



A Review of BiOBr-Based Photocatalysts for Wastewater Treatment

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<https://doi.org/10.18280/ijht.420123>

ABSTRACT

Received: 30 December 2023

Revised: 24 January 2024

Accepted: 29 January 2024

Available online: 29 February 2024

Keywords:

wastewater treatment, photocatalytic process, BiOBr synthesis, BiOBr characteristics, BiOBr photo-degradation activity

Semiconductor based photo-catalysts which was an efficient process to treat water and wastewater. There are various techniques can be used for enhancement of photo-catalytic properties such as element rich strategy, defect control and facet engineering. Many methods were used for manufacturing of BiOBr, including ion-exchange method, solvothermal method, wet-chemical method, ultra-sonication method, co-precipitation method and hydrothermal method. Various operational parameters have such as initial pH of solution, catalyst dosage and inorganic ions have been employed to show their roles on the photo-degradation efficiency of pollutants. Elemental doping and coupled semiconductors are the widely using methods for enhancing BiOBr performance. It was concluded that the binary and ternary composites have the ability to enhancement the photo-catalytic activity and increased the degradation efficiency more than 50% compared with pure BiOBr. According to suitable band structure, bismuth bromide oxide is a promising candidate to treat wastewater efficiently by photo-catalytic technique.

1. INTRODUCTION

Industries and human activities have been responsible about air, water and land contaminations. Environment pollution particularly, water pollution has increased due to rapid increase in the population, which is a major threatens of the humanity existence [1, 2]. Wastewater or polluted water contains various pollutants such as organic, inorganic pollutants and pharmaceuticals [3-6]. Wastewater contaminated with dyes is highly hazardous for biological health and ecosystem, and the water bodies should be treating from these pollutants. In developing countries, waterborne pathogens such as bacteria and viruses were responsible for 80% of illnesses including giardiasis, diarrhea, typhoid fever, dysentery and salmonellosis [7]. It is necessary to kill these microorganisms and remove other pollutants to obtain drinking water [8]. However, various methods such as electrochemical reduction [9], membrane filtration [10-15], precipitation [16], electro-dialysis [17], photo-catalysis [18, 19] and electro-deionization [20] have been used for removing various pollutants and treatment of wastewater [11, 12]. The major disadvantages of these technologies except photo-catalysis are complicated process, large intake of energy, by-products formation and wastes, low removal efficiency and expensive so these technologies are not preferred in wastewater treatment. Photo-catalysis is an effective and green process, which is the best treatment process due to its cost-effectiveness and high efficiency; it has obtained the chemical energy from sunlight energy. Photocatalysis can be used in various applications like dye removal, microbial inactivation and eliminating pollutants from air, etc. [18].

By a series of reactions, photo-catalysis process has the ability to convert complex pollutants into very simple and harmless molecules. So, this technique has been an environment friendly and economically [21]. For current research and development, one of the common techniques for wastewater management is a nanotechnology [22].

Bismuth was discovered in the 1660s which is a white substance, it has an atomic mass of 208.98 [23-25]. Many important Bismuth ores are Bi_2S_3 and Bi_2O_3 [26]. Unlike many of heavy metals which are carcinogenic, highly toxic and cause fragility of bones, failure of kidney and lung damaging [27-32], bismuth is non-carcinogenic and non-toxic [33], bismuth is less toxic than table salt, which is regarded as a green element [27-29]. For long time, bismuth has been employed in several applications as pigments, cosmetics and medicines [30, 31], as well as using of bismuth vanadate in paints as a pigment [32]. In this aspect, many researches have been used bismuth in several applications as a replacement for toxic lead [34].

BiOBr has small band-gap ($E_g=2.64-2.91$ eV) [35-37]; so, it has the ability for maximum visible sunlight energy harvesting photo-catalyst [38]. Bismuth is non-toxic and chemically stable as well as cheap [39-41]. There are several shapes of BiOBr, including nanoflowers [42], nanobelts [43] and nanoflakes [44] and nanospheres [45] which have been manufactured by several methods like solvothermal [43], hydrothermal [46], ion thermal [47], and co-precipitation [48]. Now, bismuth is being used for various purposes, such as photo-catalytic wastewater treatment [49], water splitting [42], indoor-gas purification [50] and alcohol selective oxidation [51].

Bismuth bromide oxide belongs to BiOX ($X=\text{I, Br, Cl, F}$)

family, which crystallizes with layered tetragonal matlockite structures [52]. BiOX compounds are characterized by good magnetic and electrical properties, which have been used in various fields such as photo-chromic devices, solar cells, ferroelectric materials and pigments [53, 54].

In each layer, four atoms of halogen surround the bismuth center (having weak interlayered interactions) and four atoms of oxygen (having strong covalent bonds) [55], as shown in Figure 1.

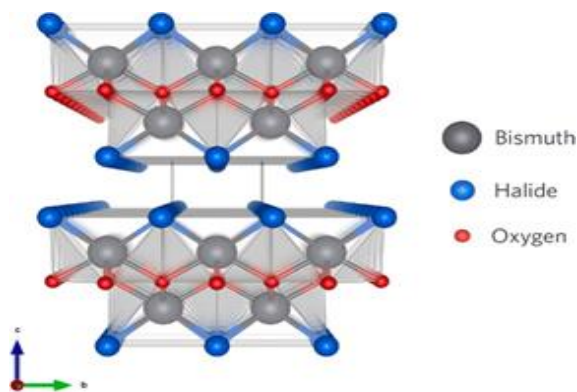


Figure 1. BiOX (X = I, Br, Cl, F) crystal structure systems [56]

For improving the overall degradation process, another form of composites has been used which are called ternary composites. The advantages of ternary composites are to suppress the recombination of photo-induced charges and provide more active sites than binary composites [57-61].

The aims of this study are reviewing recent works on the BiOBr synthesis methods, the characteristics of fabricated BiOBr, the pristine BiOBr photo-catalytic activity and operating parameters affecting on the degradation process using BiOBr composite.

2. SEMICONDUCTOR PHOTO-CATALYSIS

Semiconductor is a material that has conductivity between conductors and insulators. Semiconductor photo-catalysis is a photo-chemical reaction whereby a quantum of light (visible, ultraviolet or infrared radiation) was absorbed by a semiconductor to initiate a chemical reaction [62]. Photo-catalytic is a heterogeneous process, which uses different semiconductors like oxides (ZnO, CeO₂, TiO₂, WO₃, ZrO₂, Fe₂O₃, etc.) and sulfides (ZnS, CdS, etc.) in the presence of visible light [63-65]. Titanium dioxide (TiO₂) is characterized as low energy consumption, photo and chemical stability, high photo-catalytic activity, ease of availability, low operation temperature and nontoxic byproducts formation [66-69]. The basic elements should be provided for completing of a photo-catalytic reaction are: a photo-catalyst, source of light and the transformation of the chemical reaction partners. A semiconductor photo-catalyst must be photo-active and photo-stable, biologic and chemical inert, inexpensive and ecologic friendly to promote its functions [70].

