

Review

Bismuth oxyhalide layered materials for energy and environmental applications



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ARTICLE INFO

Keywords:

Layered materials
Bismuth oxyhalides
Photocatalytic
Solar energy conversion
Structure tuning

ABSTRACT

Photocatalytic solar energy conversion is considered one of the most promising pathways to address the global energy shortage and environmental crisis. Layered bismuth oxyhalides are a new class of photocatalytic materials with a strong light response to boost solar energy conversion due to their appealing energy band structure and unique layered structure. This critical review summarizes recent progress in designing and tuning new bismuth oxyhalide materials to boost solar energy conversion. We start with methods to prepare and tune bismuth oxyhalides to enhance photocatalysis: structural engineering *via* control of the bismuth-rich state, elemental doping, defect control, interface engineering, solid solutions, inner coupling, and heterojunction construction. Then advancements in versatile photocatalytic applications of bismuth oxyhalide-based photocatalysts in the areas of oxygen evolution, hydrogen evolution, CO₂ reduction, nitrogen fixation, organic syntheses, disinfection, and pollutant removal are discussed. Finally, the major challenges and opportunities regarding the future exploration of bismuth oxyhalide-based materials in photocatalysis are presented. The present review will deepen understanding regarding bismuth oxyhalides and open new directions in designing and optimizing advanced bismuth oxyhalide-based materials for energy and environmental applications.

1. Introduction

Photocatalysis is regarded as one of the most promising approaches for solving energy and environmental issues by the efficient utilization of solar energy [1–4]. Hydrogen (H₂) can be prepared through water splitting, and carbon-based fuels can be obtained by the photoreduction of CO₂ with the consumption of only inexhaustible solar energy. The key requirement for achieving a high-efficiency photocatalytic process is the rational design of efficient photocatalysts [5–9]. Two-dimensional (2D) layered materials with suitable bandgaps may represent ideal photocatalytic materials to address the energy issue [10]. 2D layered materials such as nitrides (e.g. h-BN, g-C₃N₄) [11,12], dichalcogenides (e.g. MoS₂, WSe₂) [13], trichalcogenides (e.g. TiS₃, In₂S₃) [14], monochalcogenides (e.g. GeSe) [15], thiophosphates (e.g. FePS₃, NiPS₃) [16], and halides (e.g. InBr₃, VI₂) are of great interest owing to their extensive technological promise and potential applications ranging from electronics to catalysis [17–21]. These layered materials are composed of continuous layers of covalently bonded atomic-layer planes, separated

successively by *van der Waals* gaps. Bismuth oxyhalides, BiOX (X = Cl, Br and I), a new class of promising layered materials for photocatalytic energy conversion and environmental remediation, have been intensively investigated [22–24]. They belong to the family of main-group multicomponent metal oxyhalides V-VI-VII, which crystallize in a tetragonal matlockite (PbFCl-type) structure. They are chemically stable, nontoxic, and corrosion-resistant. The crystal structure of BiOX is built by interlacing [Bi₂O₂] slabs with double halogen slabs to form a layered structure (Fig. 1). The intralayer atoms are connected by strong covalent bonding, and a weak *van der Waals* interaction exists among the interlayers. The open-layer crystalline structure enables enough space to polarize the related atoms and orbitals to form inherent internal static electric fields along the crystal orientation perpendicular to the [Bi₂O₂] and [X] slices [25]. The static electric fields that formed can promote effective separation of photogenerated electron-hole pairs, a key factor in photocatalysis that is responsible for the superior photocatalytic activity of BiOX [26]. Density functional theory (DFT) calculations show that the valence band (VB) maximum of BiOX primarily

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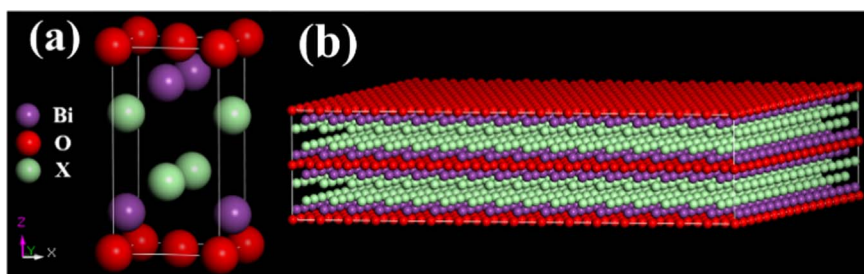


Fig. 1. Structural model illustrations of BiOX (X = Cl, Br, I): (a) single crystal cell, (b) two-dimensional layered structure with a two-layer thickness.

consists of hybrid O 2p orbitals and X np ($n = 3, 4,$ and 5 for X = Cl, Br, and I) orbitals, whereas the conduction band (CB) minimum mainly consists of Bi 6p orbitals [27].

Unlike TiO_2 , which is active only in the ultraviolet (UV) region, BiOX possesses a variable bandgap of ~ 3.3 eV for BiOCl, 2.7 eV for BiOBr, and 1.8 eV for BiOI, enabling it to be a type of light-active photocatalyst. The unique characteristics of BiOX make it a very promising photocatalyst for various applications. Since Zhang's group [28] reported 3-dimensional (3D) hierarchical BiOX microspheres assembled with 2D nanoplates *via* the ethylene glycol (EG)-assisted solvothermal method for the effective degradation of methyl orange (MO), BiOX-based photocatalysis has become a hot research topic [29]. To date, various BiOX micro/nanostructures have been prepared, such as nanocrystals, nanowires, nanofibers, nanobelts, nanosheets, nanoplates, thin film, flower-like hierarchical structures, hollow microspheres, and porous nanospheres [30–51]. To enable potential industrial applications, numerous attempts have been made to further improve the properties of BiOX by increasing the light harvesting, promoting the separation and transfer of photogenerated charge carriers, and boosting the catalytic efficiency between the active species and other substances in BiOX materials. The obtained BiOX materials display outstanding performance for photocatalytic applications such as pollutant removal [28], oxygen evolution [52], hydrogen evolution [53], CO_2 reduction [54], nitrogen fixation [55], selective organic synthesis [35], and disinfection [56].

