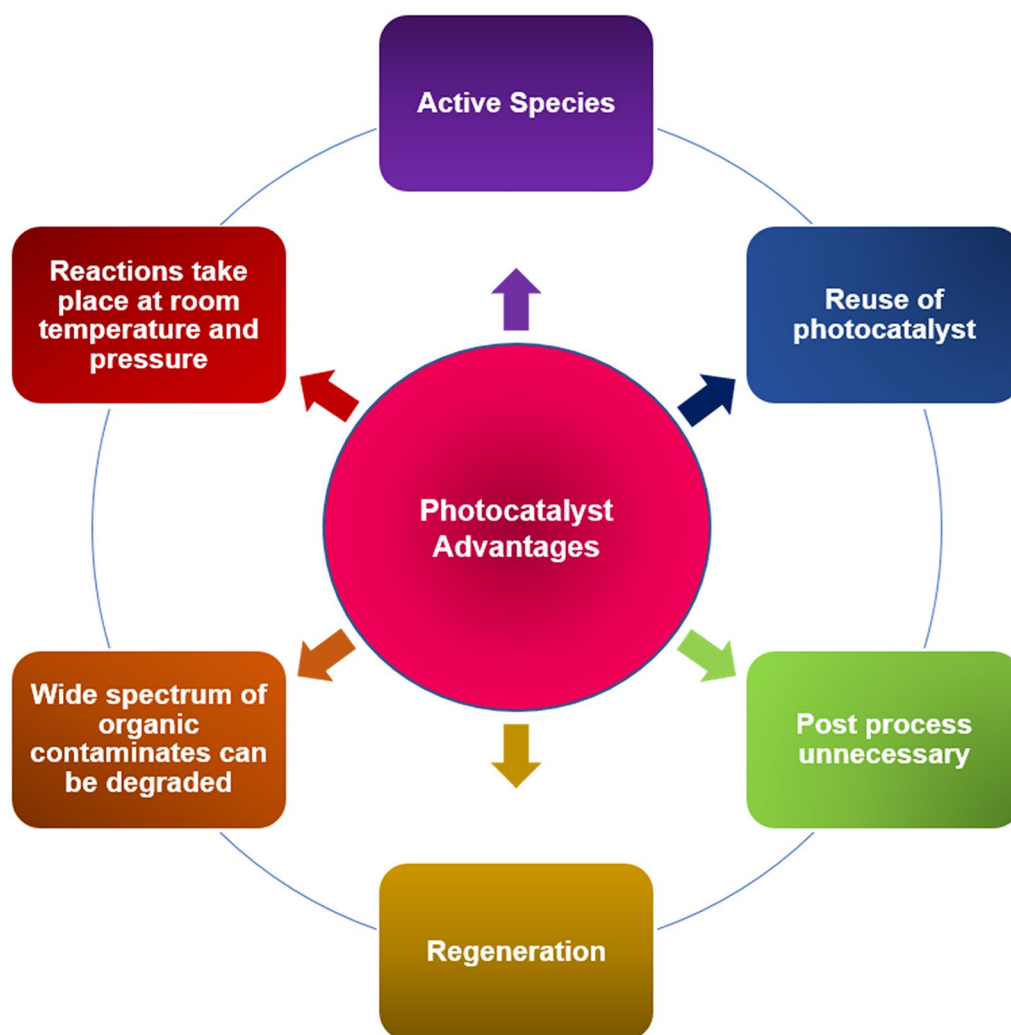


Fig. 1 Properties that make a material suitable for photocatalysis

the use of harmful chemicals or the production of harmful by-products [13].

Additionally, photocatalysis can be used to degrade a wide range of waste materials, including organic pollutants, heavy metals, and dyes. However, one of the main challenges of photocatalysis is the selection of an appropriate photocatalyst material that is efficient and stable under the conditions required for waste degradation [14]. Another challenge is the design of the photocatalytic reactor to ensure efficient use of light energy and proper mixing of the waste material with the photocatalyst [15]. Nevertheless, photocatalysis shows great potential as an effective and sustainable solution for waste degradation.