The photocatalytic activity of semiconductors has been affected by several parameters such as phase structures, crystalline, particles size, defects and composition. It was reported the influence of these factors on the ZnO activity [71]. There is a difference in the photocatalytic activity at various values of specific surface area of rods: 8.02, 7.85, 7.91, and

6.04 m²/g. It was noticed that the improving of photogenerated carriers separation was related to the structure and aspect ratio of the facets. Thus recombination centers were decreased leading to higher photocatalytic efficiency [72, 73].

BiOBr is characterized as a narrow-band gap semiconductor [74] which is more suitable than other photo-catalysts such as ZrO₂, TiO₂, ZnO and SnO because it has the ability for absorbing the maximum portion of visible sunlight due to its narrow band gap. When a photo-catalyst is irradiated by light of the desired wavelength, the electrons-holes were transferred to the semiconductor surface and may recombine. This recombination has been producing heat and phonons, the number of charge carriers have been decreased and affect the photo-catalysis efficiency. So, binary and ternary composites like BiOI/BiOBr [35], GQDs/BiOBr [75] and BiOBr/W18O49/PAN [76] have been developed to enhance the photo-catalysis efficiency.

In a photo-catalytic reaction, the electrons excited from VB of the particular semiconductor to CB by providing sufficient and enough amount of photons energy (hν). The region of empty energy between these bands is called band-gap (E_g) [77]. The positions of VB and CB are essential features for determining the photo-catalytic ability of a semiconductor [78].

Generally, by irradiation of a semiconductor photo-catalyst with photons energy greater than its band-gap, this led to a photo-catalytic reaction gets initiated. The oxidative species that are generated in a photo-catalytic reaction is shown in Figure 2.

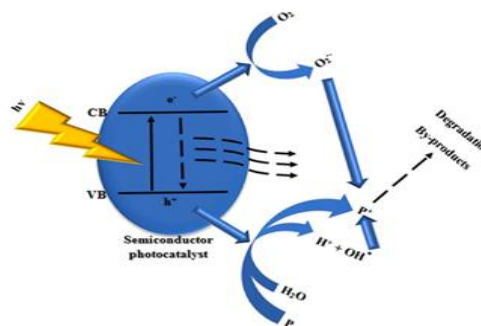
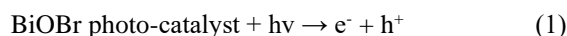
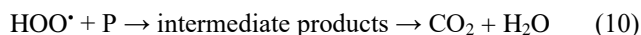
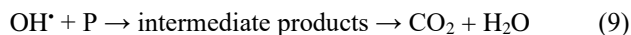
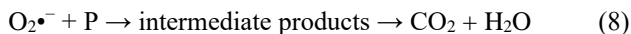
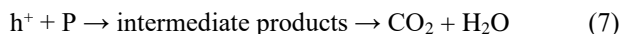
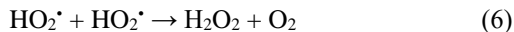


Figure 2. Schematic illustration of a typical photo-catalytic reaction [79]

In the conduction band, molecular oxygen (O₂) adsorbed has the ability to trap the electron (e⁻_{CB}), which is reduced to form superoxide radical anion (O₂^{•-}). Subsequently, (O₂^{•-}) radical can be further protonated to form hydro-peroxyl radicals (HOO[•]), then formation of hydrogen peroxide (H₂O₂) [80]. The generated (O₂^{•-}) and (HOO[•]) have the ability for degradation of organic pollutants (P) directly. On the other hand, the holes are capable of either producing intermediate products by directly oxidizing P or producing hydroxyl radicals (OH[•]) by oxidizing H₂O. Eqs. (1)-(10) below show a photo-catalytic reaction mechanism [81]:





3. SYNTHESIS OF BIOBR COMPOSITES

The photo-catalytic efficiency of composites can be enhanced by coupling of two or more semiconductors due to enhancing the surface area as well as synergistic role of each semiconductor. Various factors that influence the morphology, size, and properties of BiOBr composites such as pH and temperature. It was found that at pH equal to 8, best BiOBr photocatalytic activity has been obtained for photodegradation of rhodamine B. [82]. Temperature parameter has important effect on the size, and properties of BiOBr composites. At increasing the value of temperature, the BiOBr activity was increased and then decreased due to has a good crystallization. While for BiOCl, the efficiency was decreased due to decreasing in specific surface area and pore size [83].

BiOBr composites have been manufactured mainly by several processes such as: Ion exchange [84], solvothermal [85], wet chemical [86] ultra-sonication [87], co-precipitation [88] and hydrothermal methods [89].

3.1 Ion-exchange method

This method includes formed a new product by using a desired chemical species instead of present ionic species. For instance, using ion exchange method for synthesizing of Bi/BiOBr/AgBr. Firstly, Bi/BiOBr were fabricated by dissolving sodium bromide (0.001 mol) in 30 ml of ethylene glycol and heated at 180°C for 15 h in Teflon-lined autoclave. Then, the synthesized particles were separated by centrifugation, washed with pure water/absolute ethanol for many times and dried for 4h at 80°C. After that, it was used ion exchange method to synthesizing of Bi/BiOBr/AgBr. In which, 0.001 mol of AgNO₃ was dissolved in 80 mL ethylene glycol (80 mL), then added of synthesized Bi/BiOBr microspheres (0.4 g) to the mixture and stirred for 12h. Then, the synthesized particles were separated by centrifugation, washed three times and dried at 80°C for 4 h [84].

Figure 3 shows the Bi/BiOBr, BiOBr/AgBr and Bi/BiOBr/AgBr phase structures were characterized by XRD [90-92].

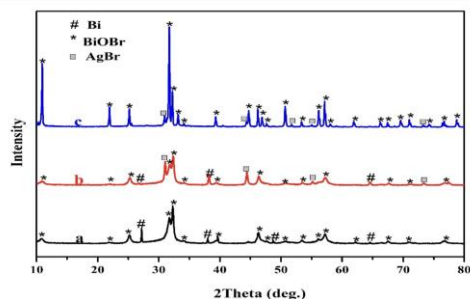


Figure 3. XRD patterns of (a) Bi/BiOBr; (b) Bi/BiOBr/AgBr and (c) BiOBr/AgBr

3.2 Solvothermal method

A Teflon-lined autoclave was used for heating the precursors. To produce precursor solutions, a variety of the solvents are used such as ethanol [93], ethylene glycol [94] and glycerol [95]. For instance, the solvothermal route has been used to synthesis CdS/BiOBr binary composites. Firstly, CdS has been synthesized by dissolving (2.666 g) of C₄H₆CdO₄·2H₂O and (2.256 g) of CH₃CSNH₂ in de-ionized water (30 mL) and stirring for 1 h. During stirring, CH₃CSNH₂ solution was added into the cadmium nitrate solution. Then, the mixture was heated at 120°C for 24 h in Teflon-lined autoclave. The synthesized particles were separated by centrifugation, washed with pure water/absolute ethanol for three times and dried for 6 h at 60°C.

