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Enhanced Photocatalytic Activity of the Bi₂O₃-NiO Heterojunction for the Degradation of Methyl Orange under Irradiation of Sunlight

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Abstract: Recently, the development of visible-light-responsive catalysts for the photodegradation of organic pollutants has captured the attention of researchers globally. The ineffectiveness and high costs associated with conventional methods and techniques used for the abatement of water pollution have forced researchers to develop effective and low-cost innovative techniques for this purpose. Photocatalysis is considered an effective protocol for this purpose. Therefore, this study was conducted for the development of the Bi₂O₃-NiO heterojunction as a visible-light-responsive photocatalyst for the degradation of methyl orange. Ni(NO₃)₂·6H₂O (Fluka) and Bi(NO₃)₃·5H₂O (Merck) were used as precursor materials for the synthesis of NiO-Bi₂O₃. After fabrication, the Bi₂O₃-NiO heterojunction was characterized using XRD, EDX, SEM, FTIR, and TGA techniques. Then, it was employed as a catalyst for the photodegradation of methyl orange under sunlight irradiation. The fabricated Bi₂O₃-NiO showed higher photocatalytic activity than Bi₂O₃ and NiO with 100, 67, and 46% degradation of methyl orange, respectively. The rate constant determined by the non-linear method of analysis for the photodegradation of MO in the presence of Bi₂O₃-NiO was 3.2-fold and 1.7-fold of the rate constant with NiO and Bi₂O₃, respectively. The higher photocatalytic activity of Bi₂O₃-NiO than of its individual components in the present study is also attributed to the separation and transfer of positive holes and electrons. The recycling of spent Bi₂O₃-NiO under similar experimental conditions exhibited the same photocatalytic activity suggesting the stability of the fabricated Bi₂O₃-NiO photocatalyst.

Keywords: photodegradation; methyl orange; heterojunction; Bi₂O₃; NiO

1. Introduction

The rapid expansion of the textile and chemical industries and the rapid increase in world population has resulted in a dramatic condition regarding the pollution of water reserves. Other sources such as science and technology, agriculture, and domestic activities also contribute to water pollution. However, the contribution to aqueous contamination by these various industries is very significant [1–7]. Various industries such as pharmaceuticals, leather, textile, food, etc., are responsible for water pollution. The effluents of these

industries transmit various organic and inorganic pollutants to the environment. Water pollution due to the release of organic dyes from different industries is the most dangerous type of pollution that causes a threat to living organisms and society. Aqueous pollution due to dyes is a critical issue worldwide. These dyes severely affect the environment due to their carcinogenic nature, high toxicity, and persistent nature [8–12]. Various traditional methods including physical, chemical, and biological methods have been attempted for the remediation of dyes-polluted water. However, these methods have some drawbacks such as high cost and ineffectiveness of treatment processes. The ineffectiveness and high costs associated with conventional methods and techniques used for the abatement of water pollution have forced researchers to develop effective and low-cost innovative techniques for this purpose. Photocatalysis has merged as a green, low-cost, and effective technique for the destruction of organic pollutants like dyes, pesticides, and pharmaceuticals present in wastewater recently [13–16]. The photocatalytic technique is an effective protocol for the abatement of organic pollutants like dyes and pesticides. The photocatalytic technique is a chemical process that is based on the production of hydroxyl radicals that take part in the complete mineralization of organic molecules into simple inorganic molecules [17–20]. The photocatalytic protocol has the following advantages over conventional methods:

1. Direct conversion of organic pollutants molecules into simple inorganic molecules like water and carbon dioxide
2. Mineralization of a wide range of organic pollutants irrespective of selectivity
3. No production of hazardous side products.

