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Next-Generation Visible-Light Perovskite Composites for Efficient Wastewater Remediation: A Review

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Abstract | **Review Article**

Pollutants like dyes, drugs, pesticides, and heavy metals such as Cr(VI) that are found in wastewater are very hard to degrade using conventional treatment processes especially in trace amounts. Photocatalysis offers an efficient degradation technique using light to degrade the pollutants. The standard photocatalyst used is titanium dioxide (TiO_2) that acts under ultraviolet light (which makes up 4-5% of the solar spectrum). Perovskite oxides are desirable due to the fact that their compositions can be altered to suit visible light; however, they suffer from rapid recombination when used on their own. This review article focuses on how oxide perovskites, mostly titanates, are combined with other substances, mainly graphitic carbon nitride (g-C_3N_4), in type II, Z-scheme, and S-scheme heterojunctions to enhance light absorption and charge separation. The MOFs are included as a reference material for Cr(VI) photoreduction. Four major gaps have been identified after reviewing the articles considered in this study. There are few articles on each individual perovskite family that cannot be compared to one another. The charge-transfer process in perovskites is assigned without enough proof. No byproducts such as Cr(III) are reported in Cr(VI) photoreduction studies. Priorities for closing these gaps are proposed so that laboratory results can move toward practical water treatment.

Keywords: Perovskite photocatalysts; visible light; heterojunction; wastewater remediation; S-scheme; emerging contaminants.

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1. INTRODUCTION

Contamination of water bodies is a serious worldwide issue which is increasing in magnitude and complexity (Zawadzki, 2022). The main sources of water contamination are industrial wastewater, agricultural runoff, and municipal and pharmaceutical waste. They include dyes, pesticides, pharmaceutical and personal care products that are discharged into surface and groundwater (Zawadzki, 2022); many of them are chronic pollutants at minute concentrations (Zawadzki, 2022). Every existing conventional method of treatment has certain strengths and weaknesses (Crini & Lichtfouse, 2019); therefore, the removal of trace contaminants from water has become as relevant an issue as that of bulk pollutants' removal. For instance, pesticides and phenolics have always been used as examples of pollutants degraded by photodegradation methods (Ahmed *et al.,* 2011). Heavy metal ions also pose problems. Cr(VI), a hexavalent chromium, is very mobile and also possesses carcinogenic and teratogenic qualities (García *et al.,* 2022). There is ongoing research to remove heavy metal ions from industrial effluents (Barakat, 2011). Reduction of metal ions, like Cr(VI), requires a unique approach in terms of chemistry compared to organic compounds; that is, the goal should be reduction and not oxidation of the metal ion. The use of semiconductor photo catalysis in this scenario would be very appropriate since the same technique could be used for degrading organic substances as well as reduction of metal ions using light energy (Schneider *et al.,* 2014).

Due to the wide range of components present in wastewater, one technique cannot be applied universally (Nkwachukwu & Arotiba, 2021). The need for complementary methods such as advanced oxidation

processes has led to the development of research on semiconductor photocatalysis as a technique for treating water and wastewater (Nkwachukwu & Arotiba, 2021). The efficiency of a photocatalytic reaction process is based on three interrelated properties: the ability of the material to absorb light, the separation and use of charge carriers, and the stability of the material during the reaction (Schneider *et al.,* 2014).

Since the work of Fujishima and Honda (1972), TiO_2 has been the standard photocatalyst. With a band gap of around 3.2 eV, its activation is limited to ultraviolet light, which accounts for 4–5% of the solar spectrum. As such, the search for visible-light photocatalysts is an essential task (Etacheri *et al.,* 2015). Perovskites (ABO_3) have shown great potential in this regard. Perovskite's stable structure can accommodate various metal ions, thus allowing the manipulation of the band structure (Nkwachukwu & Arotiba, 2021), with the latter along with band gaps, band edges, and lifetimes being crucial to their performance (Clabel *et al.,* 2022). However, since it is highly unlikely that single perovskites possess all necessary properties, composites and heterostructures have emerged by incorporating perovskites into other phases (Patra *et al.,* 2023). Over time, heterojunction design has progressed from type-II and Z-scheme architectures (Xu *et al.,* 2018) to S-scheme designs in an attempt to clear some uncertainties about charge transfer mechanism (Xu *et al.,* 2020).