Conventional perovskite structures with the chemical formula of ABO_3 have been known to contain lead (Pb), and recent research is all focussed on replacing Pb with non-toxic materials for fuel cells, photovoltaics,

memory devices, and photocatalysts [3, 16]. As the Pb-based perovskites have great power conversion efficiency, non-Pb-based perovskites must be studied to combat the instability and large energy band gap [17]. Lead-free perovskites have been synthesized by various methods, such as sol–gel, solid-state synthesis, and hydrothermal methods, to increase the chances of tuning the optical and structural properties to cater to the respective needs of the society, where perovskite oxides and halides are deemed to be effective [18–20]. Double perovskites are known for their flexible structural, optical, and morphological properties due to their lattice framework having a general form AA'BB'O_6 . They are more useful for hydrogen evolution due to their higher conduction band potential than simple perovskites. The existence of double metal provides structural variation, which is very promising for optical applications [21].

This mini-review introduces state-of-the-art studies that utilize lead-free double perovskites for photodegradation using visible light sources.

Double perovskites for visible light photocatalysis

Double perovskites enlarge the possible elements that can be utilized in the perovskite family and have appeared as promising proxies with elevated photocatalytic performances, as they can be more flexible regarding their bandgaps and edges [22–24]. Nair et al. have provided a comprehensive review of the recent research on synthesizing double perovskites and their applications [17]. There have been experimental results of lead-free perovskites showing better photocatalytic results owing to their non-toxicity. Yin et al. have provided a comprehensive review of oxide perovskites, showing improved performance in photocatalytic activities. They have summed up the typical characteristics of double perovskite oxides having higher light absorption and favorable tolerance to structural defects to be the factors that help in upgrading their photocatalytic behavior [25]. Ma et al. have commented on the adaptability of double perovskite oxides regarding their A or B site ions which makes regulation of desired properties less challenging, like the tuning of bandgap and varying their optoelectronic nature [26]. Jiang and co-workers have computed and identified stable double perovskite oxides, which can surpass existing perovskite oxides in their optoelectronic and photocatalysis significance [27]. Halder et al. have discussed how direct-to-indirect transition in bandgap structures influences the efficiency of catalytic behavior of perovskites by doping Ru in $\text{Ba}_2\text{HoSbO}_6$ [28].

Double perovskite nanoparticles have a high surface area-to-volume ratio, which makes them suitable for device-level fabrication in photocatalysis. They can be incorporated into thin films, coatings, or membranes to create high surface area photocatalytic surfaces, which can be used in waste degradation, water treatment, or air purification. Deposition techniques such as spin-coating, spray deposition, or inkjet printing can fabricate thin films or coatings [29, 30]. Incorporation into porous membranes or filters can create high surface area photocatalytic surfaces for water or air treatment [31]. Device-level fabrication of double perovskite nanoparticles is still in the early stages, and much work is needed to optimize the fabrication processes and understand their underlying mechanisms. Nonetheless, this research area has great potential for developing practical and efficient photocatalytic systems for environmental remediation and renewable energy conversion.

$\text{K}_2\text{Ta}_2\text{O}_6$

Usually, most tantalate oxides prepared by the solid-state method require high temperatures, with their secondary oxides being undesirable [32]. $\text{K}_2\text{Ta}_2\text{O}_6$, a pyrochlore-lattice structure, is known to be the secondary phase that forms at lower temperatures when synthesizing KTaO_3 , which is not desirable due to its low dielectric properties, high ferroelectric properties [33], and defect structure. It has a lower electron–phonon coupling effect, which shows enhanced charge separation, leading to better photocatalyst behavior than KTaO_3 [34]. $\text{K}_2\text{Ta}_2\text{O}_6$ is usually easily synthesized by hydrothermal or the sol–gel method due to the ease of not requiring elevated temperature [35–38]. Because most tantalum oxides have band gaps unsuitable for the absorbance of visible light for any optical applications, surface modification or doping $\text{K}_2\text{Ta}_2\text{O}_6$ with elements like noble and rare metals have been adopted to reduce the band gap energy [39]. Such reduction in bandgap energy has helped in charge separation and generation of active species of hydroxyl and superoxide radicals, boosting the catalytic behavior. Krukowska et al. have doped Y, Yb, Ho, Pr, E rare earth metals, Au, Ag, Pt, Pd, Rh, Ru as monovalent metals and Au/Pt, Ag/Pd, Rh/R as bimetallic nanoparticles in their works, and have examined the response of $\text{K}_2\text{Ta}_2\text{O}_6$ [40].