The photocatalytic technique is based on the in situ production of excitons in the valance band and conduction band of semiconductors photocatalysts under ultraviolet or visible light. These excitons generate highly reactive OH radicals through subsequent reactions. These OH radicals then mineralize the pollutants species. Depending upon the bandgap energy of the semiconductors photocatalysts, both ultraviolet and visible light can be used in the photocatalytic techniques. The semiconductor metal oxides like TiO_2 , ZnO , SnO_2 , CeO_2 , CuO , MgO , perovskite metal oxides, etc., have been used as traditional photocatalysts for a long time [21–25]. However, the traditional photocatalysts are not successful; the ineffective degradation of pollutants is due to their wide band gap energies. The traditional photocatalysts can only be used under the irradiation of ultraviolet light. As visible light is abundantly available, due to the sun (44% of sunlight is in the visible range), the development of a photocatalyst that can be effectively employed for the destruction of pollutants under the irradiation of sunlight is essential [26–28]. Semiconductors with narrow band gap energy can be employed for this purpose because such types of semiconductors can absorb visible light [29]. Therefore, the bismuth oxide, Bi_2O_3 , which is a narrow band gap semiconductor, has gained the interest of researchers recently. It has desirable characteristics such as appropriate band gap energy, stable structure, low cost, and an environmentally benign nature. It has been widely used in many fields such as for thin films, photovoltaic cells, fuel cells, and catalysis. The bismuth oxide exists in various forms, however, the $\alpha\text{-Bi}_2\text{O}_3$ (band gap 2.85 eV) and $\beta\text{-Bi}_2\text{O}_3$ (band gap 2.58 eV) are more important for catalytic applications [30–32]. However, due to the low energy difference in the valence band and conduction band of Bi_2O_3 , the photo-induced positive holes and electrons quickly recombine. As a result, the photocatalytic performance of Bi_2O_3 is badly affected. Therefore, researchers have attempted to reduce the recombination of positive holes and electrons by modification of the structure of Bi_2O_3 . One of the attempted modifications is the construction of heterojunctions between Bi_2O_3 and other semiconductors. The construction of a heterojunction between two semiconductors produces an electric field at the interface. This electric field assists in the separation of photo-induced positive holes and electrons resulting in the enhancement of photocatalytic activities [33–35]. This study reports the enhancement in photocatalytic activity of Bi_2O_3 for the photodegradation of methyl orange by the construction of a heterojunction with NiO . NiO is a p-type wide band gap (3.6–4.0 eV) semiconductor with high electro-optical proficiency and chemical stability. It has been used in several applications such as gas sensing, solar cells, and cataly-

sis [36–40]. The driving force for the construction of heterojunction between NiO and Bi₂O₃ was to develop a solar light-driven photocatalyst for the photodegradation of dyes. The Bi₂O₃-NiO heterojunction exhibits higher photocatalytic activity due to two reasons: (1) It harvests an extended portion of the UV-visible spectrum. (2) It suppresses the unwanted recombination of photo-induced positive holes and electrons due to its synergic effect.

2. Materials and Methods

2.1. Synthesis of Bi₂O₃-NiO

Analytical-grade chemicals were used in this study. All the chemicals and reagents were used as received, i.e., without further purification. Ni(NO₃)₂·6H₂O (Fluka) and Bi(NO₃)₃·5H₂O (Merck) were used as precursor materials for the synthesis of NiO-Bi₂O₃. Typically, equimolar quantities of both Ni(NO₃)₂·6H₂O and Bi(NO₃)₃·5H₂O were dissolved in 10 mL concentrated HNO₃. Then, the mixed solution was diluted with 100 mL of distilled water. The resultant solution was hydrolyzed by dropwise addition of 1M NaOH solution under continuous stirring. The addition of NaOH to the mixed solution produced a gel. The gel formed was filtered, washed, and dried at 100 °C for 24 h. Finally, the substance formed was calcined first at 300 °C and then at 600 °C for 4 h at each temperature. Pristine NiO and Bi₂O₃ were also prepared in the same way. The prepared samples were kept in glass vials for further study.

2.2. Characterization of Bi₂O₃-NiO

The NiO-Bi₂O₃ was characterized using XRD, EDX, SEM, FTIR, and TGA techniques. The instruments used for the characterization of prepared Bi₂O₃, NiO, and Bi₂O₃-NiO samples are listed in Table 1.

Table 1. Instruments used for characterization of Bi₂O₃, NiO, and Bi₂O₃-NiO samples.