A number of important limitations exist. Current literature reviews consider only one family of perovskites or one aspect: perovskite oxides for organic pollutants degradation (Nkwachukwu & Arotiba, 2021); effect of band gap and carrier lifetimes (Clabel *et al.,* 2022); or g-C_3N_4-titanate composite materials (Patra *et al.,* 2023). A comprehensive review of the composite design for perovskites in relation to various pollutants is missing. Moreover, proposed Z and S schemes of electron transfer should be verified more rigorously (Xu *et al.,* 2020). The reporting is also inadequate: in studies that use MOF for photoreduction of chromium (VI) ions, for instance, formed Cr(III) ions are not detected (García *et al.,* 2022). Without systematic data on stability, metal leaching, and efficiency in natural effluents, high efficiency obtained under laboratory conditions cannot be applied confidently in practice.

This review is narrative and does not follow a systematic approach. The study covers perovskites oxide materials, especially titanates based, in combination with g-C_3N_4 as well as heterojunctions of perovskites and g-C_3N_4 with the use of dyes, antibiotics, and Cr(VI) as examples of pollutants. Halide perovskites are not included in the review since their stability in water is another issue. Chapter 2 highlights the basics of photocatalysis and perovskites oxides. Chapter 3 compares different approaches in the design of composites. Chapter 4 covers the application of such composites in degradation of organic pollutants and Cr(VI).

2. Fundamentals of Photocatalysis and Perovskite Materials

2.1 Principles of semiconductor photocatalysis and reactive species

The structure of the semiconductor consists of the valence and conduction bands, which are separated by the band gap (Hoffmann *et al.,* 1995). Absorption of a photon with energy equal to or more than the band gap results in the movement of the electron into the conduction band, leaving the hole (Schneider *et al.,* 2014). The carriers should then be dissociated and moved to the surface, where they react with adsorbed species (Hoffmann *et al.,* 1995). Detailed information on basic concepts and modes of photocatalysis can be found elsewhere (Yang & Wang, 2018; Zhu & Wang, 2017).

On the surface level, holes can oxidize organic compounds, while electrons can reduce dissolved compounds, like oxygen and metal ions (Schneider *et al.,* 2014). Reactive species that result from this process will break down organic pollutants, such as dyes, pesticides and pharmaceuticals (Zawadzki, 2022); the way dyes are degraded by TiO_2 is explained thoroughly (Ajmal *et al.,* 2014). Reduction process transforms Cr(VI) into a more favorable state (García *et al.,* 2022). Since reduction and oxidation are taking place at the same time, the ratio of electron and hole utilization influences efficiency, and the reduction products have to be considered (García *et al.,* 2022).

However, the major competing process is recombination. In cases where recombination happens before the particles reach the surface, the energy absorbed is wasted (Hoffmann *et al.,* 1995). Lifetime of the carriers is very important, and this is the reason why materials with same bandgap have different activities (Clabel *et al.,* 2022).

2.2 Band gap, band edges and the visible-light requirement

The band gap plays a significant role in determining the region of the solar spectrum that is used by the photocatalyst. Due to its large band gap of about 3.2 eV, pristine TiO_2 is not efficient in utilizing sunlight; hence, it is vital to have a smaller band gap or even reduce the band gap further (Etacheri *et al.,* 2015). Having a small band gap is not all what is required. It is necessary to ensure that the valence and conduction band edges provide the necessary driving force to facilitate the reaction (Clabel *et al.,* 2022).