Angineni et al. have experimented with doping three transition metal ions, M^{2+} ($\text{M} = \text{Mn}, \text{Co}, \text{and Ni}$), by ion exchange method after synthesizing $\text{K}_2\text{Ta}_2\text{O}_6$ by facile hydrothermal method [35]. They have observed tailing in the material despite confirming the absence of impurities [41], attributing them to lattice defects and varying metal oxidation states. All the prepared samples have cubic crystal structure agglomerated spherical shapes, ranging from 36.64 to 38.25 nm. On photodegradation of methylene blue dye with all the catalysts, 53% of the dye was degraded using the undoped $\text{K}_2\text{Ta}_2\text{O}_6$, while Mn-doped $\text{K}_2\text{Ta}_2\text{O}_6$ degraded 93%, Co-doped $\text{K}_2\text{Ta}_2\text{O}_6$ degraded 86% and Ni-doped $\text{K}_2\text{Ta}_2\text{O}_6$ 69%. They have also verified that hydroxyl radicals are the reactive oxygen species by observing the addition of IPA scavenger (Fig. 2: a1–a6).

Angineni et al. doped Gd^{3+} ions in $\text{K}_2\text{Ta}_2\text{O}_6$ to comprehend how doping allows visible light photocatalytic performance [36], using methylene blue dye as the pollutant. They have shown that all doped samples have lower energy band levels than parent $\text{K}_2\text{Ta}_2\text{O}_6$, with 0.075 mol% Gd^{3+} doped $\text{K}_2\text{Ta}_2\text{O}_6$ having 4.68 eV being the least, attributed to the formation of new band energy levels. Irregular-shaped agglomerated particles having cubic morphology with a size range of 37.34 nm to 37.68 nm were synthesized. 0.075 mol% Gd^{3+} doped $\text{K}_2\text{Ta}_2\text{O}_6$ had the least crystallite size of 35.92 nm and showed the best photodegradation, i.e., 79% MB degrading efficiency for 3 h of irradiation and maximum adsorption of 12%