No	Characterization Technique	Instrument	Model
1	XRD analysis	X-ray diffractometer	JOEL-JDX-3532, Japan
2	EDX analysis	EDX spectrophotometer	JSM5910, UK
3	SEM analysis	Scanning electron microscope	JEOL-JSM 5910, Japan
4	FTIR analysis	IR spectrophotometer	Bruker VRTEX70, USA
5	Thermal gravimetric analysis	TGA analyzer	Perkin Elmer 6300 TGA analyzer, USA
6	Analysis of dye solution	UV-visible spectrophotometer	Hitachi U-2800, Japan

2.3. Photocatalytic Activity

The photocatalytic activity of Bi₂O₃-NiO was explored through the photodegradation of methyl orange under sunlight. The photodegradation of methyl orange was studied in a Pyrex glass beaker. In the first step, blank experiments were performed. In the first blank experiment, the dye solution was stirred under sunlight for an estimation of photolysis. A sample was taken after 30 min and analyzed with a UV-visible spectrophotometer. There was no removal of methyl orange due to photolysis. The second blank experiment was performed for confirmation of the leaching of the catalyst. It was confirmed by stirring 0.1 g Bi₂O₃-NiO in 20 mL distilled water for one hour. After stirring, the Bi₂O₃-NiO was separated and the clear liquid was analyzed for the existence of Bi or Ni ions. The analysis confirmed that there was no leaching of the metal ions from Bi₂O₃-NiO. In the second step, an optimized amount of catalyst was suspended in a 50 mL solution of methyl orange and stirred in the dark to maintain adsorption equilibrium. The initial concentrations of the dye solution used were 50, 100, and 150 mg/L. A sample was taken and analyzed with a UV-visible spectrophotometer. About 10% removal of methyl orange was determined due to adsorption. Then, the beaker was placed in sunlight under continuous stirring in the third step. Samples were taken and analyzed with a UV-visible spectrophotometer.

3. Results and Discussion

3.1. Characterization

The XRD spectra of Bi_2O_3 , NiO, and Bi_2O_3 -NiO are given in Figure 1. The existence of sharp peaks in the XRD spectrum of Bi_2O_3 indicates the crystalline nature of the prepared sample. The XRD spectrum of Bi_2O_3 is dominated by diffraction peaks corresponding to planes of tetragonal α - Bi_2O_3 at 2θ 24.71° (121), 30.68° (012), 33.59° (-112), 46.84° (041), and 47.61° (-104) [41–44]. The XRD pattern of NiO also shows the crystallinity of the fabricated sample with diffraction peaks and hkl planes of cubic NiO at 2θ 37.31° (111), 43.32° (200), and 62.78° (220) according to PDF 01-089-5881 [45–47]. The XRD of the Bi_2O_3 -NiO heterojunction shows the diffraction peaks of both pure Bi_2O_3 and NiO which confirms the successful fabrication of the heterojunction. Based on the comparison of the XRD of the fabricated Bi_2O_3 -NiO heterojunction with the available literature, it was concluded that the heterojunction had been successfully fabricated with a tetragonal Bi_2O_3 phase and cubic NiO phase with space groups P-421c and Fm-3m, respectively. The 2θ values and hkl planes corresponding to Bi_2O_3 observed in the XRD pattern and Equation (1) were used for the calculation of the lattice parameters of the tetragonal structure. Equation (1) was applied to the observed data through the non-linear method using the “Solver” software (Microsoft 365) of MS Word. The lattice parameters of the tetragonal structure were calculated as $a = b = 2.32 \text{ \AA}$ and $c = 1.34 \text{ \AA}$. Similarly, using Equation (2) and the observed XRD data of NiO, the lattice parameters for the cubic structure were calculated as $a = b = c = 0.98 \text{ \AA}$ [42].