2.3 Structure and compositional flexibility of perovskite oxides

Perovskite oxide systems have a formula of ABO_3, where two types of metal ions exist in different locations in a structurally stable lattice (Nkwachukwu & Arotiba, 2021). The selection or substitution of these

metal ions is responsible for tuning band gap, band edge and carrier lifetime (Clabel *et al.,* 2022). Examples of reported photocatalytic systems include lanthanum-, ferrite-, titanate- and tantalum-based perovskites (Nkwachukwu & Arotiba, 2021). Visible-light active perovskites have been comprehensively reviewed (Kanhere & Chen, 2014), and titanate perovskites have been systematically reviewed in terms of synthesis, characterisation and photocatalytic applications (Paramanik *et al.,* 2022). Alkaline earth titanates have been reviewed in the context of water purification (Solís *et al.,* 2021), and improvement of $BaTiO_3$ based photocatalyst for water treatment have been critically evaluated (Ray *et al.,* 2021).

Experimental research explains how sensitive activities are to composition and structure. Titanate perovskite nanoparticles have been made via a low-temperature process to be used as photocatalysts (Alammar *et al.,* 2015). Structural, optical, and photocatalytic features of $ATiO_3$ ceramics, where A is Ca, Sr, and Ba, were compared (Karakozov *et al.,* 2021). Differences in structures of $CaTiO_3$ and $SrTiO_3$ were related to their different efficiencies in VOC mineralization (Yang *et al.,* 2021). Adding Na or Na/Fe to $BaTiO_3$ modified its dielectric, ferromagnetic and sunlight photocatalytic characteristics (Yakout, 2020). In theory, composition provides the way to produce catalysts which can absorb visible light and retain redox capability. However, in practice, enhancing one characteristic inevitably has an effect on another, so optimization requires considering multiple characteristics simultaneously (Clabel *et al.,* 2022).

2.4 Heterojunction concepts for improved charge separation

Considering recombination and limited visible light response, combining two or more semiconductor materials in heterojunction form becomes an effective practice (Patra *et al.,* 2023). This is meant to achieve spatial separation of electrons and holes so that more of them can be brought to the surface (Xu *et al.,* 2018). While in type II junctions carriers get separated across both materials, carriers in this case possess less redox power (Xu *et al.,* 2018). However, direct Z-scheme approach allows for maintaining higher redox power carriers through recombination of less efficient ones at the interface (Xu *et al.,* 2018); review of such systems for photoelectrochemical applications has been carried out extensively (Li *et al.,* 2022). Later on, the S-scheme concept has been proposed to address the problem of uncertainty in the description of charge transfer mechanisms in earlier approaches (Xu *et al.,* 2020)

