Chapter 7

Perovskites based Nano Heterojunctions for Photocatalytic Pollutant Removal

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Abstract

The development of new generation photocatalytic materials used for the betterment of human as well as environment. Perovskites and perovskites related nano-hetero-junction shows great interest for photocatalytic organic and inorganic pollutant removal. This chapter discusses its crystalline structures varying from cubic (high symmetry) to triclinic (very low symmetry). Various methods have been utilized for synthesis of perovskites such as sol-gel, hydrothermal, vapor deposition methods, solid-state reaction routes from oxide and high pressure technique. The first technique is used for the synthesis of perovskite is ceramic route in which the mixture of oxide was treated at high temperature and processed later by ceramic powder method. Various photocatalyst such as nitrides, sulphides, phosphides, oxide and mixed oxide are employed for photocatalytic water splitting or hydrogen generation. Future perspectives of perovskite-related photocatalyst are included in this chapter.

Keywords

Perovskites, Nano-Hetero-Junction, Sol-Gel, Hydrothermal, Ceramic Route, Hydrogen Generation

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1. Introduction

Various environmental problems such as water and air contamination and energy shortage are being faced by human being as well as aquatic life. Water pollution has grabbed much attention of environmentalists and researchers worldwide. Large quantities of organic pollutants were discharged into the surrounding soil and water by numerous industries [1]. Perovskite based photocatalysts have found great interest in energy and environmental applications. Perovskites was firstly discovered by German and Russian mineralogist Gustav Rose and Lev Perovski in 1839. Perovskite refers to the crystal structure of calcium

titanate and all compounds with the same crystal structure are perovskites. It has a general formula ABX_3 , where A is an organic cation, B is a metal cation and X stands for the halide anions [2]. For last few years, various experiments and investigation were done to study the perovskites ABX_3 . This type of structure enhance its importance in the field of electrical ceramics, refractories, material science [3], astrophysics [4], particles accelerated [5], heterogeneous catalysis [6] and environment [7] etc. Significant substitution of one or both A and B cation sites accept by the perovskite structured sites, which retained their original crystal structure. This type of process shows partial replacement of the cation sites with foreign metal ions, which modifying their electrical, structural, microstructural, and magnetic properties [8]. Perovskite-type oxides and perovskite-like oxides grasp attention in chemistry as well as in physics.

Perovskites have wide range of crystalline structures varying from cubic to triclinic. Various solid state methods are used for synthesis of perovskites such as sol-gel, hydrothermal, high-pressure technique and vapors deposition methods. Perovskite materials show various fascinating properties for both theoretical and application point of view. These compounds are used in fuel cells as sensors and catalyst electrodes and are candidates for memory devices and spintronics applications. Tanaka and Misono discussed in detail numerous advantages of perovskite photocatalysts [9] as listed follows:

- a) They are formed from a large variety of compositional and constituents elements, but their basic essential, basic structure are similar.
- b) Their well-defined bulky structures are characterized well and their surface properties are easily explored.
- c) Their valency, stoichiometry and vacancy can be varied widely.
- d) Physical and solid-state chemical properties have been accumulated [9].

Photocatalytic performance of the catalyst is affected by various factors such as electron and hole effective mass, diffusion length, exciton lifetime, exciton binding energy, electron-hole separation and transport within the lattice. Due to its distinctive crystal structure and electronic properties perovskite oxides show excellent results as a photocatalyst under visible light irradiation. The crystal structure of perovskite shows excellent band gap values and band edge potential which is useful in specific photocatalytic reaction. Separation of photogenerated charge carrier can be influenced by lattice distortions in perovskite. Numerous groups of perovskites such as vanadium-and niobium, titanate, tantalite and ferrites possess high visible-light activity.

The first technique used for perovskite synthesis was ceramic route in which mixture of oxides was treated at high temperature. The first perovskite $CaTiO_3$ was fabricated by sol-

gel route. Exceptional chemical tolerance properties were exposed by ion doped calcium titanate, which can be used for LEDs [10]. Also LaFeO_3 grabs more attention because of its narrow band gap due to this it is easily excited by visible light and decomposing organic molecule and water [11]. On the other hand LaFeO_3 shows good chemisorptions of nitrogen oxide (NO_x) [12].

BiFeO_3 shows excellent photocatalytic activity for dye degradation as well as water splitting under visible light irradiation [13]. In addition, it also shows visible-light-driven photo-Fenton degradation activity. Strontium titanate (SrTiO_3) is widely used for photocatalysis because of its tremendous structural stability, high photo activity and strong photo-corrosion resistibility [14]. It has wide band gap and low quantum yield which is overcome by formation of heterojunctions via coupling with two or more photocatalyst [15] or by adjusting band gap via doping or co-doping with cations [16, 17]. Rare earth based perovskites with composition LnFeO_3 (Ln = rare earth) shows various applications such as gases separators [18], sensors [19], catalyst [20] etc. Various groups investigated their photocatalytic activity because of their excellent photo degradation rate [21] and photo-Fenton like reactions [22].

Hybrid organic and inorganic perovskite (HOIPs) form a subclass of ABX_3 in which the replacement of A-site and X-site are done by the organic amine cations or organic linkers respectively. In HOIPs the organic component in this structure shows additional functionalities and structural flexibility that cannot be achieved by pure inorganic perovskites. Hybrid perovskite have a reported cubic phases of MAPbX_3 (MA = methylammonium and $\text{X} = \text{Cl, Br or I}$) [23].

The current chapter is focused on the structure, types, properties and synthesis of the perovskites. It also discusses the photocatalytic activity of the perovskites. Different methods are used for the synthesis of perovskites based nano-hetero assemblies. These are useful for organic and inorganic pollutant removal.