$$\frac{1}{d_{hkl}^2} = \frac{h^2 + k^2}{a^2} + \frac{l^2}{c^2} \quad (1)$$

$$\frac{1}{d_{hkl}^2} = \frac{h^2 + k^2 + l^2}{a^2} \quad (2)$$

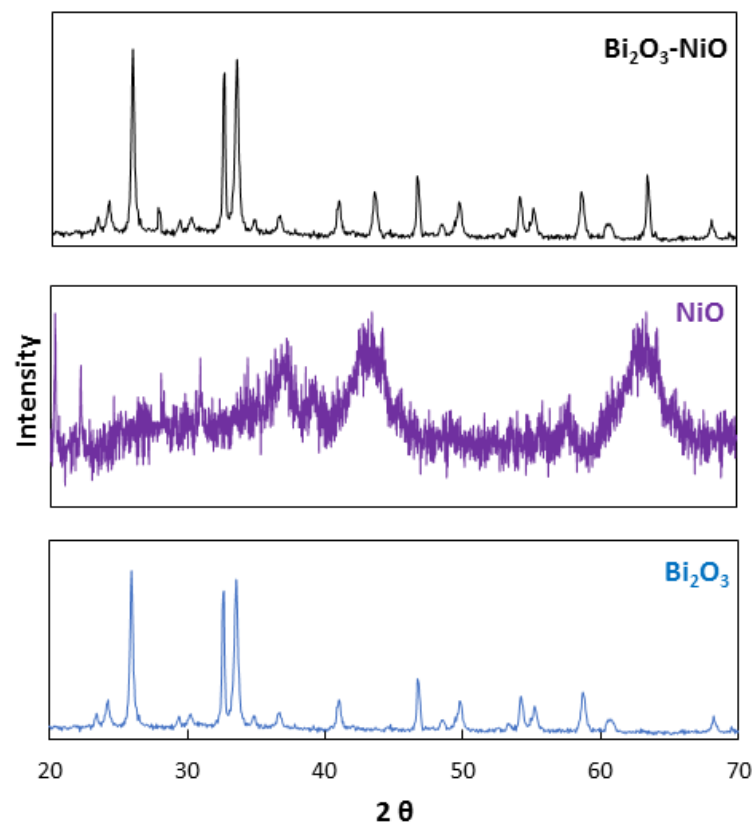


Figure 1. XRD analysis of Bi_2O_3 , NiO, and Bi_2O_3 -NiO samples.

The stability of the fabricated samples was investigated by thermal gravimetric analysis (TGA). Figure 2 shows the results of the thermal gravimetric analyses. All three fabricated samples were quite stable over a wide range of temperatures. A minute weight loss (about 2%) observed in each sample is attributed to the evaporation of adsorbed moisture content.