Herein, we present a comprehensive overview of recent advances in the design, regulation, and energy and environmental applications of BiOX-based materials. We start with a synthetic method of making new BiOX nanostructures. Then, regulating approaches for tuning the photocatalytic performance of BiOX-based materials are thoroughly discussed, ranging from a bismuth (Bi)-rich strategy, to elemental doping and defect control, interface engineering, solid solution and inner coupling, and heterojunction construction. This review also introduces different applications of BiOX-based materials in the fields of photocatalytic oxygen evolution, photocatalytic hydrogen evolution, CO_2 photoreduction, photocatalytic nitrogen fixation, photocatalytic organic syntheses, photocatalytic disinfection, and photocatalytic removal of pollutants. Finally, concluding remarks and perspectives on the future exploration of BiOX-based materials are presented.

2. Synthetic methods for BiOX-based materials with engineered structure

The main approaches to making BiOX micro/nanostructures include the hydro/solvothermal [57,58], precipitation [59], calcination [60], reverse microemulsion [29], microwave [61], template [30], and sonochemical methods [62]. Various BiOX micro/nanostructures such as 1D nanorods/wires, 2D nanoplates/sheets, and 3D hierarchical architectures have been prepared using these methods. Generally, BiOX materials are inclined to form a 2D nanosheet structure owing to the inherent layering characteristic of crystalline structures. The 3D hierarchical architectures are always assembled from 2D nanosheets that form a porous and hollow structure. Taking the widely used Bi $(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$ as the Bi source as one example, during the formation

process, a Bi^{3+} cation tends to react with H_2O to yield BiONO_3 and H^+ . When BiOX materials are formed in polyhydric alcohols (e.g. ethylene glycol), the Bi^{3+} cation can coordinate with EG to yield alkoxide complexes and H^+ at the initial stage. As a result of the strong binding interaction between the H^+ cation and oxygen, the (001) surface with a high density of terminated oxygen atoms is capped by H^+ , and the crystal growth along the c axis is inhibited, leading to a (001) facet exposed BiOX crystal. Taking into account the unique crystal structure of BiOX, it is desirable to reasonably tune the exposed crystal facets or control the number of layers of the nanosheet structure to make assembly possible. Herein, we emphasize three effective strategies for making high-performance BiOX nanostructures: crystal facet tailoring, ultrathin structure control, and porous and hollow structure assembly.

2.1. Crystal facet tailoring of BiOX nanostructures

Crystal facet tailoring is an effective method of tuning/optimizing the photocatalytic properties of semiconductor materials. Adjusting the specific surface configuration and atomic structure enables effective regulations of the electro- and photo-catalytic properties of different exposed facets. Exposed facets with higher surface energies support higher catalytic activity. However, during the process of crystal growth, facets with high surface energy often disappear [63]. To make crystals with a large percentage of exposed reactive facets, capping agents, such as organic surfactants and fluorine ions, are typically used. For example, TiO_2 crystals with different exposed crystal facets can be tailored by fluorine ions [64].

The preparation of BiOX with specific exposed crystal facets was realized using H^+ as a capping agent [23]. BiOCl nanosheets with exposed (001) facets were simply synthesized *via* a hydrothermal process by using $\text{Bi}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$, KCl as a reactant and H_2O as a solvent without any additional agents. As shown in Fig. 2a and b, an individual nanosheet was produced with the angle at 45° in the selected area electron diffraction (SAED) pattern, which is consistent with the theoretical value of the angle between the (110) and (200) planes, suggesting the exposure of (001) facets. A high-resolution transmission electron microscope (HRTEM) image (Fig. 2c) shows an interplanar lattice spacing of 0.275 nm, corresponding to that of a (110) facet, which reveals the (001) facet exposure based on the symmetries of tetragonal BiOCl (Fig. 2d–f). In aqueous solution, $\text{Bi}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$ reacted with H_2O to yield BiONO_3 and H^+ , then the Cl^- reacted with BiONO_3 to form the Cl-Bi-O-Bi-Cl layer. Because the self-generated H^+ built a strong binding interaction with the oxygen atoms in the exposed facets, (001) facets with a high density of terminated oxygen atoms were induced. Thus, BiOCl nanosheets dominated by (001) facets were produced. Adjusting the pH value to 6 allowed (010) facet-dominant BiOCl nanosheets to predominate. Through the addition of OH^- ions to neutralize the H^+ ions, a thermodynamically stable (010) faceted surface with low surface energy was achieved. Under UV-light irradiation, (001) facet-dominant BiOCl nanosheets displayed higher photocatalytic activity than (010) facet-dominant BiOCl nanosheets for the degradation of MO by direct semiconductor photoexcitation. However, (010) facet-dominant BiOCl nanosheets displayed higher activity than did (001) facet-dominant BiOCl nanosheets under visible-light irradiation *via* indirect dye

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