2. Perovskites

2.1 Structure

Generally, the structural formula of perovskite materials is ABX_3 and A_2BX_4 , where A and B are cations and X is the anion, by the formation of anionic bond A and B are binds together. A possesses larger ionic radius than the B. The structure of the perovskite is analyzed as follow: A is placed on edge, and at the center of octahedron B is located. The cationic radii are $r_A > 0.09 \text{ nm}$ and $r_B > 0.051 \text{ nm}$ [6]. Anionic X is represented as oxygen or non oxygen material such as fluorine ($-\text{F}$) and methyl ($-\text{CH}_3$).

Figure 1: Typical structure of Perovskite.

The structural distortion in perovskite is determined via electronic configuration of metal ions and the ratio of A and B ion size. In perovskites, there are two types of structural distortions, one is off-centering of B ion in BO_6 octahedral and another one is the tilting of BO_6 octahedral. The first type corresponds to displaced phase transition and the second corresponds to order-disorder phase transition [24, 25]. To identify the formability of perovskite structure the tolerance factor (t') was calculated as suggested by Goldschmidt [26].

$$t' = \frac{(r_A + r_0)}{\sqrt{2} \cdot (r_B + r_0)} \quad (1)$$

Where r_A and r_B ionic radii of A and B cation and r_0 ionic radius of oxygen anion. When $t' < 1$ gives the tilting mode and $t' > 1$ show centering of smaller B cation which causes tilting of BO_6 octahedron. Due to contraction of BO_6 octahedron there off-centering occur due to large A and smaller B ion. At room temperature the best ideal perovskite-type structure is cubic [27].

The majority of the perovskite material shows the band gap greater than 3 eV which are further modified its band gaps by the doping of elements, which can be used as high photocatalytic efficiency. Depending on the details of the octahedral rotations there is a transformation of crystal phase in ABX_3 perovskite materials which displayed lattice distortion in following succession such as orthogonal, tetragonal, rhombohedral,

monoclinic and triclinic phase, [26]. Some of the perovskite materials examples are given below:

Niobium based perovskites such as KNbO_3 having cubic, orthorhombic and tetragonal with band gap of 3.12-3.24 [28]. CuNbO_3 is a monoclinic crystalline structure with band gap 2.0 eV [29]. SrNbO_3 having cubic structure with band gap of 2.79 eV [30]. Iron based perovskites such as LaFeO_3 is a cubic and orthorhombic structure with a band gap of 2.1 eV [31]. Orthorhombic structure was shown by the bismuth based perovskites such as AgBiO_3 with 2.5 eV band gap [32]. Table 1 shows various perovskites with their band gaps and crystalline structure.

Table 1. Perovskites with their band gaps and crystalline structure.

Perovskites	E_g (eV)	Crystalline Structure	Reference
SrTiO_3	3.1-3.7 eV	Cubic	[33]
CaTiO_3	3.6 eV	Orthorhombic	[34]
PbTiO_3	2.75 eV	Tetragonal	[35]
MnTiO_3	3.1 eV	Rhombohedral	[36]
KTaO_3	3.6 eV	Cubic and orthorhombic	[37]
AgTaO_3	3.4 eV	Rhombohedral	[38, 39]
LaTiO_3	4.7 eV	Cubic and Rhombohedral	[28]
KNbO_3	3.12-3.24 eV	Cubic, tetragonal and orthorhombic	[29]
CuNbO_3	2.0 eV	Monoclinic	[30]
SrNbO_3	2.79 eV	Cubic	[31]
LaFeO_3	2.1 eV	Orthorhombic	[32]
AgBiO_3	2.5 eV	Orthorhombic	[40]

2.2 Types of perovskites

ABX_3 perovskite crystalline structure shows flexibility and its ability to accommodate a broad range of cation or anion vacancies. And show the origin of large variety of

perovskite-based compounds with a wide range of physical properties. The oxide phases have been divided into two types: one is ternary ABO_3 and its solid solutions and the another one is newer complex type compounds $(\text{A}^{x+}\text{B}^{y+})\text{O}_3$ where B' and B'' are two different element in different oxidation states and $x + y = 1$. On basis of oxidation states the oxides are classified into $\text{A}^{1+}\text{B}^{5+}\text{O}_3$, $\text{A}^{2+}\text{B}^{4+}\text{O}_3$, $\text{A}^{3+}\text{B}^{3+}\text{O}_3$ and oxygen and cation deficient species [41, 42]. The details are sorting in flowchart given below Figure 2.

The complex perovskite type compounds $\text{A}(\text{B}'_x\text{B}''_y)\text{O}_3$ divided into different compounds which enclose twice the lower valence state element then the higher valence state element $\text{A}(\text{B}'_{0.65}\text{B}''_{0.33})\text{O}_3$, those which have twice the higher state element then the lower state element $\text{A}(\text{B}'_{0.33}\text{B}''_{0.65})\text{O}_3$, and at the last on is when both B element in equal amount $(\text{B}'_{0.5}\text{B}''_{0.5})\text{O}_3$ and oxygen deficient phase $\text{A}(\text{B}'_x\text{B}''_y)\text{O}_{3-z}$. The valences of the A and B cations are 2^+ and 4^+ respectively. But in some cases if the B^{3+} has six coordinates their valences can be 3^+ . The bulking of the $(\text{AO}_3)^{4-}$ layers because of variation at the A cation position which causes distortion and displacement of the oxygen anion. Because of bulking, there is a distortion of octahedral when B cation took as a center.

The B cation has an ability of stiffness to endure this effect, and the transition metal oxide, because of its multi valency or the special 3d and 4d electronic configuration, fills the B cation position. Due to this reason, the transition metal oxides show extraordinary properties and perovskite type structure.

2.3 Distinctive properties of perovskites

Perovskite based materials are used for various applications such as sensors, piezoelectric, ultrasonic and under water devices, wastewater treatment, high temperature heating applications, frequency filters for wireless application etc. Depending on the application, perovskites can be synthesized in diverse forms such as bulk, nanocrystalline and thin films. There is a possibility to synthesize to multi component perovskite via partial substitution of cation in position A and B. They acquire several peculiar properties such as pyroelectric, polar properties, ferroelectric, optical properties and many more. Some of the detailed properties are explained below.