To synthesizing of BiOBr, (1.0902 g) of bismuth nitrate was dissolved in 30 mL of ethyl glycol and mixed with 0.078 M of KBr. Then, a Polyvinylpyrrolidone (0.3020 g) was added into the Bi(NO₃)₃·5H₂O and stirred for 30 min. After that, 0.078 M KBr solution was added to the mixture under stirring for 1 h. Then, the mixture was heated at 160°C for 12 h in Teflon-lined autoclave. The synthesized particles were separated and washed with pure water/absolute ethanol for many times and dried for 6h at 60°C.

Finally, to synthesizing of CdS/BiOBr binary composites, take (0.3033 g) of synthesized CdS and added to 0.07 M of KBr solution. Then, a Polyvinylpyrrolidone (0.3207 g) was added into 0.0868 M of Bi(NO₃)₃·5H₂O. Separately, both mixtures were separately stirred for 1 h. After that, the suspension of CdS and KBr was added into the bismuth nitrate solution with stirring for 1 h. The synthesized particles were separated and washed with pure water/absolute ethanol for many times and dried for 6h at 60°C [96]. By using solvothermal method, there are various BiOBr based composites can be synthesized like BiOCl/BiOBr [97], PANI/BiOBr/ZnFe₂O₄ [98] and Fe₃O₄/mSiO₂/BiOBr [95].

Also, the BiOBr physicochemical properties were influenced by the initial reaction pH value. For instance, synthesizing of BiOBr catalysts with different values of reaction pH. It was noticed that the average thickness and width of BiOBr nanosheets were inversely proportional with pH value, as shown in Table 1, even though the basic samples units were nanosheets, see Figure 4 [99].

Table 1. BiOBr sizes and specific surface areas at various pH [99]

pH Value	Width (μm)	Thickness (nm)	S _{BET} (m ² /g)
1	2-4	110-130	2.51
3	1-3	80-90	5.18
5	1-2	60-80	5.80
7	0.5-2	40-60	11.91

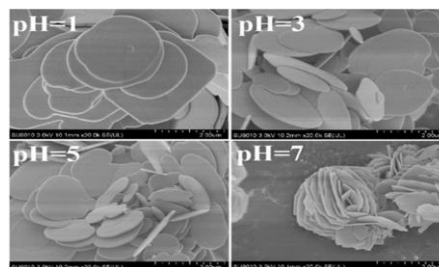


Figure 4. FE-SEM of BiOBr samples at various values of pH [99]

Moreover, with the high value of pH, the (001), (002), (003) and (004) diffraction peaks (shown in Figure 5) weakened due to the reduced exposure percentage of [001] facets. Also, it was concluded that BiOBr has been nucleation at pH value was increased leads to a decrease in crystallite size, so the BET specific surface area is directly proportional to the pH value, see Table 1.

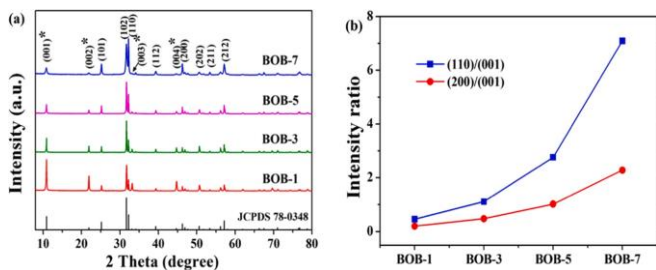


Figure 5. (a) BiOBr XRD patterns and (b) their peak intensity ratios of BiOBr samples [99]

3.3 Wet-chemical method

In this method, it was synthesized of composites in suitable liquid such as deionized water. For instance, Gao et al. fabricated of $\text{BiPO}_4/\text{BiOBr}$ binary composites by using wet-chemical method. In which, 0.01 mol of synthesized BiOBr was dissolved in ethanol (40 mL). Then, it was added the concentrated H_3PO_4 to the solution with and stirred for 20 min. The obtained particles were washed and dried at 80 C for 12 h to obtain $\text{BiPO}_4/\text{BiOBr}$ composites [100]. The BiOBr phase structures and $\text{BiPO}_4/\text{BiOBr}$ were characterized by XRD, and the results are shown in Figure 6.

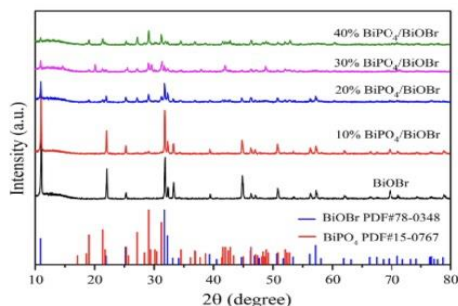


Figure 6. XRD patterns of BiOBr and $\text{BiPO}_4/\text{BiOBr}$ [100]

3.4 Ultra-sonication method

Ultrasonic processing has been used to process the mixtures of reaction for a suitable time. For example, Cheng et al. [101] used this method for synthesizing of $\text{BiOBr}/\text{Bi}_2\text{O}_2\text{CO}_3$ binary composite. Firstly, $\text{Bi}_2\text{O}_2\text{CO}_3$ was synthesized by dissolving 1.4553 g of $\text{Bi}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$ in 60 mL of diluted HNO_3 . Then, it was added a urea (3.6 g) to the solution and sonication treated for 10 min. The solution was heated at 160°C for 10h in a Teflon-lined autoclave. The produced particles were washed and dried at 80°C.

To synthesize of BiOBr, 1.4553 g of $\text{Bi}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$ was dissolved in 30 mL of pure water and treated ultra-sonically for 20 min. On the other hand, 0.309 g of sodium bromide was dissolved in 30 mL of pure water and dropped this solution to the above solution and treated the mixture ultra-sonically for 10 min. The solution was heated at 160°C for 10h in a Teflon-

lined autoclave. The produced particles were washed and dried at 80°C.

Finally, to synthesize of $\text{BiOBr}/\text{Bi}_2\text{O}_2\text{CO}_3$ binary composites, 0.515g sodium bromide was dissolved into 50 mL of pure water. Then, 0.5 g of synthesized $\text{Bi}_2\text{O}_2\text{CO}_3$ was dispersed in the above solution and treated the mixture ultra-sonically for 40 min. The produced particles were washed with water and dried at 80°C [101].

3.5 Co-precipitation method

This method required less time and low temperature, which is environment friendly as compared to other methods.

Zhao et al. [102] used this method for synthesizing of $\text{BiOBr}/\text{TiO}_2$ composite. In which, during an appropriate amount of potassium bromide has been dissolved into 20 mL of de-ionized water, an appropriate amount of $\text{Bi}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$ was added into de-ionized water (20 ML) to form suspension X_1 . After that, TiO_2 nanobelts (40 mg) were dispersed in to potassium bromide with stirring to form suspension X_2 . After that, the suspension X_2 was added slowly into the suspension X_1 under stirring at room temperature for 30 min. Then, the synthesized particles were separated and washed with pure water/ethanol solution and dried at 60°C to form $\text{BiOBr}@/\text{TiO}_2$ hetero-structures.