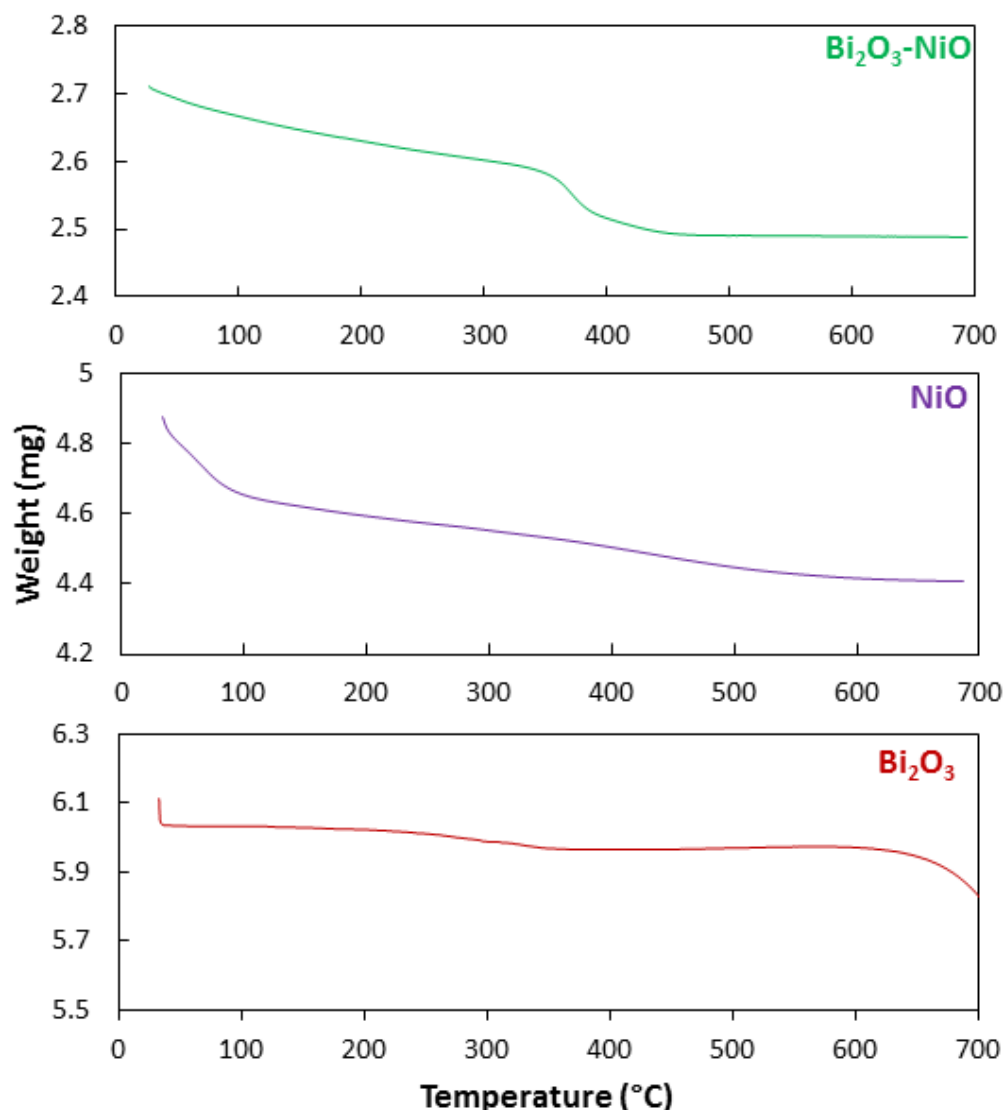


Figure 2. TGA analysis of Bi₂O₃, NiO, and Bi₂O₃-NiO samples.

The fabrication of the samples was further confirmed by elemental composition analyses using energy-dispersive X-ray (EDX) spectroscopy. Figure 3 shows the results of the energy-dispersive X-ray (EDX) analyses of the samples. The results confirmed that Bi₂O₃ comprised Bi and O. Similarly, the NiO comprised Ni and O only. The Bi₂O₃-NiO heterojunction is composed of Bi, Ni, and O. The demonstrated EDX results confirmed the successful fabrication and purity of the samples. A trace amount of C was also detected in the Bi₂O₃-NiO heterojunction which might be due to impurity in the precursor material or contamination during the synthesis process.