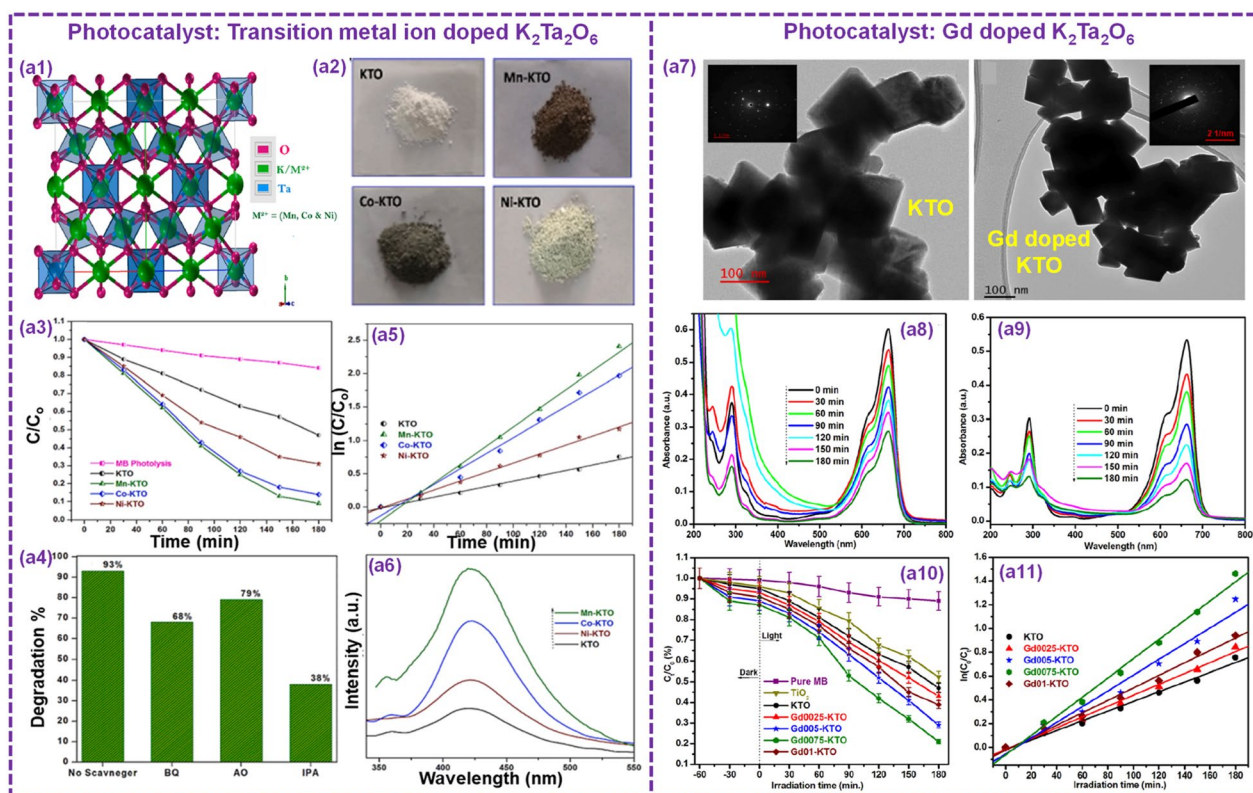


Fig. 2 **a1** Schematic structure of KTO lattice, **a2** Digital photographs of all catalysts, **a3** Degradation curves and **a4** The first-order reaction kinetics plot for degradation of MB under visible light irradiation, **a5** Effects of different scavengers on the photocatalytic degradation of MB over Mn-KTO. **a6** Fluorescence spectra of all catalyst suspensions in the TA solution for 120 min of irradiation time ($\lambda_{\text{exc}} = 320$ nm). Reprinted from [35] with permission from Elsevier. Copyright (2022) Elsevier. **a7** TEM images of KTO and Gd0075-KTO (insets show the corresponding SAED patterns). **a8** The changes of temporal UV-Vis absorption spectra during the degradation of MB aqueous solution over KTO and **a9** Gd0075-KTO, **a10** Photocatalytic degradation profiles of MB as a function of the irradiation time under visible light over all catalysts and **a11** pseudo-first-order kinetics for degradation of MB under visible light irradiation for 180 min (Reprinted from [36] with permission from Elsevier. Copyright (2022) Elsevier)

after 1 h (Fig. 2: a8–a9). Using scavengers to understand the nature of reactive species, it is reassuring that for $\text{K}_2\text{Ta}_2\text{O}_6$, hydroxyl radicals are the dominant species contributing to the degradation process. The prepared samples also retained their stability after 5 cycles of repeated experiments, emphasizing the degradation ability of the Gd^{3+} doped $\text{K}_2\text{Ta}_2\text{O}_6$ nanoparticles. The structural advantages of $\text{K}_2\text{Ta}_2\text{O}_6$ as photocatalytic materials over other double perovskites are ongoing, as limited work is presently done.