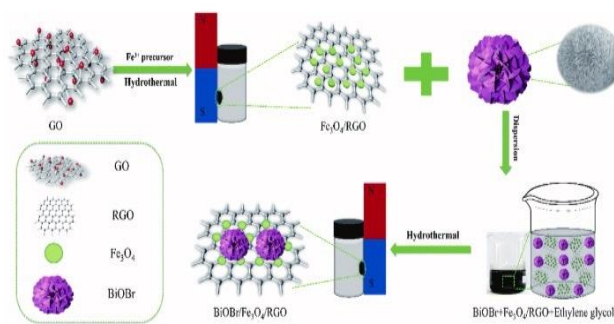


Figure 7. Mechanism of using $\text{PANI}/\text{BiOBr}/\text{ZnFe}_2\text{O}_4$ for photo-catalytic degradation of nitrobenzene [59]

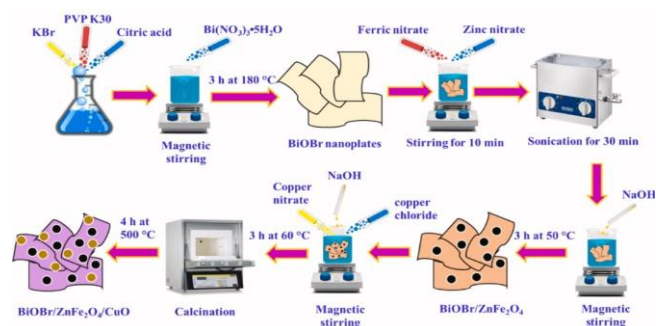


Figure 8. Schematic illustration explains of the constructing of $\text{BiOBr}/\text{ZnFe}_2\text{O}_4/\text{CuO}$ nano composites [103]

3.6 Hydrothermal method

Water has been used as a solvent in hydrothermal method. For instance, Jiang Y. et al. used this method for fabricating BiOBr/BiOI binary composites for the photo-catalytic crystal violet dyes degradation. The general procedure includes adding of precursors into water/nitric acid and transfer the mixture to an autoclave and heat at (110-260°C) for 12 h. Then,

a variety of hetero-structures are produced [89]. Moreover, this method has been used for synthesizing of Ternary composites of BiOBr/Fe₃O₄/rGO [60], as explain in Figure 7. Also, Figure 8 shows the fabricating BiOBr/ZnFe₂O₄/CuO photo-catalyst by a hydrothermal method [102]. The phase structures of BiOBr, ZnFe₂O₄, CuO, and BiOBr/ZnFe₂O₄/CuO. were characterized by XRD [103], see Figure 9.

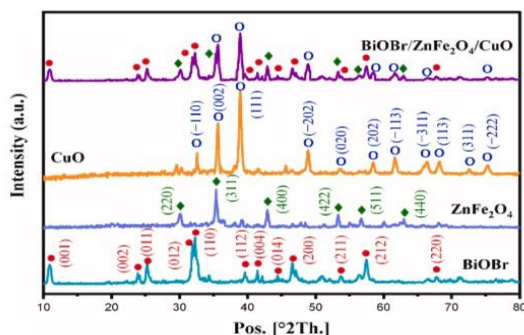


Figure 9. XRD patterns of BiOBr, ZnFe₂O₄, CuO, and BiOBr/ZnFe₂O₄/CuO [103]

4. APPLICATION OF BIOBR IN WASTEWATER TREATMENT

In the photocatalytic process, the photo-catalyst was interacted to the incident visible light for creating the electron-hole (e^- / h^+) pairs [104, 105]. The visible light has the ability to promoted the valence band electron to the conduction band, while a hole (h_{VB}^+) is generated by the interaction of visible-light. These charges have the ability for reacting with adsorbed species and migrate from the bulk to the surface of photocatalysis [106, 107].

Binary and ternary composites have the ability to enhancement the photo-catalytic activity and increased the degradation efficiency more than 50%. For example, Han et al. [108] showed that the chlorophenol degradation was increased from 0.8% (by using BiOBr composite) to more than 92% (by using BiOBr/NaBiO₃). Band structure, band gap and interface of the composites have the ability for enhancement of photocatalytic efficiency. Formation of narrow binary composite band gap is the main factor for enhancement the photo-catalytic activity by increased sunlight harvesting, it was shown that the BiOBr and NaBiO₃ band gabs are 2.880 eV and 2.60 eV, respectively, which was reduced to 2.52 eV when the formation of BiOBr/NaBiO₃ binary composite. Conclusively, efficient visible light harvesting resulted in binary composite due to modulated band gap.

Although the binary composites have the ability for enhancing the photo-catalytic activity, but still not perfect in practical applications. So, researchers have been increased charge separation and enhanced surface area by synthesizing of ternary composites. For example, in the case of Bi/BiOBr/AgBr composites, the degradation percentage of wastewater pollutants was increased from 48% (BiOBr) to more than 95%. Furthermore, metal-oxygen bond has the ability to recovery of composites after pollutants degradation and has the ability for increasing surface area, like BiOBr/Fe₃O₄/RGO [60], Fe₃O₄/BiOBr/BiOI [94], Fe₃O₄/mSiO₂/BiOBr [95] and Fe₃O₄/BiOBr/CQDs [109]. Conclusively, ternary composites have more adsorption area for pollutants and more ability for degrading pollutants than

binary composites, for example, the rhodamine B degradation efficiency by Bi/BiOBr/AgBr was 4.9 and 1.4 times faster than BiOBr/AgBr and Bi/BiOBr, respectively [84]. Similarly, BiOBr/TiO₂/G has the ability for degradation of various wastewater pollutants more efficiently than BiOBr/TiO₂ [89, 110-113] and the degradation efficiency of g-C₃N₄/BiOI/BiOBr was a better than BiOI/BiOBr [102, 114-118]. Also, Bi₂O₂CO₃/Ti₃C₂T_x heterostructure has the ability to degrad 95.4% of levofloxacin, while the degradation efficiency was 68.1% by using Bi₂O₂CO₃ [119].

Several operating parameters like dose of a semiconductor, pollutants concentration and intensity of visible light irradiation have been studied in the literature. Higher catalyst concentrations provide reactive radical's generation. On the other hand, due to particles aggregation, the effective path-length of radiation was reduced when using excess of catalyst [120-122].

5. PHOTO-CATALYTIC ACTIVITY OF BIOBR

The previous studies indicated that the treatment of water contaminated with synthetic dyes become difficult due to stability of dyes in water. Recently, nanoparticles under visible light have the ability for removing dyes from wastewater [22]. Various organic pollutants were degraded by using BiOBr as catalyst with visible light. However, the hierarchical structure, surface area, crystallization are the main factors that affecting the degradation efficiency of pollutants. For instance, Xia et al. [123] used BiOBr nanospheres for degrading of 10 mg/L of (RhB), it was found that the degradation percentage was 100% when using BiOBr nanospheres as catalysts. While, the degradation percentage was 75% after 105 min by using hollow spheres.

Xue et al. [124] used 3D-BiOBr hierarchical microspheres for degrading 7 mg/L of Rhodamine Blue (RhB) by using 1:1, 1:2 and 2:1 molar ratios of Br:Bi for 35 min detention time, it was found that the degradation efficiencies were 91.1%, 88.6% and 95.9%, respectively. Due to high percent of Bi compared with Br in the second mixture, the degradation efficiency as low compared with other efficiencies.