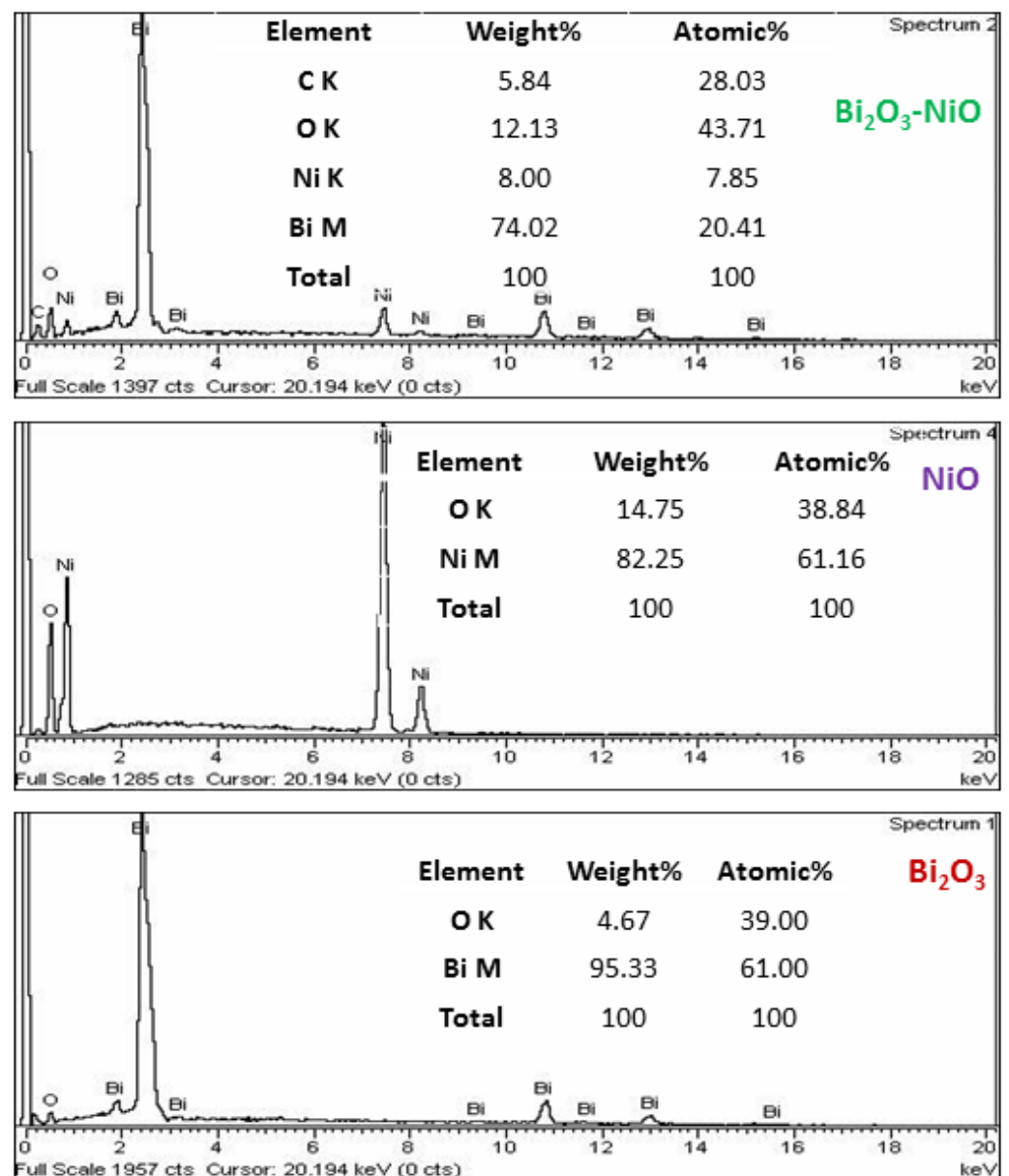


Figure 3. EDX analysis of Bi_2O_3 , NiO , and $\text{Bi}_2\text{O}_3\text{-NiO}$ samples.

The morphology and particle size analyses were carried out using scanning electron microscopy (SEM). Figure 4 demonstrates the scanning electron micrographs of Bi_2O_3 , NiO , and $\text{Bi}_2\text{O}_3\text{-NiO}$ samples. Particles of both Bi_2O_3 and NiO are non-agglomerated and irregular in shape. The SEM of $\text{Bi}_2\text{O}_3\text{-NiO}$ indicates that both Bi_2O_3 and NiO retain their identity and particle shape in the heterojunction.