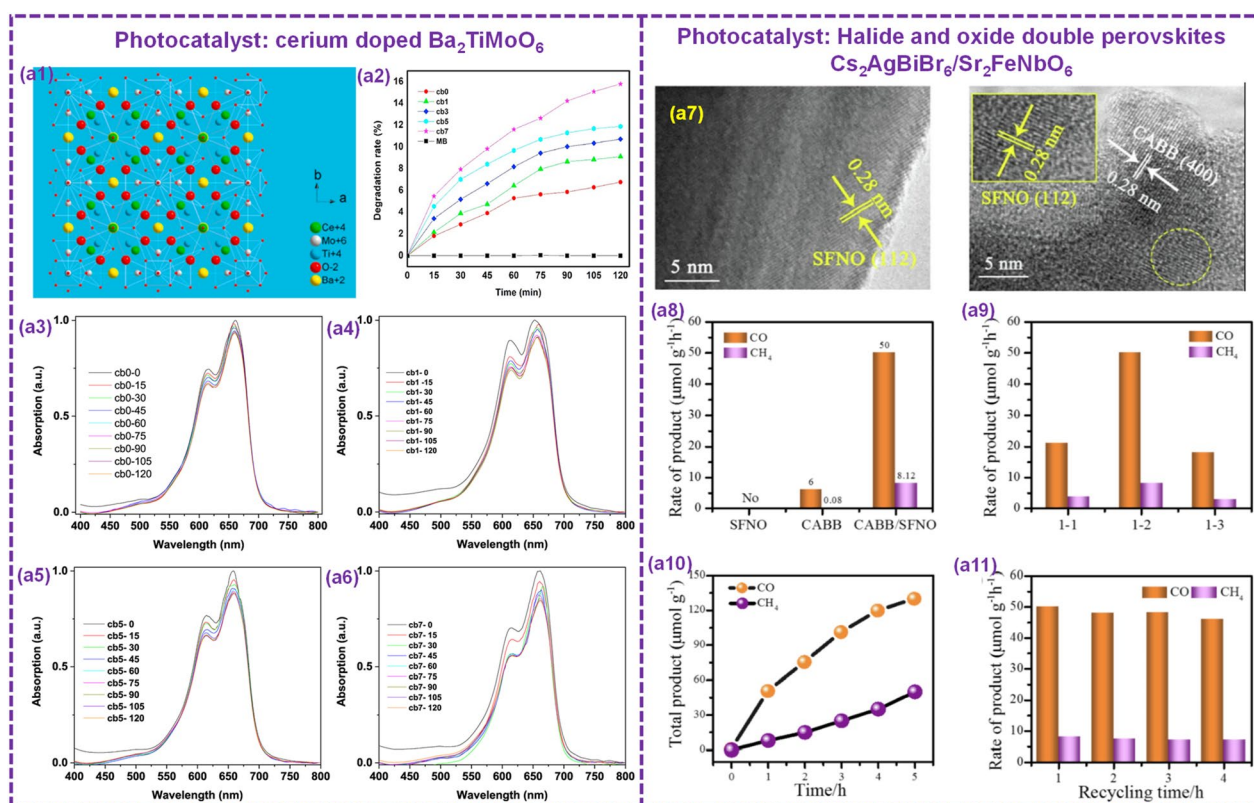
$\text{Ba}_2\text{TiMoO}_6$

$\text{Ba}_2\text{TiMoO}_6$ double perovskite has been reported to exhibit ferroelectric hysteretic behavior at room temperature [42]. It behaves as a p-type semiconductor but manifests a decrease in the current flow when Ce^{3+} ions are doped in the host lattice [43] when synthesized by the solid-state reaction method. Ghrib et al. have reported well-synthesized Ce^{3+} doped $\text{Ba}_2\text{TiMoO}_6$ by solid-state reaction method and annealed twice to obtain

homogeneous powder samples [44]. They have obtained spherical-shaped irregular grains with a size ranging from 8–22 nm, having energy levels from 3.41–3.6 eV. The maximum degradation shown by 7% Ce^{3+} doped $\text{Ba}_2\text{TiMoO}_6$ is almost 16% for industrial pollutant methylene blue degradation in the presence of hydrogen peroxide (H_2O_2). The significance of this prepared material is its ability to absorb energy in the visible electromagnetic spectrum and show evident results in 120 min. Ghrib et al. have emphasized the importance of the Ce 4f level due to doped ions, with hydroxyl radicals being the reactive species enforcing degradation (Fig. 3: a1–a6).