2.3.1 Optical properties

Perovskite is a class of materials having excellent optical and photoluminescence properties. Single domain crystal of BaTiO_3 shows optical properties studied by Merz at different temperature [43]. At temperature from 20°C to 90°C the crystal has a reflective index at constant value ~ 2.4 and reached at maximum value 2.46 at 120°C . Thickness of single crystal BaTiO_3 is 0.25-1 mm which transmits from $0.5\ \mu$ to $6\ \mu$. Higher adsorption was found for wavelength greater than $11\ \mu$ and weak adsorption at $8\ \mu$. Noland studied

that the single crystal strontium titanate show optical properties produced by a flame fusion [44]. Between $0.20\ \mu$ to $17\ \mu$ in wavelength the optical coefficient was measured. 70% of better transmission was measured between $0.55\ \mu$ to $5\ \mu$. This crystal shows 2.407 of reflective index at $5893\ \text{\AA}$.

Figure 2: Types of Perovskites.

In the paraelectric phase the electro-optic properties of KTaO_3 , BaTiO_3 and SrTiO_3 were measured [45]. The electro-optic coefficients of these perovskite are constant with temperature. The term of induced polarization was expressed when the distortion of the optical indicatrix varies from material to material. Last few years researchers show a great interest in materials for laser applications. One of the best deals is using perovskite laser host materials.

Perovskite type oxides phosphors are broadly used in field emission display (FED) and plasma display panel (PDP) devices because on the surface of the phosphors particles a adequately conductive to discharge electric charges [46].

Recently BaZrO_3 is well known environment friendly photoluminescence (PL) materials which is used to emit light easily in visible region and prepared easily at low cost [47].

2.3.2 Piezoelectricity

Piezoelectricity is the ability of some materials to generate an electric charge in response to applied mechanical stress. Piezoelectric ceramics is an important group of piezoelectric materials, of which PZT is an example [48]. They show perovskite crystal structure such as tetragonal/rhomohedral close to cubic. The general formula is $\text{A}^{2+}\text{B}^{4+}\text{O}_3^{2-}$ where A is large divalent metal ion i.e. barium along with lead and B denotes a tetravalent metal ion such as titanium or else zirconium. Various examples of piezoelectric materials are topaz, wood, quartz analogic crystal, Gallium orthophosphate (GaPO_4), Barium titanate, Lead zirconate titanate etc.

2.3.3 Superconductivity

Superconductivity is a phenomenon of zero electrical resistance and expulsion of magnetic flux fields taking place in certain material cooled below critical temperature [49]. Many important physical properties are demonstrated by the oxide perovskite. Perovskite type structure provides a tremendous support for superconductivity. One of the best examples of superconductivity perovskite is High T_c copper oxide [50]. Because of perovskite, various intermetallic compounds used as a superconducting material have been eclipsed.

Some of the best example of superconductor is Sodium, potassium, rubidium and cesium tungsten bronze. Superconducting transition in three samples of reduced strontium titanate was measured by Schooley and co-workers [51].

2.3.4 Multiferroicity

It is class of materials which exhibiting simultaneous ferromagnetic, ferroelectric and ferroelectric ordering. Among other multiferroic, bismuth ferrite (BiFeO_3), a rhombohedrally distorted perovskite, is acquiring great attention because of its ferroelectric order and anti-ferromagnetic order above room temperature [52]. In transition metals doped BaTiO_3 achieved multiferroic in new era, from experimental as well as theoretical studies [53]. These materials shows both physical and chemical properties which can be enhanced by extrinsic and intrinsic dopants such as transition metal cations doping, excess oxygen vacancies [54].

2.4 Synthesis

2.4.1 Sol-gel method

Various techniques of sol-gel are used for preparing perovskites such as alkoxide, alkoxide-salt and the pechini methods. Due to adaptability in preparing perovskites, pechini method is widely used. One of the greatest advantages of this method is high purity and homogeneity of perovskites structure coupled with an accurate control of the composition of final materials [88].

The flexibility of sol-gel route, principally the pechini method that delivers homogeneous solutions, helps the formation of perovskite structure and makes it very attractive. This method leads formation of pure crystal perovskite structure at 1000°C . For preparing perovskite materials this technique, is one of the most widely used and studied as the chemical routes.

2.4.2 Solid state reactions

Solid state reaction is one of the most conventional methods of synthesis of ceramic compounds. In this method the oxide powder, carbonate or salt are mechanically mixed, heated at high temperature around 1200°C (Figure 3). During crystalline grains this process is carried out for 8 to 24 hours so that the mobility of cation helps to form the perovskite structure [55]. The reaction occurs at the interface of the mixed solids from the bulk to the interface between the particles.

For the preparation of perovskite, the solid-state reaction is one of the oldest methods that we used. In literature, the availability of the experimental data is one of the advantages of this method. The properties of the perovskite are correlated with preparation or sintering conditions. Sintering effect helps perovskites to form crystal structure. For example $\text{BaCo}_{0.7}\text{Fe}_{0.2}\text{Ta}_{0.1}\text{O}_{3-\delta}$ sintered above 900°C resulted in pure perovskite structure, where unreacted materials remain at lower temperature [56].

Solid-state reaction has some limitations such as it related with the particle size and broad particle distribution of final material. This method shows the precursor powder particles undertake widespread milling as well as mixing. After sintering $\text{BaCo}_{0.7}\text{Fe}_{0.2}\text{Ta}_{0.1}\text{O}_{3-\delta}$ the grain sizes is approximately 1-2 μm were obtained [57]. To overcome this problem, the fine powder is synthesized by incessant milling at high rotation, which helps to avoid the external heating known as mechanochemical route [58]. Even with these disadvantages, solid state reaction is simple and fast method to synthesized perovskites.

Figure 3: Solid State Reaction.