BiOBr photo-catalytic activity was also found to be impacted by its hydrothermal pH value. For instance, Ye et al. [125] used BiOBr with three different values of pH (0.4, 6 and 10) for degradation of 10 ppm of RhB. It was found that the degradation efficiencies after 2h were 67%, 83% and 99%, respectively. It was concluded that the BiOBr photo-catalytic activity has a direct relationship with pH value because the BiOBr band structure, size and its surface area are affected by its hydrothermal pH value.

By comparing the BiOBr activity with other commonly used photocatalysts, the Rhodamine (RhB) degradation efficiency was 92% by using Ni-ZnS/g-C₃N₄ heterojunction after 75 min [126]. Another study showed that by using Iron oxide/CdS to degrade Xylenol blue (XB), the photodegradation efficiency was 90.2% after 3 hr [127]. Also, it was reported that the BiOBr/ZnFe₂O₄/CuO photocatalyst has optimum photocatalytic properties, which has the ability for destroying 98% of malachite green in 90 min [103].

Band-gap, mesoporous structure, surface area and synergistic effect between TiO₂ and BiOBr are the main factors that affect the photocatalytic activity of BiOBr nanoparticles such as band-gap, mesoporous structure, surface area and synergistic effect between TiO₂ and BiOBr [128].

6. ENHANCING THE PHOTO-CATALYTIC PERFORMANCE OF BIOBR

The electron-hole recombination is an important process in semiconductors due to it plays a crucial role in photocatalytic reactions [129]. The electron-hole recombination cause losses in copper zinc tin sulfide charge and energy [130].

To enhance the BiOBr performance, there are many efforts and methods have been used for this purpose. The widely used methods are discussed below.

6.1 Elemental doping

Several dopants have been used to enhance the photocatalytic activity of BiOBr, as tabulated in Table 2. The most important efforts include inhibits recombination of electron-hole by doping an appropriate number of cations [131]. Some of the cations can be introduced to improve other properties, such as redox-potential of the photogenerated radicals and the ability of BiOBr for harvesting of visible light [132]. However, it was noted that photo-catalytic activity was decreased in the presence of excess cationic dopants because these dopants serve as recombination centers [133].

To inhibit the electron-hole recombination, it was used Ag-TiO₂-xNx to separate of electrons and holes under UV-visible light. The higher efficiency of this composite is reacted to the electrons and holes density. So, the Ag loading has the ability for decreasing of the recombination and enhancing the photocatalytic activity. The electron acceptors can be inhibit the fast electron-hole recombination by accelerating the electron transfer [134, 135].

Many researchers studied the BiOBr photo-catalytic activity loaded with noble metals. For instance, using photo-deposition method for loading palladium nanoparticles onto the surface

of BiOBr. It has been further loading of palladium nanoparticles onto BiOBr surface for enhancing the Pd-doped BiOBr absorbance intensity. The phenol degradation was 67% only by using BiOBr for 5h. While, when using 0.5 Pd-BiOBr, it was successfully degraded all phenol [136]. Moreover, using photo-reduction method for dispersing Pd NPs onto BiOBr surface [137].

Other forms of cationic dopants are the transition metals that were reportedly introduced into BiOBr, this method has the ability for improving the absorbance intensity in the visible region as well as inhibit recombination of electron-hole. For instance, synthesizing of Fe-doped BiOBr photo-catalysts for methyl orange degradation, it was noticed that the photocatalytic activity of Fe-doped BiOBr more effective than that of pristine BiOBr [131]. Similarity, synthesizing of Zn-doped BiOBr catalysts with a 300 W Xe lamp to degrade RhB dye [133]. Many authors studied the potential for leaching of dopants, it was reported that the catalytic site of combined W dopants and Co vacancies was less active than the W dopants alone. However, the leaching-induced Co vacancies with residual W dopants cause decreasing of current density and the created catalytic sites is more active than generated by W dopants alone. As a result, the photocatalytic activity of combined W dopants and Co vacancies has been enhanced [138].

Literature has also used the anionic dopants onto BiOBr surface to inhibits recombination of electron-hole. For instance, B-doped BiOBr samples was synthesized for the RhB dye degradation. It was reported high degradation efficiency of B-doped BiOBr compared with alone pristine BiOBr [139]. Also, degradation of RhB dye by S-doped BiOBr. It was found that the degradation rate of S-doped BiOBr was 0.0960 per min. While, it was 0.0176 per min of pristine BiOBr [140].

Table 2. The photo-degradation efficiencies of common BiOBr and doped BiOBr catalysts

Dopant	Source of Light	Experimental Conditions [Pollutant]; Dosage; Time of Irradiation	Photo-Degradation Efficiency, %		Reference
			BiOBr	Doped BiOBr	
Er	300 W Xe lamp with a light cut-off filter ($\lambda > 400$ nm)	[CIP]=10 mg/L; 100 mg/L; 360 min	50	61	[132]
Ag	500 W Xe lamp	[MO]=10 mg/L; 200 mg/L; 120 min	58.5	98.6	[137]
Fe	150 W halogen-lamp with light intensity of 200 mW/cm ²	[MO]=10 mg/L; 1000 mg/L; 120 min	75	100	[131]
Ti	11 W lamp with a light cut-off filter ($\lambda \geq 400$ nm)	[RhB]=10 mg/L; 1000 mg/L; 180 min	-75	100	[141]
Pt	300 W Xe lamp to provide visible light (320 nm $< \lambda < 680$ nm)	[PNP]=10 mg/L; 1000 mg/L; 30 min	-25	100	[52]
Cu	200 W Xe arc lamp with a light cut-off filter ($\lambda \geq 420$ nm)	[NOR]=10 mg/L; 1000 mg/L; 90 min	-20	-45	[142]
Cu	200 W Xe arc lamp with a light cut-off filter ($\lambda \geq 420$ nm)	[NOR]=10 mg/L; 1000 mg/L; 90 min	-20	-45	[142]
Co	Xe lamp (500 W) with a light cut-off filters ($\lambda \geq 400$ nm)	[RhB]=10 mg/L; 1000 mg/L; 120 min	34	99.5	[143]
Mn	Daylight lamp with a light cut-off filters ($\lambda \geq 400$ nm)	[RhB]=0.2 g/L; 1000 mg/L; 140 min	78	96.5	[144]
Nb	300 W Xe lamp with a light cut-off filters ($\lambda \geq 420$ nm)	[RhB]=10 mg/L; 200 mg/L; 20 min	51	-100	[145]
S	11 W lamp with a light cut-off filter ($\lambda \geq 400$ nm)	[RhB]=10 mg/L; 400 mg/L; 60 min	-45	-100	[146]
I ^a	500 W Xe lamp with a light cut-off filter to provide visible light ($\lambda = 400$ nm)	[MO]=10 mg/L; 2000 mg/L; 180 min	-20	-70	[147]
S	1000 W Xe lamp with a UV cut-off filter ($\lambda \geq 420$ nm)	[RhB]=20 mg/L; 500 mg/L; 50 min	50	100	[140]
Bi	300 W perfect Xe lamp with a UV cut-off filters	[TC]=NA;	-30	-100	[148]

	($\lambda \geq 400$ nm)	800 mg/L; 20 min			
B	150 W tungsten lamp with a UV cut-off filters ($\lambda \geq 420$ nm)	[RhB]=15 mg/L; 1000 mg/L; 30 min	71.1	99.3	[139]
Y	25W Eco-living day light fluorescent	[CIP]=20 mg/L; 1000 mg/L; 60 min	71.7	87.6	[149]

*CIP = Ciprofloxacin; RhB = Rhodamine B; MO = Methyl Orange; TC = Tetracycline; PNP = p-nitro-phenol; NA = Not Available; W = Watts.