$\text{Sr}_2\text{FeNbO}_6$

$\text{Sr}_2\text{FeNbO}_6$, known for its applications in cell electrode materials for solid-oxide fuel cells [45–47], is also efficient as a visible light n-type semiconductor photocatalyst with a bandgap of 2.04 eV [48]. This double perovskite has been synthesized by Jeong et al., with a band gap of 2.06 eV by solid-state method, and it has



been proven to photo reduce and oxidize water to evolve hydrogen and oxygen, indicating its use as a visible light application material for degradation of wastes [49]. Idris et al. have successfully synthesized a heterostructure of lead-free double-perovskites $\text{Cs}_2\text{AgBiBr}_6/\text{Sr}_2\text{FeNbO}_6$ for photocatalytic CO_2 reduction [48]. It has been observed that the lead-free double perovskites have band gaps of 2.04 eV for $\text{Sr}_2\text{FeNbO}_6$ and 2.17 eV for $\text{Cs}_2\text{AgBiBr}_6$. The heterostructure has shown photocatalytic behavior in the reaction cell saturated with pure CO_2 in visible light in an ethyl acetate solution. They have prepared the heterostructure by an electrostatic self-assembly method, which has resulted in the formation of pure cubic phases for both $\text{Sr}_2\text{FeNbO}_6$ and $\text{Cs}_2\text{AgBiBr}_6$ with irregular aggregated polyhedral nanoparticles in the size range of 20–50 nm. When one compares the performance of individual photocatalysts with the heterostructure, it has been observed that the reduction rates are drastically higher for the heterostructure, implying possible application in pollutant removal. The stability of the heterostructure has also been examined over the time of 5 h for

four cycles and confirmed with XRD and SEM mapping (Fig. 3: a7–a11).

$\text{La}_2\text{FeTiO}_6$

$\text{La}_2\text{FeTiO}_6$ synthesized by the sol–gel method has been reported to have still dielectric properties at room temperature to 200 °C [50]. It is a ternary metal oxide, which can be considered a possible replacement for TiO_2 photocatalysts. It is placed in the same category as other double perovskites where the B-site cation is doped, resulting in the change in band gaps exhibiting varying properties. Hu et al. [51] have synthesized LaFeO_3 perovskite and corresponding double perovskite $\text{La}_2\text{FeTiO}_6$ by sol–gel method to degrade p-chlorophenol in visible light. They found that the irregular polyhedron $\text{La}_2\text{FeTiO}_6$ nanostructures with a diameter of around 70 nm and crystallite size of 26.3 nm can degrade the organic effluent better than LaFeO_3 . $\text{La}_2\text{FeTiO}_6$ could degrade >62% compared to LaFeO_3 's degradation of 49% in visible light for 5 h. They were the first to report the superiority of $\text{La}_2\text{FeTiO}_6$ over LaFeO_3 for photodegradation.

Nunes et al. have experimented with varying the amount of Ti in $\text{LaTi}_{1-x}\text{Fe}_x\text{O}_3$ (where $0 \leq x \leq 1$) and observed the changes in degrading Acid Orange 7 under ultraviolet and visible light [52]. They obtained agglomerated nanoparticles with non-homogeneous distribution and shapes. It is also observed with the increasing value of Ti and decreasing value of Fe in the prepared double perovskite, the energy band gap has steadily decreased from 3.97 eV for $x=0$ to 2.2 eV for $x=1$ (Fig. 4: a1), with increasing photocatalytic activity (3% in 1 h) in the presence of visible light, and high photodegradation of 47% achieved by the $\text{La}_2\text{Ti}_2\text{O}_7$ particles, in 1 h under UV light (Fig. 4: a2).