2.4.3 Hydrothermal method

The hydrothermal method is carried out in autoclave at high-pressure i.e. 15 MPa. At high pressure, it is a combination of both temperature between the boiling point of the water and the material critical temperature. Basically, it is a sol-gel type route which gives a good control of particle size. The basic example of this method is CaTiO_3 synthesized at 15 atm and 150°C and further calcined at 1300°C [59].

Through the analysis of XRD pattern it was found that there is no impurities in the sample and the phases has a similar cell parameters. In case of specific material hydrothermal method is not necessary [59].

2.4.4 Co-precipitation method

Supersaturation condition was required by the co-precipitation methods in which solution with soluble cations are mixed with another solution called precipitation agent as shown in Figure 4. In co-precipitation method there is need to control some important parameter such as pH, mixing rate, temperature and concentration to reach desire physical properties such as morphology and particle size.

Figure 4: Co-precipitation method.

This method is used in preparing perovskites with good homogeneity and purity. This is used to control on the chemical reaction which is necessary to obtain compounds without deficiency of metal cation.

This method is used while preparing $\text{La}_{0.8}\text{Sr}_{0.2}\text{Co}_{0.6}\text{Fe}_{0.4}\text{O}_3$ (LSCF) by using metal hydrate precursors dissolved in water, where potassium hydroxide used as a precipitation agent [60]. This method is used for synthesis of nanostructure materials such as LSCF [61]. And sphere-like NdFeO_3 nanocrystals [61] with modified particle size and morphology.

There are various other methods which are used to prepare advanced ceramics that can also be used for perovskites materials such as spray and freeze drying, spray pyrolysis, microwave assisted synthesis and sintering, combustion synthesis and physical vapor deposition (PVD). These methods are not widely used, and they are under development for the preparation of perovskites materials.

3. Perovskites as a photocatalyst

Photocatalysis is a phenomenon that utilized the energy input from incident radiation and the catalytic properties to be carried or accelerate certain chemical reaction on the surface of the materials [62, 63] (Figure 5). From last many years various chemical reaction have been studied, which are useful for generation of energy and environment cleaning applications. To design and develop the new photocatalytic material we should understand the mechanism of photocatalytic reaction.

Figure 5: Basic principle of overall photocatalysis [64].

The perovskite crystals structure offers an excellent framework to acquire the band gap values to enable visible light adsorption and band edge potentials helps for precise photocatalytic reactions. The separation of photogenerated charge carrier is influenced by the lattice distortion in perovskite compound. Some groups of materials, which show visible light activity, are shown below.

3.1 Titanate based perovskite

To date, titanate perovskites have been mostly studied for photocatalytic applications. Most of the titanate perovskites show band gap (E_g) more than 3.0 eV and have excellent photocatalytic activity under UV radiation [65]. Titanate MTiO_3 ($M = \text{Sr, Ba, Ca, Mn, Co, Fe, Pb, Cd, Ni}$) show excellent photostability and corrosion. Calcium Titanate is one of the most widespread perovskites with band gap of 3.6 eV. Cu doped CaTiO_3 was widely used as a visible light-driven photocatalytic water deposition [66].

Under visible region, CoTiO_3 shows band gap 2.28 eV which has been studied for photocatalytic $64 \mu\text{mol g}^{-1} \text{h}^{-1}$ of O_2 evolution reaction was obtained without any co-catalyst [67]. The band gap of NiTiO_3 is around 2.16 eV and the light adsorption spectra show peak in visible region which correspond to crystal field splitting [68]. Under visible light irradiation degradation of nitrobenzene is done by the CaTiO_3 nanorods [68].

Under visible region FeTiO_3 having band gap 2.8 eV. $\text{FeTiO}_3/\text{TiO}_2$ are used for the photocatalytic degradation of 2-phenol under visible light irradiation. The separation of

electron hole recombination and hole capturing phase are done by TiO_2 [69]. The junction is shown in Figure 6.

The synthesized CdTiO_3 nanofibers having band gap 2.8 eV are studied for the degradation for rhodamine [70]. PbTiO_3 is visible active photocatalyst having band gap 2.75 eV. Composite of nano- TiO_3 and micro PbTiO_3 are studied for photocatalytic performance and effective charge separation [71]. The ferroelectric behaviour of PbTiO_3 shows electron-hole separation at interface. Under visible light irradiation MnTiO_3 shows improved charge separation and degradation of rhodamine B [36].

Figure 6: Energy band diagram of Type-B Heterojunction $\text{TiO}_2/\text{FeTiO}_3$ [69].

3.2 Ferrite perovskites

Most of the ferrite perovskite have native band gap under visible region. Under visible light irradiation LaFeO_3 attain 2.1 eV band gap and it is used for the degradation of pollutants as well as hydrogen evolution. Pt co-catalyst loaded LaFeO_3 fabricated by sol-gel method which show exceptional yield of hydrogen evolution under 400W tungsten light source [72] (Figure 7). LaFeO_3 shows better photocatalytic activity than Fe_2O_3 because of its comparative electron properties such as electron hole separation, mobility, photoexcited lifetime etc. Under visible light irradiation GaFeO_3 has been reported to show overall water splitting having yield 0.10 and 0.04 $\mu\text{mol.h}^{-1}$ [73]. YFeO_3 having band gap 2.43 eV and showed four times better photocatalytic activity than TiO_2 -P25 under visible light irradiation >400 nm for the degradation of RhB [74]. Ferrite based perovskite used for magnetic recovery of particles, which is useful in particle application.

Figure 7: Photocatalytic reactor for hydrogen generation studies [72].

3.3 Vanadium and Niobium based perovskites

Under UV irradiation Vanadium and Niobium based perovskites show excellent photocatalytic activity. Both KbNO_3 and NaNbO_3 have 3.14 eV and 3.08 eV band gap values respectively. Also the suitable modification of band structure as shown in Figure 8 results in effective visible light photocatalysis [75]. N-doped NaNbO_3 used for the degradation of 2-propanol under visible light irradiation [76]. Also, nitrogen doped KbNO_3 helps for water splitting and organic pollutant degradation [77].