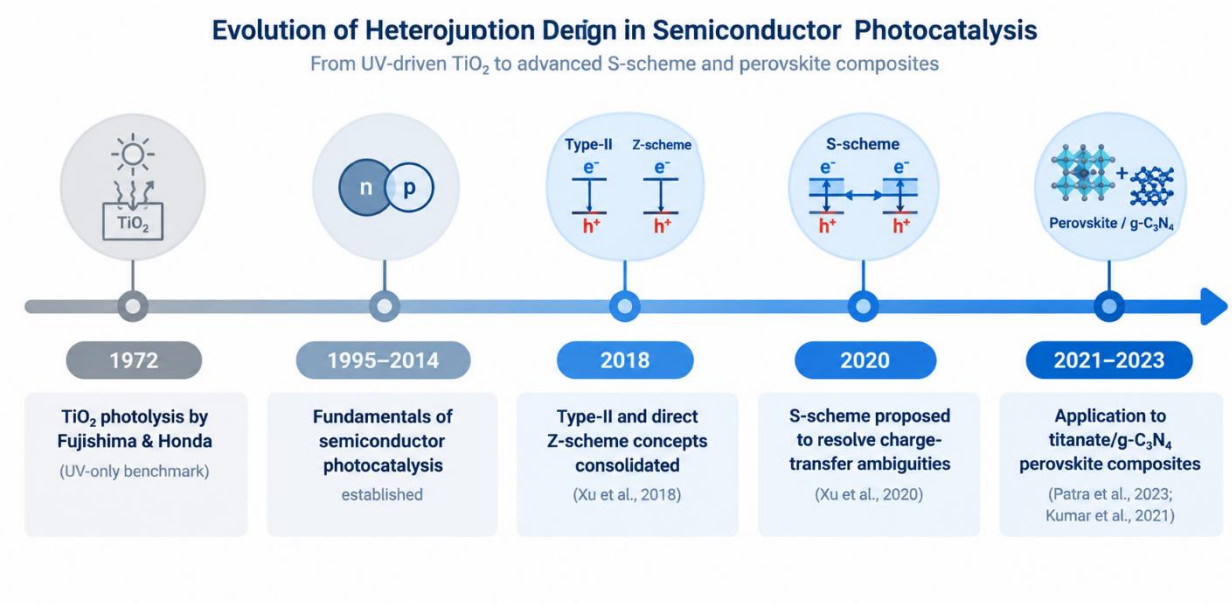


Figure 1: Timeline of heterojunction design evolution from UV-only TiO_2 to type-II, Z-scheme, and S-scheme perovskite composites

3. Composite Design Strategies for Visible-Light Perovskite Photocatalysts

3.1 Rationale

The combination of high visible light absorption efficiency, good charge separation capability, and stability is difficult for any individual photocatalyst. Properties of perovskites are correlated, and optimization of one of these properties usually leads to deterioration of the other one (Clabel *et al.,* 2022). Composites should help to enhance the utilization of visible light and charge separation, which are two main bottlenecks for the single-phase catalysts (Patra *et al.,* 2023; Schneider *et al.,* 2014).

3.2 Perovskite–g-C_3N_4 composites

Graphitic carbon nitride (g-C_3N_4) is a non-metallic polymeric photo catalyst that works under visible light (Wang *et al.,* 2009), and g-C_3N_4 based photo

catalysts have been systematically reviewed for environmental applications (Ong *et al.,* 2016). The combination of titanate perovskites with g-C_3N_4 has been systematically reviewed for water purification application: in which both phases are placed in proximity to each other, so that their interface separates charges (Patra *et al.,* 2023).

A number of studies provide the variety of this approach (Table 1). The $SrTiO_3$/g-C_3N_4 heterojunction has been used for degrading organic pollutants by means of the Z-scheme mechanism as a result of scavenger trapping experiments (Konstas *et al.,* 2018). The $CaTiO_3$/g-C_3N_4 photocatalysts have been studied for triphenylmethane dyes degradation under simulated sunlight (Chen *et al.,* 2020) as well as for methylene blue and levofloxacin (Zhao *et al.,* 2023). The $SrZnTiO_3$/g-C_3N_4 heterostructure has been found to operate with the light source-dependent charge-transfer mechanism (Kim & Jo, 2019). The $SrTiO_3$/g-C_3N_4 system modified by plasmonic Ag was called the Z-scheme polymeric-perovskite heterojunction (Sohrabian *et al.,* 2023). Moreover, the nZVI-doped Al_2ZnTiO_6 double perovskite/g-C_3N_4 photocatalyst has been developed for degradation of organic and inorganic contaminants under visible light (Kadkhodayan & Alizadeh, 2023).

Composite	Target pollutant(s)	Feature reported	Source
$SrTiO_3$/g-C_3N_4	Organic pollutants (methylene blue as model)	Z-scheme proposed from scavenger experiments	Konstas *et al*. (2018)
$CaTiO_3$/g-C_3N_4 (2D/1D)	Triphenylmethane dyes	Simulated sunlight; face-to-face contact	Chen *et al.* (2020)
$CaTiO_3$/g-C_3N_4	Methylene blue, levofloxacin	Dye and antibiotic tested	Zhao *et al.* (2023)
$SrZnTiO_3$/g-C_3N_4	Dyeing-industry pollutants	Light-source-dependent charge transfer	Kim & Jo (2019)
nZVI-doped Al_2ZnTiO_6/g-C_3N_4	Organic and inorganic contaminants	Double perovskite; visible light	Kadkhodayan & Alizadeh (2023)
g-C_3N_4/$Bi_4Ti_3O_{12}$/$Bi_4O_5I_2$	Ofloxacin (antibiotic)	Dual Z-scheme; 87.1% ofloxacin removal with H_2 evolution	Kumar *et al.* (2021)

These reactions vary in terms of perovskite, modification, target pollutant, and light source, so they cannot be compared. Whether the results for titanate-based composites can be applied to perovskites based on ferrite, lanthanum, or tantalum compounds (Nkwachukwu & Arotiba, 2021) have also not been compared yet.