$\text{BiFe}_{1-x}\text{Co}_x\text{O}_3$

BiFeO_3 is a known multiferroic material used for memory devices, piezoelectric sensors, photodetectors, and magnetoelectric devices [53, 54]. But it is known to have a

low energy band gap, which makes it suitable for visible light applications [55]. In an attempt to tweak the band gap for effortless application in photocatalytic activities, cobalt has been doped in the host perovskite to observe the structural and optical changes [56]. Prasannakumara et al. have synthesized Co-doped BiFeO_3 by sol-gel method and observed that with an increase in the concentration of Co ion doping, the energy band gap has steadily increased from 1.74 eV for an undoped thin film to 1.91 eV for $\text{BiFe}_{1-x}\text{Co}_x\text{O}_3$ where $x=0.08$ is the maximum doping.

Ponraj et al. have prepared $\text{BiFe}_{1-x}\text{Co}_x\text{O}_3$ ($x=0, 0.025, 0.05, 0.075$ and 0.10) and observed their photocatalytic ability by degrading Acid Red-85 dye. They have observed a decrease in crystallite size from 57 to 32.29 nm with increasing Co ion concentration with rhombohedral structure [57], attributing it to the size difference between Co^{3+} ions compared to Fe^{3+} ions. They reported that the

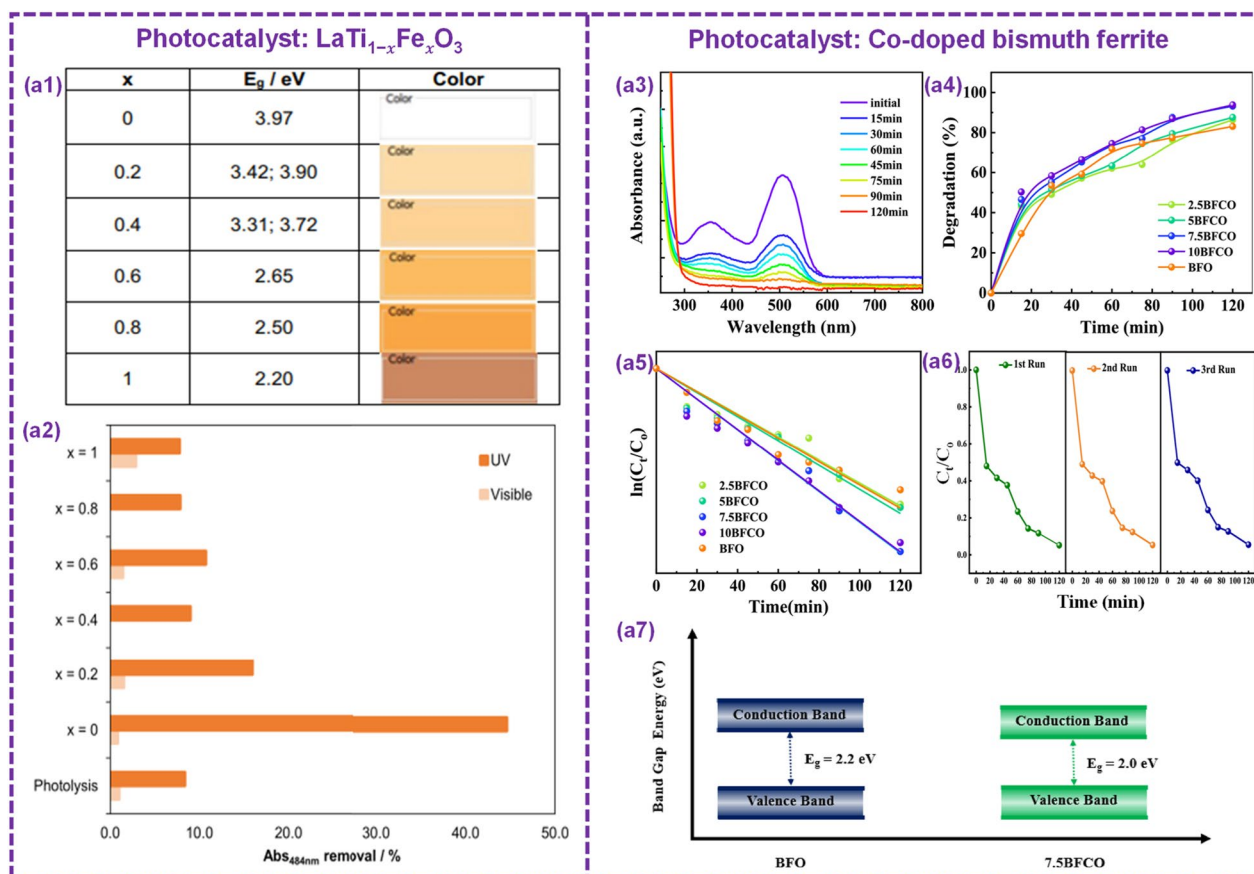


Fig. 4 a1 Table E_g values and color of the $\text{LaTi}_{1-x}\text{Fe}_x\text{O}_3$ oxides. a2 Absorbance removal of the AO7 solutions ($C_i = 10 \text{ mg L}^{-1}$) obtained with the photocatalyst powder suspensions ($C = 0.5 \text{ g L}^{-1}$) under UV and visible light after 1 h. Reprinted from [52] with permission from Elsevier. Copyright (2022) Elsevier. a3 Absorbance Spectra of AR-85 as a function of time for 7.5BFCO photocatalyst. a4 Percentage of degradation of Acid Red-85 dye using $\text{BiFe}_{1-x}\text{Co}_x\text{O}_3$ ($0 \leq x \leq 0.10$) as photocatalysts. a5 Pseudo first order kinetics fitting for $\text{BiFe}_{1-x}\text{Co}_x\text{O}_3$ ($0 \leq x \leq 0.10$). a6 Recyclability of 7.5BFCO for AR-85 degradation for three runs. a7 Schematic diagram of the possible mechanism involved for electronic energy band in BFO samples (Reprinted from [57] with permission from Elsevier. Copyright (2022) Elsevier)

degradation efficiency of $\text{BiFe}_{1-x}\text{Co}_x\text{O}_3$ ($x=0.075$) was the highest at 93.79% in 2 h, even higher than 1 mol% coped sample due to presence of impurities (Fig. 4: a3–a7). The prepared samples have also shown reusability for 3 cycles, with the reaction following the pseudo-first order kinetics. They have credited the improvement in photodegradation ability of the double perovskite to the increased oxygen vacancies and reduced crystallite sizes.