Figure 8: Band structure of NaTaO_3 , KTaO_3 , NaNbO_3 , KNbO_3 [75].

AgVO_3 show both type of crystalline structure such as $\alpha\text{-AgVO}_3$ and $\beta\text{-AgVO}_3$ and having band gaps 2.5 and 2.3 eV respectively [78]. $\beta\text{-AgVO}_3$ shows superior photocatalytic activity than α -phase. Volatile organic compounds (VOCs) were degraded and O_2 evolution was done by AgVO_3 . $\beta\text{-AgVO}_3$ nanowires show tremendous photocatalytic performance in degradation of RhB [79]. Composite of $\text{AgBr}/\text{AgVO}_3$ show degradation of RhB [80]. The mechanism is shown in Figure 9.

Figure 9: Proposed mechanism of photocatalysis of the $\text{AgVO}_3@\text{AgBr}@\text{Ag}$ nanobelt heterostructure [80].

3.4 Oxynitride based perovskites

Perovskite type SrNbO_2N nanoparticles were constructed by solvothermal route by post-nitridation treatment. SrNbO_2N having narrow band gap of 1.94 eV exhibit high photocatalytic activity for H_2 evolution (H_2 release rate is $148.3 \mu\text{mol h}^{-1}\text{g}^{-1}$) and high degradation efficiency of methylene blue under visible light irradiation i.e. 100 % in 70 min [81]. Tantalum, niobium and titanium oxynitride perovskites have been reported as visible active photocatalysts for water splitting. Photocatalytic activity of LaTiO_2N was first reported by Domen *et al.* [82] for water splitting in the presence of methanol (e^- donor) and Ag^+ (e^- acceptor) and has been further investigated for acetone decomposition [83]. BaTaO_2N and CaTaO_2N are active in water splitting in the presence of methanol or I^- as electron donor [84]. CaNbO_2N was found active for H_2 and O_2 evolution from methanol and aqueous AgNO_3 respectively [85]. Table 2 shows various bare perovskites for utilization as photocatalysts.

Table 2: Bare perovskites photocatalysts and their utilization

Sr.No.	Photocatalyst/Dosage	Pollutants/ concentration	% removal	Source of irradiation	Ref.
1.	SrTiO ₃ , sulfur doped SrTiO ₃ , and Ag doped SrTiO ₃ (0.5 g)	Degradation of 4-chlorophenol (10 ppm)	99 % within 90 min for Ag doped SrTiO ₃	Under visible light	[86]
2.	PbTiO ₃ (50 mg)	Acid red, acid black (5ppm)	69.7 and 96.05 % in 90 min	Ultrasonic probe power of 100-600 W	[87]
3.	Ag-LaTiO ₃ (1.2 gL ⁻¹)	Atrazine (50 ppm)	100 % in 40 min	Visible light	[88]
4.	PbTiO ₃ (500 mg)	Methyl orange (500 mL, 10 mg/L)	91 % in 180 min	Xe lamp 500 W	[89]
5.	KNbO ₃ (0.5 g)	Rhodamine B (10 mg/L)	89 % in 120 min	Xe lamp 500 W	[90]
6.	KNbO ₃ (1 g/L)	Acid Red dye (5 mg/L)	69.2 % in 5 h	Ultrasonic irradiation (300 W)	[91]
7.	LaFeO ₃ 500 mg	Rhodamine B (10 mg/L)	100 % in 180 min	Visible light	[92]
8.	LaFeO ₃ (10 mg)	Diclofenac (20 mg/L)	98.2 % in 90 min	Visible light	[93]
9.	Erbium doped CaTiO ₃ (0.1 g)	Methylene Blue (10 mg/L)	Rate: 4.54 × 10 ⁻⁵ s ⁻¹ in 2.5 h	Hg-Xe lamp 300W	[94]
10.	AgBiO ₃ (0.1-0.5 g/L)	4-nitrophenol and E.coli (4-ml)	90 % in 5 h and 1 h	Metal halide lamp	[95]
11.	La(1-x) AxTiO ₃ . 5-δ (A= Ba, Sr, Ca) (0.1 g)	Congo red dye (100 ppm)	70.92 % in 60 min	Xe lamp 300 W	[96]
12.	Pt-CaTiO ₃ (0.5 g)	Photo-conversion of nitrobenzene to aniline (8.13×10 ⁻⁴ mol/L)	100 % in 60 min	Xe lamp 300 W	[97]
13.	MnTiO ₃ (0.005 g)	Methylene blue (1×10 ⁻⁵ M)	70 % in 240 min	Sunlight irradiation	[98]
14.	F-MnTiO ₃ (6 mg)	RhB (5×10 ⁻⁶ mol/L)	90 % in 240 min	Xe lamp 500 W	[36]
15.	SrTiO ₃ (20 mg)	CO ₂ reduction (10 mL)	26.4 μmol/g	Xe lamp 300 W	[99]
16.	Cr-SrTiO ₃ (0.1 g)	Cr(VI) reduction	100 % in .5 h	Hg lamp 500 W	[100]
17.	Ag-SrTiO ₃ (30 mg)	CO ₂ reduction (10 mL)	80.24 μmol/g	Xe lamp 300 W	[101]
18.	BaTiO ₃ (10 mg)	Tetracycline (20 mg/L)	96 % in 180 min	UV light	[102]

3.5 Other perovskite systems

There are many more perovskites photocatalyst such as compound of bismuth, cobalt, antimony and nickel having band gaps in visible region. Under visible light irradiation pentavalent bismuth perovskites are known to be active photocatalyst. Perovskite such as LiBiO_3 , NaBiO_3 , KBiO_3 and AgBiO_3 having band gaps of 1.63, 2.53, 2.04, 2.5 eV respectively for the degradation of organic pollutant [32]. NaBiO_3 shows better photocatalytic performance for phenol and methylene blue [32]. The growth of *Microcystis aeruginosa* was effectively restricting under solar light source with the help of AgBiO_3 photocatalyst [103]. LaNiO_3 has been used for degradation of MO (>400) [104]. AgSbO_3 has been used for degradation of MB, RhB and 4-chlorophenol [105]. $\text{Gd}_2\text{Ti}_2\text{O}_7/\text{GdCrO}_3$ nanoassembly act as a p-n junction photocatalyst [106]. GdCrO_3 are responsible for the visible light adsorption with band gap 2.5 eV [107]. Some of the bare perovskite systems are listed in Table 2.