3.3 Type-II and direct Z-scheme heterojunctions

How well the carriers are used within the composite depends on the charge transfer path in the interface region. In type-II heterojunctions, carriers (electrons and holes) travel to different regions, thereby separating the carriers; however, this leads to reduced redox activity of the separated carriers (Xu *et al.,* 2018). The direct Z-scheme strategy has been proposed to overcome this issue in that the weaker carriers would recombine at the interface while the stronger carriers will remain in different regions (Xu *et al.,* 2018). A dual Z-scheme system of g-C_3N_4/$Bi_4Ti_3O_{12}$/$Bi_4O_5I_2$, for instance, involved both ofloxacin decomposition and H_2 evolution under visible light irradiation (Kumar *et al.,* 2021). Therefore, the partner material and the interface itself are crucial for perovskite composites.

3.4 S-scheme heterojunctions

The S-scheme model was developed in order to clarify the ambiguities in describing the process of charge transfer in the type-II and Z-scheme approaches. Like the Z-scheme approach, it seeks to maintain the carriers with the highest redox potential, and it provides a better understanding of the charge transfer path (Xu *et al.,* 2020). Currently, S-scheme-based hybrids have found applications in titanate-based materials; for instance, S-scheme-based $FeTiO_3$/g-C_3N_4 (iron titanate, and not perovskite) hybrid has been demonstrated for the degradation of tetracycline (Xu *et al.,* 2022). However, the mechanisms suggested for Z-scheme and S-scheme approaches are frequently disputed and require further verification.

3.5 Metal–organic frameworks as comparison materials

MOFs do not belong to perovskites; however, MOFs can be considered comparison materials because they were investigated in water purification and Cr(VI) photoreduction (García *et al.,* 2022). In terms of the Cr(VI) photoreduction literature, most studies are devoted to MOFs, whereas perovskite-based composites have not been extensively examined in relation to such pollutants (García *et al.,* 2022).

3.6 Critical comparison

The approaches mentioned above have a common target – to expand light response range and enhance charge separation (Patra *et al.,* 2023; Xu *et al.,* 2018). The main difference between the two strategies is in the way how they combine carrier separation and redox properties (Xu *et al.,* 2018; Xu *et al.,* 2020). Reviews dedicated to perovskites discuss either one class of perovskites or one particular issue (Clabel *et al.,* 2022; Nkwachukwu & Arotiba, 2021; Patra *et al.,* 2023), while,

as shown in Table 1, primary studies also have their individualities.