6.2 Coupled semiconductors

The formation of hetero-junction with semiconductors is another widely approach has been used for improving the photo-catalytic performance of photocatalysts [150].

The synergistic effects on the photo-catalytic performance have been studied. It was reported synergetic effect on g-C₃N₄ structure, the g-C₃N₄ has the ability to create gap states. while the g-C₃N₄ band edge can be tune by coupling effect at g-C₃N₄/MoS₂ interface, this modification has the ability for inhibiting the electron-hole recombination by affecting the electron distribution [151].

Metal-organic frameworks (MOFs) have several properties make it more suitable for photocatalytic applications, but there stability is the most important challenge, the stability of these compounds is limited. To overcome these limitations, many authors developed composite materials such as utilizing molybdenum disulfide (MoS₂), in combination with (MOFs). It was concluded that the MoS₂/MOF heterojunctions were high photochemical stability and effective compared to pure MOFs due to there. It was indicated that these composites have the ability for using in practical applications [152].

such as tabulated in Table 3. Bismuth-based semiconductors are the most common coupling semiconductors have been used in photocatalytic process. For example, Qiu et al. [153] used solvothermal method for loading different amounts of bismuth sub-carbonate (Bi₂O₂CO₃) onto BiOBr nanosheets to form Bi₂O₂CO₃/BiOBr (BOC/BOB). The degradation efficiency of fabricated Bi₂O₂CO₃/BiOBr p-n heterojunction composites was a better than pristine BiOBr or pristine Bi₂O₂CO₃ degradation efficiencies under visible light. There are many factors which enhanced the performance of composites, including: the p-n hetero-junction, suppressed recombination

and facilitating charge separation. Also, Su and Wu [154] used hydrothermal method to construction of BiOBr/Bi₄O₅Br₂ composites for photo-catalytic degradation of CIP at various pH (pH=4, 5, 6 and 7). It was noticed that the highest degradation efficiency can be obtained by synthesized of the BiOBr/Bi₄O₅Br₂ composite at pH 7.

Another widely approach is loading metal-free carbonaceous materials, like carbon quantum dots (CQDs) on to BiOBr. For example, Xia et al. [155] used different weight ratio of CQD to BiOBr for synthesizing a series of CQDs/BiOBr to degrade of RhB dye. It was noticed that the pristine BiOBr degradation efficiency was 37% only after 30 min. While, the degradation efficiency of CQDs/BiOBr was 100%, as shown in Figure 10.

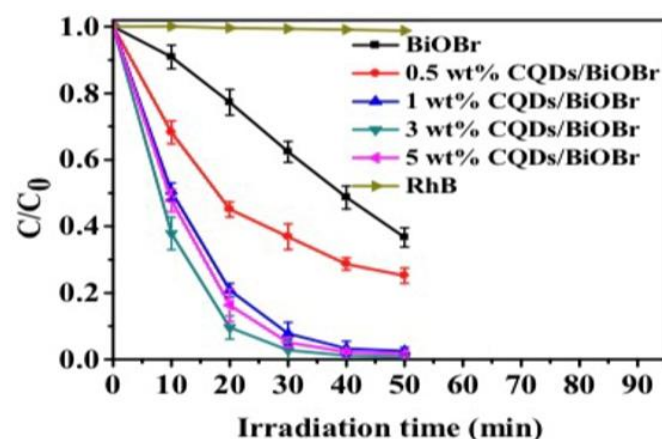


Figure 10. The photo-catalytic degradation of RhB in the presence of 1CQDs/BiOCl composites [155]

Table 3. The photo-degradation efficiencies of common BiOBr and coupled BiOBr catalysts

Coupling Catalyst	Source of Light	Experimental Conditions Dosage; [Pollutant]; Time of Irradiation	Photo-Degradation Efficiency, (%)		Reference
			Pristine	Coupled	
			BiOBr	BiOBr	
Bi ₄ O ₅ Br ₂	Improvised photo-catalysis reactor under 500 W Xe lamp	[CIP] = 10 mg/L; 1000 mg/L; 150 min	50	91	[154]
Bi ₂ O ₂ CO ₃	500 W Xe lamp ($\lambda > 420$ nm)	[RhB] = 10 mg/L; 500 mg/L; 45 min	46.99	92.83	[156]
CdS	1 kW Xe lamp	[MG] = 15 mg/L; 600 mg/L; 100 min	~ 20	99	[157]
NiFe ₂ O ₄	350 W Xe lamp ($\lambda > 420$ nm)	[MB] = 10 mg/L; 1000 mg/L; 60 min	45	90	[158]
CoTiO ₃	Xe lamp (500 W)	[RhB] = 10 mg/L; 1000 mg/L; 50 min	~ 40	100	[159]
CQDs	Xe lamp (500 W) equipped with a light filter ($\lambda = 420$ nm)	[RhB] = 10 mg/L; 300 mg/L; 20 min	67	92	[157]
ZnO	Xe lamp (300 W) equipped with a light filter ($\lambda = 420$ nm)	[RhB] = 5 mg/L; 1000 mg/L; 130 min	58	95	[160]
CoFe ₂ O ₄	Xe lamp (300 W) as a simulated solar light source	[CR] = 15 mg/L; 1000 mg/L; 60 min	62.27	90.78	[161]
CQDs	Xe lamp (300 W) equipped with a light filter ($\lambda = 400$ nm)	[RhB] = 10 mg/L; 200 mg/L; 30 min	37	~ 100	[155]
TiO ₂	104 W Slovenia cool white lamp	[CIP] = 25 mg/L;	30.7	95.5	[162]