4. Perovskite based nano-heterojunctions

Heterojunction are analysed as the n-n, p-p and p-n combination [108]. For improving optoelectronic conversion efficiencies, many efforts have been made towards the fabrication of many heterojunctions [109-112]. The heterojunctions are classified into four categories (shown in Figure 10):

- I. Semiconductor-semiconductor (S-S) heterojunction: SrTiO_3 based heterojunction such as $\text{SrTiO}_3/\text{TiO}_2$ [113], $\text{SrTiO}_3/\text{SrCO}_3$ [114], $\text{SrTiO}_3/\text{BiOBr}$ [115] and $\text{SrTiO}_3/\text{Bi}_2\text{O}_3$ [116] were synthesized for various photocatalytic applications.
- II. Semiconductor-carbon(S-C) heterojunction, in which carbon can be derivative of graphene oxide (GO) [117], reduced graphene oxides (rGO) [118], Single-walled or multi-walled carbon nanotubes [119] and carbon aerogels [120]. For example, due to its super flexibility, extremely high specific surface and excellent electrical conductivity the Graphene (GR) in heterojunction enhances the charge separation efficiency, reduce the e^- and h^+ recombination and grant a large interface heterogeneous reaction.

Figure 10: (a) Schematic diagram of semiconductor-based photocurrent generation, (b) Doped semiconductor, (c) S-S heterojunction, (d) S-C heterojunction, (e) S-M heterojunction, (f) M-C heterojunction [121].

- III. Semiconductor-metal (S-M) heterojunctions: Ag-LaFeO₃ nanoparticles (ALFO NPs) which match with p-type characteristics exhibited remarkably high selectivity to formaldehyde, by the modification with Ag the LFO enhanced the sensing performance. Ag NPs with localized surface plasmon resonance (LSPR) or SPR properties are well known to increase the visible-light absorption [122, 123]. The surface of the photocatalyst is modified with Ag, Au etc. not only enhances solar absorption but also facilitates surface electron excitation.
- IV. Multicomponent (MC) heterojunctions: Various MC Heterojunctions consisting of visible-light active components and an electron transfer system enhance visible light response and transfer of charge carriers. Metallic Ag deposited BiPO₄/BiOBr/BiFeO₃ (APBF) MC heterojunction was synthesized. An excellent wide spectrum photo-response gain by the plasmonic junctions is obtained which makes the best use of BiPO₄ which is otherwise a poor photocatalyst [124].

4.1 Synthesis of perovskites based heterojunctions

4.1.1 Synthesis of Ag₂S/BiFeO₃

Z-scheme Ag₂S/BiFeO₃ heterojunction was successfully synthesized by simple precipitation method. Ag₂S nanoparticle deposited over the BiFeO₃ particles inveterate by

the morphology and microstructure characterization to form $\text{Ag}_2\text{S}/\text{BiFeO}_3$ heterojunctions [125].

4.1.2 Synthesis of $\text{LaFeO}_3/\text{g-C}_3\text{N}_4$

The p-type LaFeO_3 and n-type $\text{g-C}_3\text{N}_4$ was synthesized by facile method to form p-n heterojunctions. The $\text{LaFeO}_3/\text{g-C}_3\text{N}_4$ heterojunction acquired higher photocurrent density than pristine $\text{g-C}_3\text{N}_4$ and LaFeO_3 [126].

4.1.3 Synthesis of $\text{Ag}/\text{Fe}_3\text{O}_4$ bridged $\text{SrTiO}_3/\text{g-C}_3\text{N}_4$

Z-scheme transfer and surface plasmon resonance effect of Ag augmented by iron oxide was analysed by well-constructed $\text{SrTiO}_3/\text{g-C}_3\text{N}_4$ junction bridged with $\text{Ag}/\text{Fe}_3\text{O}_4$ nanoparticles [127]. The synthesis route as shown in Figure 11 follows ultrasonication-photodeposition-volatilization-drying route.

Figure 11: Synthesis Scheme of $\text{Ag}/\text{Fe}_3\text{O}_4$ bridged $\text{SrTiO}_3/\text{g-C}_3\text{N}_4$ [127].

4.1.4 Synthesis of $\text{PbTiO}_3/\text{CQDs}$

$\text{PbTiO}_3/\text{CQDs}$ were fabricated by hydrothermal route. The Designing of highly-effective $\text{PbTiO}_3/\text{CQDs}$ binary junction possess various properties such as visible light driven photocatalyst, biocompatibility, eco-friendly response and good durability [128].

4.1.5 Synthesis of $\text{CoFe}_2\text{O}_4/\text{BaTiO}_3$

Novel multiferroic nanostructure of $\text{CoFe}_2\text{O}_4/\text{BaTiO}_3$ was fabricated via co-precipitation and sol-gel technique. The synthesis of multiferroic nanocomposites, designed to enhance the mechanical coupling between the phases magnetostrictive-piezoelectric core-shell type nanostructure [129].

4.1.6 Synthesis of $\text{Ag}@\text{BiPO}_4/\text{BiOBr}/\text{BiFeO}_3$

Metallic Ag deposited $\text{BiPO}_4/\text{BiOBr}/\text{BiFeO}_3$ ternary hetero-assembly were constructed by simple precipitation-wet impregnation-photo deposition method. Surface plasmon resonance effect of Ag helps in protection of high conduction band and Valence band in the three semiconductors [124]. The synthesis route is shown in Figure 12.