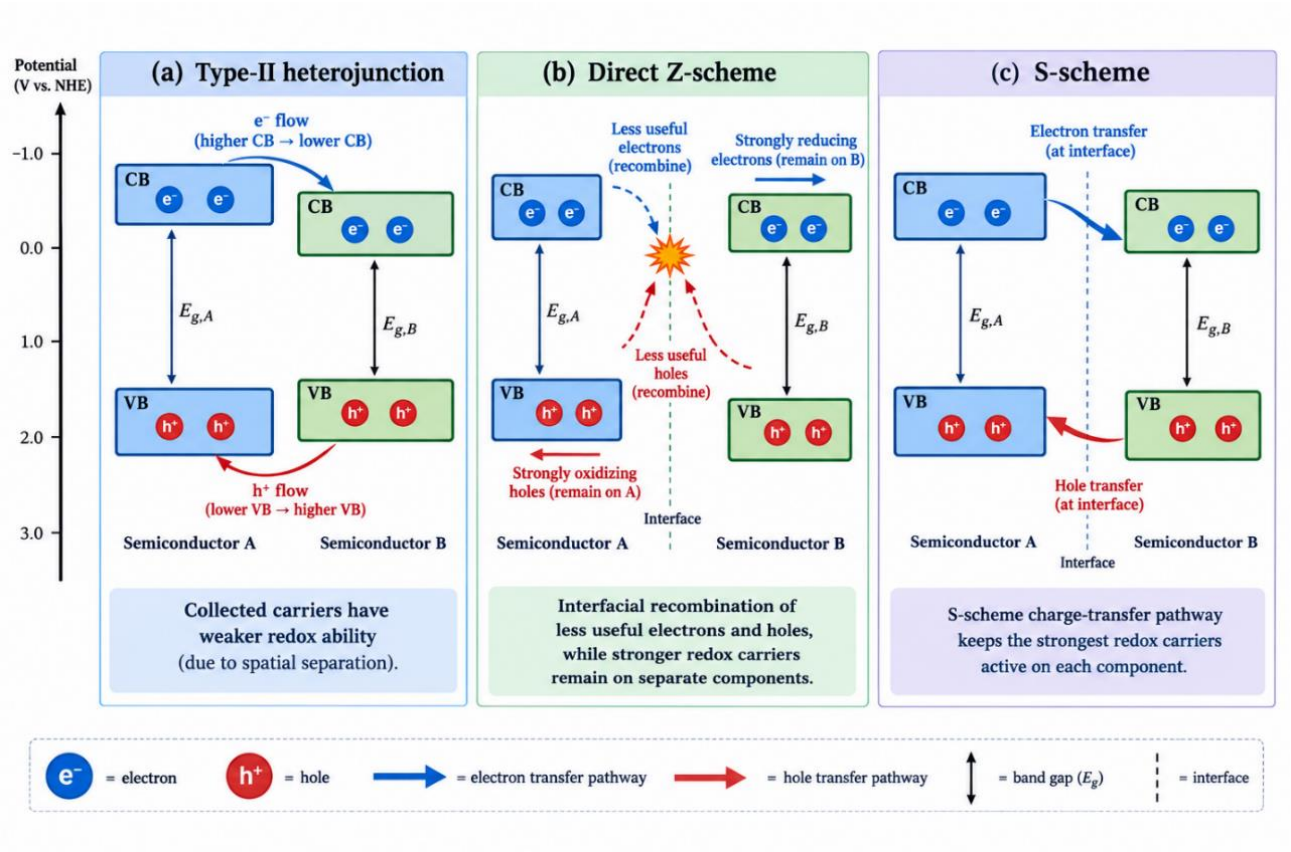


Figure 2: Schematic comparison of charge-transfer pathways in Type-II, direct Z-scheme, and S-scheme heterojunctions, highlighting the trade-off between carrier separation and redox ability