	($\lambda = 390\text{--}700\text{ nm}$)	1000 mg/L; 180 min			
WS ₂	Xe lamp (500 W) to obtain visible light with a UV cut-off filter ($\lambda > 400\text{ nm}$)	[CIP] = 20 mg/L; 1000 mg/L; 100 min	61	92	[163]
FePc	Xe lamp 350 W equipped with a UV filter ($\lambda \geq 400\text{ nm}$)	[CIP] = 10 mg/L; 400 mg/L; 4 h	~ 30	~ 60	[164]
g-C ₃ N ₄	Xe lamp 300 W equipped with a UV cut-off filter ($\lambda \geq 400\text{ nm}$)	[CIP] = 10 mg/L; 200 mg/L; 6 h	67	85	[165]
WO ₃	500 W Xe lamp equipped with a UV cut-off filter ($\lambda \geq 400\text{ nm}$)	[CIP] = 20 mg/L; 500 mg/L; 120 min	59.1	94.7	[166]
α -Fe ₂ O ₃	300 W Xe lamp equipped with a UV cut-off filter ($\lambda \geq 420\text{ nm}$)	[RhB] = 20 mg/L; 1000 mg/L; 40 min	60	95	[167]
C ₃ N ₄	300 W Xe lamp with two cut-off filters ($\lambda = 320\text{--}780\text{ nm}$)	[RhB] = 20 mg/L; 500 mg/L; 40 min	~ 65	100	[168]
BHO	300 W Xe arc lamp equipped with a 385–740 nm desired filters	[RhB] = 15 mg/L; 500 mg/L; 50 min	~ 75	100	[169]
CdS	Halide lamp (250 W) with a light cut-off filter ($\lambda \geq 400\text{ nm}$)	[RhB] = 20 mg/L; 1000 mg/L; 50 min	74	97	[170]
Bi ₂ O ₄	1300C Xe lamp	[MO] = 20 mg/L; 1000 mg/L; 10 min	~ 15	100	[171]
CdWO ₄	Xe lamp (300 W) equipped with a UV cut-off filter ($\lambda \geq 420\text{ nm}$)	[RhB] = 10 mg/L; 500 mg/L; 8 min	40	~ 100	[172]
TiO ₂	Xenon arc lamp (300 W) equipped with a UV cut-off filter ($\lambda \geq 400\text{ nm}$)	[MO] = 10 mg/L; 1000 mg/L; 80 min	~ 40	~ 91	[173]
BiSbO ₄	Xe lamp (300 W) equipped with a UV cut-off filter ($\lambda \geq 400\text{ nm}$)	[RhB] = 10 mg/L; 300 mg/L; 45 min	30	96	[174]
Graphene	xenon lamp (500 W) equipped with a UV cut-off filter ($\lambda \geq 400\text{ nm}$)	[RhB] = 10 mg/L; 200 mg/L; 24 min	50	100	[175]
BiOCl	LED light irradiation	[MB] = 10 mg/L; 1000 mg/L; 360 min	72	93	[176]
TiO ₂	300 W xenon lamp equipped with a UV cut-off filter ($\lambda \geq 400\text{ nm}$)	[RhB] = 10 mg/L; 250 mg/L; 40 min	~ 70	~ 100	[177]
Bi ₂ MoO ₆	50 W LED light (410 nm)	[MB] = 20 mg/L; 1000 mg/L; 40 min	> 20	> 90	[178]
QDs-Cu ₂ O	250 W halide lamp with equipped with a UV cut-off filter ($\lambda \geq 400\text{ nm}$)	[MB] = 10 mg/L; 1000 mg/L; 60 min	73	95	[179]
FeWO ₄	Xe lamp (300 W) with a 400 nm cut-off filter	1000 mg/L; 60 min	66.2	90.4	[180]
LaFeO ₃	200 W Xe lamp emitting simulated sunlight	[RhB] = 5 mg/L; 1000 mg/L; 30 min	95.2	95.8	[181]
Black phosphorus (BP)	300 W Xe arc lamp with a UV filter (420 nm < λ < 780 nm)	[TC] = 50 mg/L; 1000 mg/L; 90 min	~ 25	85	[182]
NaBiO ₃	Fluorescent lamp (22W)	[4CP] = 24 mg/L; 1000 mg/L; 20 min	0.8	> 92	[183]
La ₂ Ti ₂ O ₇	Xe lamp (300 W)	[RhB] = 10 mg/L; 400 mg/L; 20 min	~ 80	~ 100	[184]
CO/ZFO/BOB	150 W LED lamp	[MG] = 25 mg/L; 150 mg/L; 90 min	42	91	[185]
SnO ₂	5 W nine parallel LED lights	[RhB] = 20 mg/L; 1000 mg/L; 20 min	~ 60	~ 98.2	[186]
BiVO ₄	Xe lamp (300 W) with a 420 nm cut-off filter	[RhB] = 10 mg/L; 1000 mg/L; 15 min	~ 60	~ 95	[187]
Bi ₅ O ₇ Br	Xe lamp (500 W) with a 420 nm cut-off filter	500 mg/L; [CBZ] = 10 mg/L; 90 min	~ 25.8	~ 90	[188]
Bi(C ₂ O ₄) OH	150 W Xe lamp	1000 mg/L; [RhB] = 10 mg/L; 30 min	89.5	99.6	[189]
Bi ₁₂ O ₁₇ Cl ₂	300 W Xe arc lamp with 420 nm cut-off filter	[MO] = 10 mg/L; 500 mg/L; 20 min	~ 70	~ 92	[190]
BiOCO ₂ H	Xe lamp with a UV filter	[LEV] = 10 mg/L; 600 mg/L; 40 min	75.1	90.1	[191]
CeO ₂	300 W Xe lamp with 420 nm cut-off filter	[RhB] = 20 mg/L; 500 mg/L; 25 min	71.2	97.3	[192]
ZnO	300 W iodine-Wolfram lamps	[MB] = 10 mg/L; 1000 mg/L; 240 min	~ 42	~ 90	[193]
ZnS	350 W Xe lamp	[TC] = 20 mg/mL; 1000 mg/L; 25 min	~ 70	~ 82	[194]
Basic bismuth nitrate (BBN)	500 W Xe lamp emitting simulated sunlight	[RhB] = 10 mg/L; 500 mg/L; 45 min	61.8	91.98	[195]
CoS	44 W LED lamp with a UV filter	[GLP] = 10 ⁻⁴ mol/L;	21.9	74.7	[196]

		400 mg/L; 180 min			
Ag ₆ Si ₂ O ₇	CEL-HXUB300 Xe lamp equipped with a UV filter ($\lambda \geq 400$ nm)	[MB] = 20 mg/L; 1000 mg/L; 15 min	~ 25	98	[197]
ZnWO ₄	300 W Xe lamp equipped with a UV cut-off filter ($\lambda \geq 400$ nm)	[RhB] = 2 × 10 ⁻⁵ mol/L; 500 mg/L; 16 min	~ 50	~ 100	[198]
SnIn ₄ S ₈	266 W Xe lamp with a two glass filters (380 nm < λ < 780 nm)	[RhB] = 15 mg/L; 200 mg/L; 40 min	71.1	99.8	[199]
BiPO ₄	12 W LED light irradiation	[RhB] = 15 mg/L; 1000 mg/L; 120 min	83.51	95.66	[200]
MnFe ₂ O ₄	fluorescent lamp (150 W)	[2,4-D] = 20 mg/L; 1000 mg/L; 80 min	57.3	96.5	[201]
C60	Xe lamp (500 W) with a light filter ($\lambda > 420$ nm)	[RhB] = 10 mg/L; 1000 mg/L; 10 min	59	91	[202]
NiS	266 W Xe lamp with with a two glass filters (380 nm < λ < 780 nm)	[RhB] = 15 mg/L; 200 mg/L; 50 min	84	99.5	[203]
SnWO ₄	sunlight radiation (Natural)	[RhB] = 20 mg/L; 1250 mg/L; 60 min	65.9	97.85	[204]
Zn ₂ GeO ₄	Xe lamp equipped with a cut-off filter ($\lambda > 420$ nm)	[RhB] = 12 mg/L; 1000 mg/L; 40 min	~ 60	93	[205]
BiOI	500 W Xe lamp passed through annular quartz tube equipped with a cut-off filter ($\lambda > 420$ nm)	[MO] = 10 mg/L; 2000 mg/L; 300 min	~ 30	63.1	[206]

*CIP=Ciprofloxacin; CBZ = Carbamazepine; DC = Doxycycline; LEV = Levofloxacin; GLP = Glyphosate; 2,4-D=2,4-dichlorophenoxyacetic acid; RhB = Rhodamine B; TC = Tetracycline; MG = Malachite Green; MO=Methyl Orange; BPA = Bisphenol A; 4CP = 4-chlorophenol; CR = Congo Red; MB = Methylene Blue and W = Watts.