Figure 12: Synthesis scheme of $\text{Ag}@\text{BiPO}_4/\text{BiOBr}/\text{BiFeO}_3$ Kumar et al., 2019 [124].

4.1.6 Synthesis of $\text{AgBr}/\text{AgTaO}_3$

$\text{AgBr}/\text{AgTaO}_3$ nanocomposites were synthesized by hydrothermal route. It shows strong visible light adsorption, which enhanced photocatalytic performance then their bare counterpart [130]. Table 3 depicts various synthesis methods for perovskites based photocatalysts.

Table 3: Synthesis methods for various perovskites based photocatalysts.

Photocatalyst	Synthesis Method	Reference
Ag ₂ S/BiFeO ₃	Simple precipitation	[125]
Ag/Fe ₃ O ₄ bridged SrTiO ₃ /g-C ₃ N ₄	Hydrothermal	[127]
LaFeO ₃ / g-C ₃ N ₄	Facile Method	[126]
ZnTiO ₃ @TiO ₂	Hydrothermal	[131]
PbTiO ₃ /CQDs	Hydrothermal	[128]
CoFe ₂ O ₄ /BaTiO ₃	Co-precipitation and sol-gel technique	[129]
BiVO ₄ @ β-AgVO ₃	Hydrothermal	[132]
AgBr/AgTaO ₃	Hydrothermal	[130]
Ag@BiPO ₄ /BiOBr/BiFeO ₃	precipitation-wet impregnation-photo deposition method	[124]

4.2 Photocatalytic organic pollutant removal

Now days various environmental problems are facing by human being correlated to hazardous wastes, polluted ground water, and toxic air contaminants [133, 134]. Many industries such as textiles, dyeing, printing, food and cosmetics use dyes, which discharges a large amount of organic pollutant into land, water which show adversely effect on environment as well as human health [135-137]. By considering the toxicity and wide range of organic contaminants various methods have been developed such as physical separation/transfer [138] and chemical degradation [139]. Under visible light irradiation Cr-doped SrTiO₃ and Ag₃PO₄ heterostructured photocatalyst effectively work. The junction was used for the degradation of Isopropanol under visible light irradiation for evaluating their photocatalytic properties. 97% of degradation of isopropanol was observed within 3.8 h of irradiation [140].

The high photocatalytic active BiFeO₃ synthesized by the solid-state reaction for the degradation of methylene blue (MB) under visible light irradiation. The catalyst has narrow band gap energy of 2.1 eV, high surface area with hollow structure, high crystallization of

perovskite-type BiFeO_3 . The promotion of separation of photo-induced electron and holes led to near about 86% degradation in 180 min of irradiation [141].

CaTiO_3 NPs are used as a highly efficient photocatalyst for the removal of arsenite from aqueous solution. CaTiO_3 NPs enhance the photocatalytic activity towards the oxidation of As(III) up to 98.4% within 40 min of irradiation [142].

Under ultraviolet irradiation, photocatalytic decomposition of methylene blue over $\text{CoFe}_2\text{O}_4/\text{BaTiO}_3$ (CFO/BTO) based nano-composite was reported. The synthesized CFO/BTO showed excellent photocatalytic activity for the degradation of methylene blue (MB) which is 99.3% within 90 min in aqueous solution under UV light [143]. The specific surface area of synthesized ZTCN junction was 1.69 times that of pure g- C_3N_4 with 76% degradation efficiency for methyl orange (MO) under visible light irradiation within 180 min, about 13 times the pure ZnTiO_3 and three times pure g- C_3N_4 . The complete degradation of MB and RhB under visible light irradiation within 90 min [144].

The photocatalytic activity of KBiO_3 showed 100% discoloration of MB within 120 min KBiO_3 obtained by the sonochemical method and 67% degradation of CPF was obtained by KBiO_3 synthesized by hydrothermal method [145]. The influence of the enhanced charge separation was displaced in the photocatalytic reaction. Under visible light illumination $\text{AgNbO}_3/\text{g-C}_3\text{N}_4$ photocatalyst showed high photocatalytic H_2 -generation [146].

The highly photocatalytic activity for the degradation of Norfloxacin (NFN) was shown by the well-designed $\text{Ag@BiPO}_4/\text{BiOBr}/\text{BiFeO}_3$ under different light sources within 45 min. Under visible light, the photocatalyst show highest 99.1% of degradation of NFN. Under near infrared 30.2% degradation is achieved in 120 min, this is due to presence of Ag which improves the light adsorption [124] (Figure 13).

The highly photoactive $\text{SrTiO}_3/(\text{Ag}/\text{Fe}_3\text{O}_4)/\text{g-C}_3\text{N}_4$ (SFC) nano-hetero-assembly was used for photocatalytic hydrogen production and photodegradation of levofloxacin (LFC) under different light sources (Figure 14). SFC-3 (30wt% g- C_3N_4 and 3% $\text{Ag}/\text{Fe}_3\text{O}_4$) shows H_2 evolution of $2008 \mu\text{mol g}^{-1}\text{h}^{-1}$ which is 14 times than the single g- C_3N_4 . Under visible light irradiation there is 99.3% of LFC degradation in 90 min [127].

The synthesized photocatalyst show high photocatalytic activity for the degradation of malachite green solution. 4.75% of $\text{NaBiO}_3/\text{Bi}_2\text{O}_3$ shows best photocatalytic activity which is approximately 95.70% of malachite green degradation after 300 min [147].

Water pollution is also caused by the discharge of inorganic cations in the water bodies. Cr^{+6} causes many disease such as epithelial irritant and human cancer inducer [148, 149]. Therefore various approaches were developed for efficient removal of pollutants from

water such as adsorption [150] and photocatalysis [151, 152]. The example of binary and ternary perovskite-based photocatalyst listed in Table 4.

Figure 13: Degradation of Norfloxacin by using Ag@BiPO₄/BiOBr/BiFeO₃ photocatalyst [124].