Conclusion

The future advancement of double perovskites for visible light photocatalysis looks very promising. Several research studies have shown that double perovskites have a wide range of potential applications in solar energy conversion, including photocatalytic degradation of pollutants, water splitting, and CO_2 reduction. The unique structural and electronic properties of double perovskites make them ideal for use as photocatalysts, as they can absorb a broad range of visible and ultraviolet light and exhibit high stability and activity. In the coming years, researchers are expected to continue exploring the potential of double perovskites for visible light photocatalysis, focusing on improving their efficiency, stability, and selectivity.

One area of research that is likely to receive increased attention is the development of new doping materials to enhance the photocatalytic activity of double perovskites further. Moreover, with advancements in materials science, new synthesis routes and fabrication methods would be developed to optimize the properties of double perovskite nanomaterials, leading to their practical application in photocatalytic degradation. The synthesis of double perovskites is challenging and requires precise control over stoichiometry, temperature, and reaction time. The complex crystal structure of double perovskites makes characterization difficult and requires advanced techniques. Stability under environmental conditions, such as humidity and temperature, can also affect photocatalytic activity and practical applications. Overall, the future of double perovskites in visible light photocatalysis is very bright, and it is expected that they will play a crucial role in the development of more efficient and sustainable technologies for waste degradation and other energy conversion applications.

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Author contributions

RJ: Conceptualization, figure preparation, writing—original manuscript; PMR: writing—original manuscript; SH: data curation; HJK: Supervision, funding acquisition, writing—review. All authors read and approved the final manuscript.

Availability of data and materials

Data and material are available upon request to authors.

Declarations

Competing interests

The authors declare there is no conflict of interest related to this work.

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