It was concluded that the coupling BiOBr with optimal amount of (CQDs) has the ability for enhancing the photo-catalytic degradation efficiency due to improved optical absorption [155].

7. THE OPERATIONAL PARAMETERS THAT EFFECT ON PHOTO-CATALYTIC DEGRADATION PROCESS

There are several operational conditions that influence on photo-catalytic degradation process are discussed below.

7.1 Initial pH of solution

Initial pH of solution is an important parameter that influences the photo-catalytic degradation efficiency process. Altering the initial solution pH can be Altering by changes in the properties of photo-catalyst's surface charge and the organic pollutant. Many methods have been used for estimating the point of zero charge like potentiometric titration method [136], modified batch equilibrium method [207], salt addition method [165], etc. BiOBr surface is negatively charged at pH above 5.30 and positively charged at pH below 5.30 because the zero charge point of BiOBr was recorded as 5.30 [208].

Also, Wang et al. [209] used BiOBr catalyst for degrading of sulfurhodamine MO, it was found decreasing in the degradation efficiency as pH value increases from 2.0 to 9.0 because low adsorption of sulfurhodamine MO onto the surface of BiOBr due to electrostatic attraction between BiOBr and sulfurhodamine MO. Similarity, by using BiOBr as photo-catalyst for degradation of RhB, it was found that as different values of pH, the performance degradation process was low due to low RhB adsorption onto the surface of BiOBr [210].

Moreover, BiOBr/ZnFe₂O₄/CuO composite has been used for degradation of malachite green (MG) at various pH values (pH=3, 5, 7, 9, and 11), it was noticed that the best (MG) destruction was obtained at pH=7 [103], as shown in Figure 11.

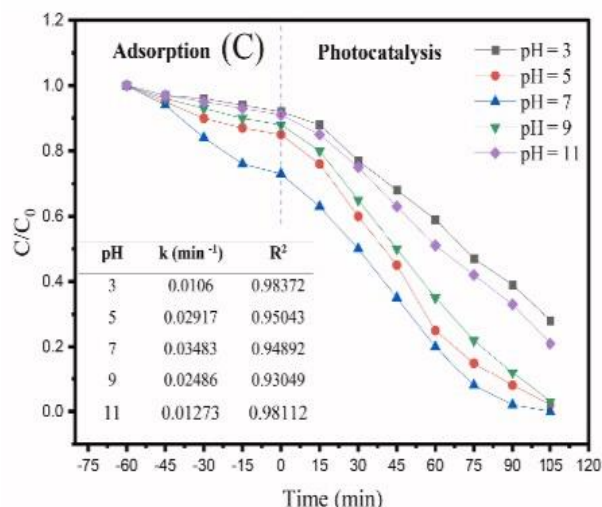


Figure 11. Effect of pH value on MG photo-oxidation over BiOBr/ZnFe₂O₄/CuO [103]

7.2 Catalyst dosage

The BiOBr photo-degradation efficiency is also affected by the dosage of BiOBr catalyst used for degrading of wastewater pollutants, which is lead to increase in the photo-catalytic reactions active surface and increase the photo-generated charge carriers [210-212].

Gondal et al. [213] used BiOBr (600-1500 mg/L) for degrading 7 mg/L of RhB dye and studied the effect of this dosage on the photo-catalytic degradation, with keeping other experimental parameters constant. For 90 min and using 1200 mg/L of BiOBr dosage, it was noticed that 94.6% degradation efficiency was obtained. Then, the degradation efficiency started to reduce. Similarly, it was used BiOBr (250-1500 mg/L) for degrading 10 mg/L of RhB dye and studied the effect of this dosage on the photo-catalytic degradation, it was reported that the optimum BiOBr dosage was 1000 mg/L at the degradation rate of 0.106 per min [210].

7.3 Inorganic ions

Wastewater often contains another pollutant which is inorganic ions [214]. Many authors have reported the effect of these ions on the degradation efficiency. For example, studying the effect of SO_4^{2-} , Cl^- , CO_3^{2-} , PO_4^{3-} and NO_3^- ions on the tetracycline photo-degradation rate using BiOBr. Due to their radical scavenging properties, Cl^- and CO_3^{2-} ions decreased the photo-catalytic activity, while NO_3^- , SO_4^{2-} and PO_4^{3-} did not show any significant inhibition, due to competition among tetracycline and inorganic ions for the BiOBr active sites [215, 216]. Similarly, the effect of SO_4^{2-} , Ca^{+2} , NH_4^+ , Cl^- , PO_4^{3-} , CO_3^{2-} and NO_3^- ions on the degradation of ibuprofen was studied. It was found that Cl^- , NH_4^+ , Ca^{+2} , SO_4^{2-} , NO_3^- and CO_3^{2-} ions have not significant inhibition effect. While in the presence of PO_4^{3-} ions, the photo-degradation rate was decreased significantly [217, 218].

8. CONCLUSIONS AND RECOMMENDATIONS

In this study, several information associated with BiOBr have been reviewed such as its synthesis, properties, photo-catalytic degradation activity and strategies for improving photo-catalytic efficiency of the BiOBr. Also, it was summarized various operational parameters that effect on BiOBr photo-catalytic degradation rate like initial pH of solution, catalyst dosage and inorganic ions.

Many methods that are used for synthesizing BiOBr, but the most employed methods are solvothermal and hydrothermal methods. By using such methods, various forms of BiOBr photo-catalysis have been synthesized with various features of dimension, pore volume, pore size and morphology. However, the large-scale fabrication of BiOBr must be encouraged with more attractive features.

There are many operational parameters that effect on BiOBr photo-catalytic degradation, including pH of the solution, BiOBr dosage and inorganic ions.

The useful strategies for improving the photo-catalytic performance of BiOBr include formation of composite materials and elemental doping. Even though, the formation of composite materials and elemental doping using facile processes are recommended. Lastly, BiOBr was frequently used for the photo-degradation of pollutants, mostly dyes. So, an important recommendation would be to consider the application of BiOBr in real wastewater treatment. As well as, should be further investigated other potential applications of BiOBr, considering the interesting features of BiOBr.

It should be focusing on developing a recyclable heterojunction with special properties under a UV light which has the ability to degrade non-biodegradable hazardous dye.

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