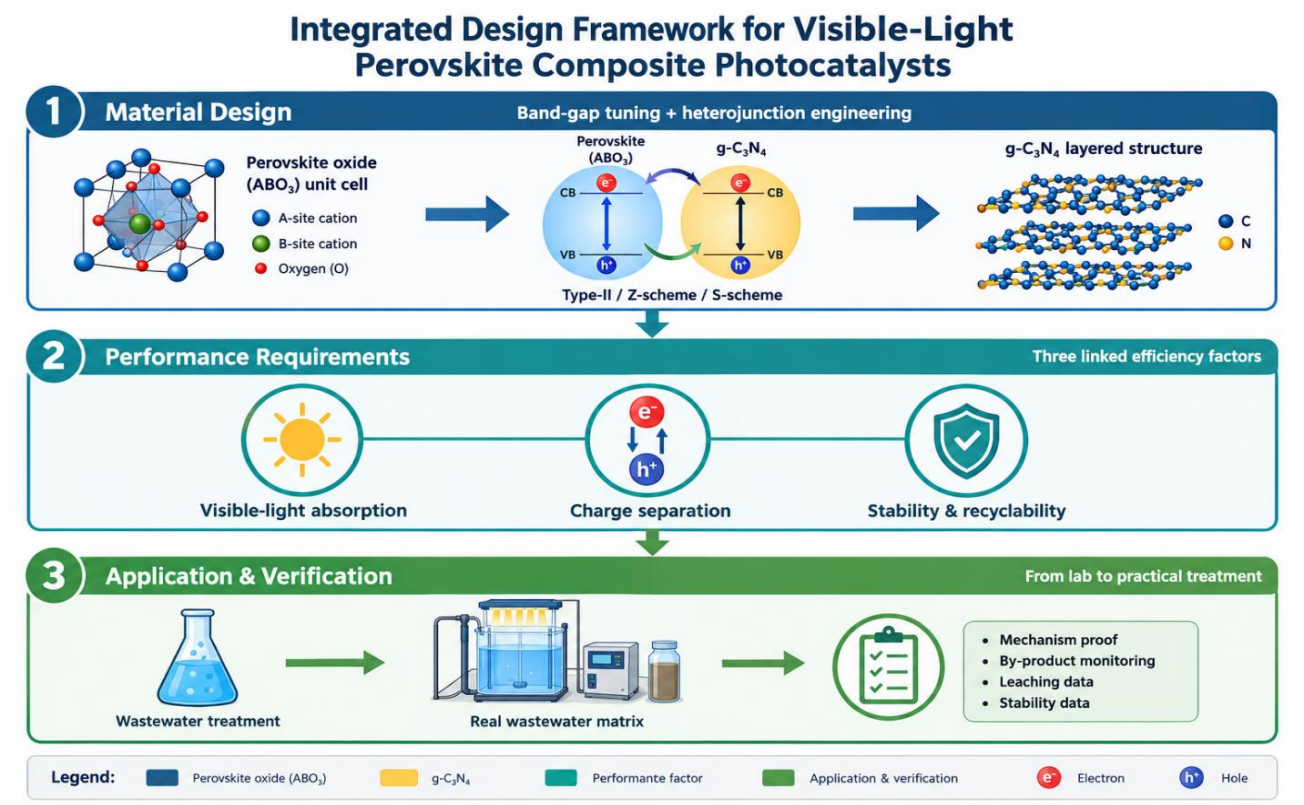


Figure 3: Integrated design framework linking material design, performance requirements, and application/verification for visible-light perovskite composite photocatalysts

4. Application to Wastewater Remediation

4.1 Organic pollutants and emerging contaminants

A visible-light-induced advanced oxidation process has been suggested as one of the technologies for removing emerging contaminants from water, such as pharmaceuticals and personal care products (Zawadzki, 2022). Operation at visible light is important because it enables using a wider range of the solar spectrum (Etacheri *et al.,* 2015; Zawadzki, 2022). Perovskite oxides have been used as catalysts in photocatalytic/photoelectrocatalytic water treatment (Nkwachukwu & Arotiba, 2021), and alkaline earth titanates have been reviewed as potential catalysts for water purification (Solís *et al.,* 2021). Perovskite zinc titanate obtained via the sol-gel process has been examined against emerging contaminants (Chen *et al.,* 2021). Studies with composites based on g-C_3N_4 have been carried out mainly for dyes and antibiotics (Table 1). TiO_2-based catalysts have been investigated, showing strong dependence of the pesticide and phenolic contaminant degradation on operating conditions (Ahmed *et al.,* 2011), thus, operating conditions must be reported.

4.2 Photoreduction of Cr(VI)

Photoreduction is used to eliminate Cr(VI) where electrons photogenerated reduce Cr(VI) to a more stable state (García *et al.,* 2022). Photocatalysis is appealing because it offers a solution to both organic compounds and toxic metals since one technique can be used to clean up both organic and metal contaminants (Schneider *et al.,* 2014), a strategy that was successfully implemented using a double perovskite/g-C_3N_4 photocatalyst for the elimination of both organic and inorganic contaminants (Kadkhodayan & Alizadeh, 2023). However, in the case of MOF, the formation of Cr(III) is never checked.

4.3 Comparing performance across studies

Comparison of pollutant removal from the literature is complicated. The primary studies apply various perovskites, partners, pollutants, and light sources (Table 1). Current reviews also focus on single classes or individual aspects of perovskite catalysts (Clabel *et al.,* 2022; Nkwachukwu & Arotiba, 2021; Patra *et al.,* 2023). Interpretation of published data may also be influenced by whether or not the suggested mechanism was validated (Xu *et al.,* 2020). For that reason, even the best results cannot be transferred to practical application as is.