Figure 14: Degradation of levofloxacin by using Ag/Fe₃O₄ bridged SrTiO₃/g-C₃N₄ photocatalyst [127].

Table 4: Binary and ternary perovskites based photocatalyst and their utilization.

Sr.No	Photocatalyst/ Dosage	Pollutants/Concentration	% Removal	Source of irradiation	Reference
1.	BiVO ₄ /SrTiO ₃ (0.05 g)	Sulfamethoxazole (10 mg/L)	91 % in 60 min	Xe lamp 500 W	[153]
2.	Ag ₃ PO ₄ /PANI/Cr: SrTiO ₃ (50 mg)	Rhodamine B (RhB) (20 mg/L) and Phenol (25 mg/L)	100 % in 10 min and 100 % in 18 min	Xe lamp 300 W	[154]
3.	SrTiO ₃ /BiOI (50 mg)	Methyl orange (MO), (10 mg/L) Oxytetracycline hydrochloride (20 mg/L)	94.6 % in 40 min 85 % in 90 min	Xe lamp 300 W	[155]
4.	SrTiO ₃ /mpg-C ₃ N ₄ (0.3 g/L)	Basic violet 10 (10 mg/L)	80 % in 120 min	Xe lamp 300 W	[156]
5.	CoFe ₂ O ₄ /SrTiO ₃ (0.1 g)	Azo dye (500 ppm)	98.13 in 120 min	Xe lamp 300 W	[157]
6.	Ag/Fe ₃ O ₄ bridge SrTiO ₃ /g-C ₃ N ₄ (0.2 g/L)	Levofloxacin (10 mg/L)	99.3 % in 90 min	Visible light (500 W Xe) UV (500 W Hg lamp) and NIR	[127]
8.	BiFeO ₃ /CuWO ₄ (0.01 g)	Methyl Orange and RhB (50 mg/L)	87 and 85 % in 120 min	LED lamp 5 W	[158]
9.	BiFeO ₃ /CuBi ₂ O ₄ /BaTiO ₃ (20 mg)	Norfloxacin (10 mg/L)	93.2 % in 60 min	Visible light	[159]
10.	g-C ₃ N ₄ /rGO/ BiFeO ₃ (0.5 g)	Chromium (5 mg/L)	100 % in 90 min	Xe lamp 300 W	[160]
11.	SrTiO ₃ /(BiFeO ₃ @ZnS) (1 g/L)	2,4-dichlorophenol (10 mg/L) and Cr(VI) (10 mg/L)	94.32 and 97.8 % in 180 min	Visible light and Xe lamp 300 W	[161]
12.	Ag-AgVO ₃ /g-C ₃ N ₄ (0.04 g)	Tetracycline (0.015 g)	83.6 % in 120 min	Xe lamp 300 W	[162]
13.	Graphene quantum dots/ AgVO ₃ (0.01 g)	Ibuprofen (10 mg/L)	90 % in 120 min	Xe lamp 350 W	[163]
14.	Ag/AgVO ₃ / BiOCl (50 mg)	Methylene blue (7 mg/L)	93.1 % in 60 min	Xe lamp 300 W	[164]
15.	β-AgVO ₃ /ZnFe ₂ O ₄ (24 mg)	Methylene blue (60 mg/L)	87 % in 60 min	Xe lamp 300 W	[165]
16.	Pt/AgVO ₃ nanowires(0.05 g)	Atrazine (100 ppm)	99 % in 60 min	UV-Visible light	[166]
17.	ZnO/AgVO ₃ (20 and 50 mg)	Ciprofloxacin and chromium (10 and 20 mg/L)	90 % in 90 and 45 min	Visible light	[167]

18.	Ag/AgVO ₃ /g-C ₃ N ₄ (0.1 g)	Ciprofloxacin (10 ppm) and hydrogen production	82.6 % and 3.75 mmol/h H ₂ production	Xe lamp 300 W and 500 W Hg lamp	[168]
19.	MoS ₂ /AgVO ₃ (50 mg/L)	Rhodamine B (10 mg/L)	83.3 % in 120 min	Xe lamp 300 W	[169]
20.	GO-Ag ₂ O/Ag ₃ VO ₄ /AgVO ₃ (0.15 g)	Rhodamine B (15 mg/L) and MO (15 mg/L)	99.2 and 92 % in 45 min	Visible light	[170]
21.	CaTiO ₃ /rGO/NiFe ₂ O ₄ (0.1 g)	Methylene orange (5 mg/L)	98.3 % in 90 min	Xe lamp 200 W	[171]
22.	CaTiO ₃ /g-C ₃ N ₄ (25 mg)	Rhodamine B (1×10 ⁻⁵ M)	97 % in 60 min	Xe lamp 300 W	[172]
23.	PbTiO ₃ /CQDs (0.05 g)	Rhodamine B (5×10 ⁻⁵ M)	100 % in 120 min	Xe lamp 300 W	[128]
24.	KTaO ₃ /FeVO ₄ /Bi ₂ O ₃ (1 g/L)	Ceftriaxone sodium (10 mg/L)	81.30 % in 90 min	Visible light	[173]

Conclusion

Perovskites and perovskites related structures propose a wide range of materials to design novel photocatalyst for energy and environmental applications. The doping and substitution of A- and B- sites as well as the O sites are utilized for reducing band gaps to prepare visible and solar active photocatalysts. The formation of multicomponent heterojunctions is an approach to improve photocatalytic activity for harvesting solar energy and charge carrier separation. The performance of the perovskite and perovskite related photocatalysts gets improved by tailoring of their particle size, crystallinity and morphology. All the physicochemical properties provide perovskites and perovskites-related materials for various applications such as potentials for organic/inorganic contaminants removal, drug delivery, catalyst in modern chemical industry, sensor etc. These represent an alternative class of materials with hybrid character for visible and solar light assisted photodegradation of pollutants and clean energy production.

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