5. Challenges and Future Perspectives

5.1 Visible-light response and recombination

The perovskites exhibit visible light response by altering band structure (Nkwachukwu & Arotiba, 2021); however, absorption is not sufficient for ensuring high activity. Band gap, carrier lifetime and the positions of band edges play critical roles in the performance of a material (Clabel *et al.,* 2022), while the loss of energy takes place due to recombination (Hoffmann *et al.,* 1995). The composites facilitate charge separation (Patra

et al., 2023) but cannot eliminate the problem of balancing these parameters.

5.2 Verifying charge-transfer mechanisms

Where a mechanism lacks credible supporting data, it becomes difficult to justify the superiority of one composite to the other and weakens rational design (Xu *et al.,* 2020). Although scavenger studies like those done by Konstas *et al.,* (2018) identify the reactive species involved, they cannot be used to justify the direction of electron flow on their own since complementary data is required. It becomes clear from the report of light source-dependent mechanism (Kim & Jo, 2019) that there is variability in the path based on testing conditions.

5.3 Complete reporting and by-product monitoring

In the MOF-based chromium (VI) removal research, there is no regular monitoring of the chromium (III) removal (García *et al.,* 2022), just as there must be in perovskite-based composites. Similarly, removal of the parent organic substance does not prove that its transformation products are benign. Therefore, all studies must provide the pollutant, its transformation products, and the working conditions (Ahmed *et al.,* 2011; García *et al.,* 2022; Zawadzki, 2022).

5.4 Stability, metal leaching and real effluents

The reality of wastewater involves the presence of a mixture of dyes, pesticides, pharmaceuticals, personal care products, and toxic metals (García *et al.,* 2022; Zawadzki, 2022); hence, tests should be performed using mixtures rather than individual compounds. The potential of photocatalysis-based water treatment technologies has been critically evaluated in terms of real conditions (Loeb *et al.,* 2019), and the advantages and limitations of traditional treatment serve as criteria that any new technology should reach (Crini & Lichtfouse, 2019). Stability, metal ion leaching to treated water, and catalyst separation should be studied systematically. The literature reviewed does not provide the data on these characteristics for perovskites composites, which is another gap.

5.5 Future perspectives

The following four areas need to be addressed. Firstly, the design of composites must rely on proven pathways of charge transfer and not on trial and error (Xu *et al.,* 2020). Secondly, results for reduced species and degradation products must be provided in full (García *et al.,* 2022). Thirdly, approaches employed for titanate-based composites (Patra *et al.,* 2023) have to be applied to ferrite-, lanthanum- and tantalum-based perovskites (Nkwachukwu & Arotiba, 2021) under similar conditions. Fourthly, promising materials have to be tested in effluent streams based on practical aspects like stability, leaching, and recovery. One possibility could be combining photocatalysts with adsorbents (Yahya *et al.,* 2018).

6. CONCLUSIONS

Perovskites based nanocomposites represent one of the best candidates for visible light wastewater treatment. Replacement of metal ions of a perovskite affects the optical properties of the material, while its combination with another material prolongs separation of charges necessary for the reaction. Advances in design of heterojunctions have allowed to go from type-II to Z-scheme and S-scheme designs with an attempt to keep only the most active charges. However, limitations discussed in the reviewed literature are mostly related not to the absence of efficient materials, but to the way of investigation and reporting. Literature review of either particular families of perovskites or specific approaches is not enough for a comprehensive comparison. The Z-scheme and S-scheme designs are controversial and require further verification. In Cr(VI) investigations, Cr(III) produced remains unreported, and therefore the efficiency of treatment of water cannot be assessed properly. The literature sources provide insufficient information on stability, metal leaching and wastewater treatment. In the future, scientists have to demonstrate the validity of the proposed mechanisms using better data, mention all products produced by the reaction and not just the initial pollutant, examine the same design for other types of perovskites in similar conditions, and test the optimal materials on actual wastewater. Only after these steps will perovskite photocatalysts become applicable to water treatment applications.

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