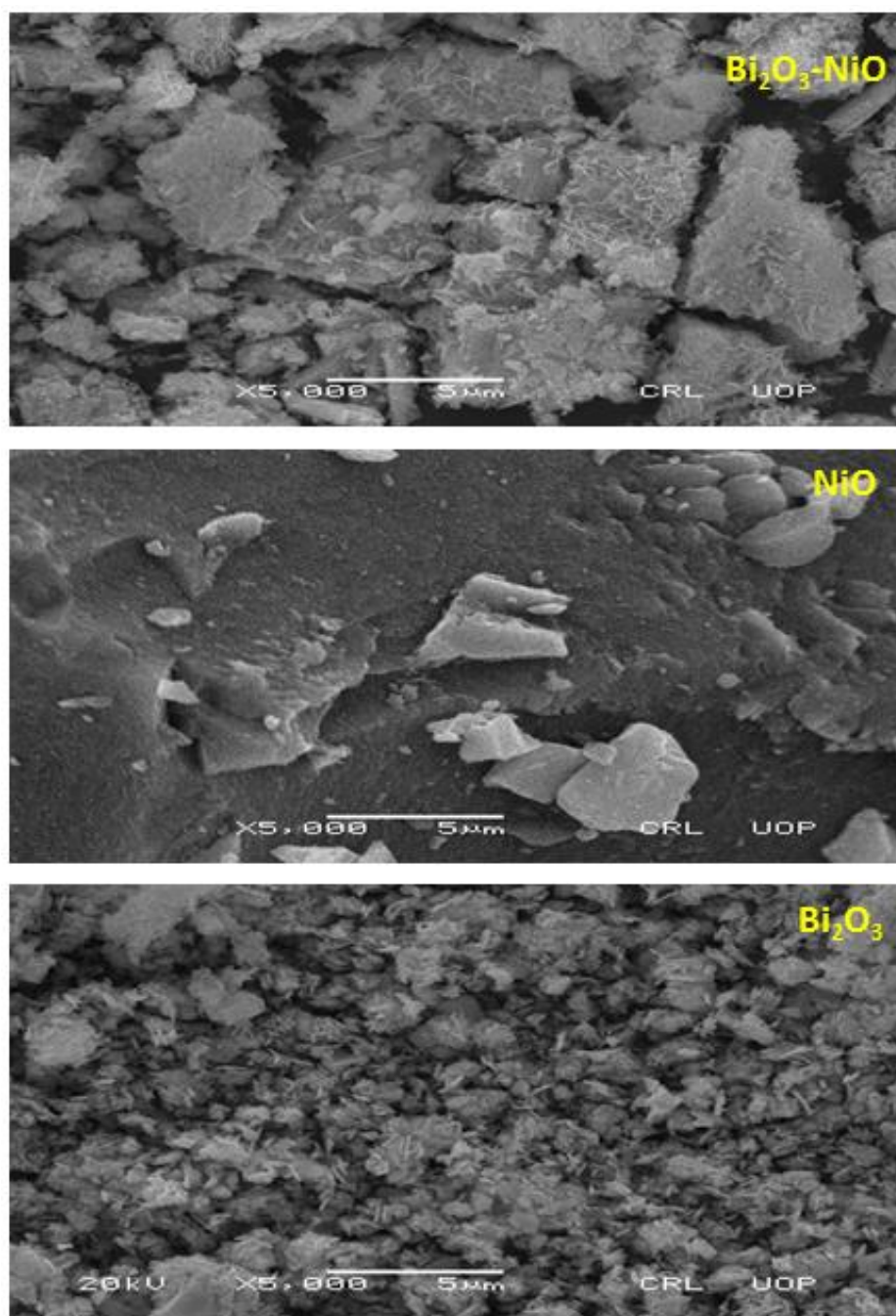


Figure 4. SEM analysis of Bi_2O_3 , NiO , and $\text{Bi}_2\text{O}_3\text{-NiO}$ samples.

3.2. Photocatalytic Activity

The comparison of the photocatalytic activity of Bi_2O_3 , NiO , and $\text{Bi}_2\text{O}_3\text{-NiO}$ was studied by performing separate photodegradation experiments using a 50 mL solution of methyl orange with a 100 mg/L concentration. The photodegradation profile of methyl orange dye in terms of M_t/M_0 (M_t : concentration or absorbance at a different time interval, M_0 : concentration or absorbance at zero time) is given in Figure 5. The data given in Figure 5 shows that the photodegradation of methyl orange dye observed with $\text{Bi}_2\text{O}_3\text{-NiO}$ is significantly higher than the photodegradation observed with pristine Bi_2O_3 and NiO . In terms of the percent photodegradation of methyl orange, the photocatalytic activity of $\text{Bi}_2\text{O}_3\text{-NiO}$, Bi_2O_3 , and NiO was observed as ~100%, 67%, and 46%, respectively. After vigilant analyses of the observed results, it was predicted that various factors contribute to the enhanced photocatalytic response of the $\text{Bi}_2\text{O}_3\text{-NiO}$ composite.

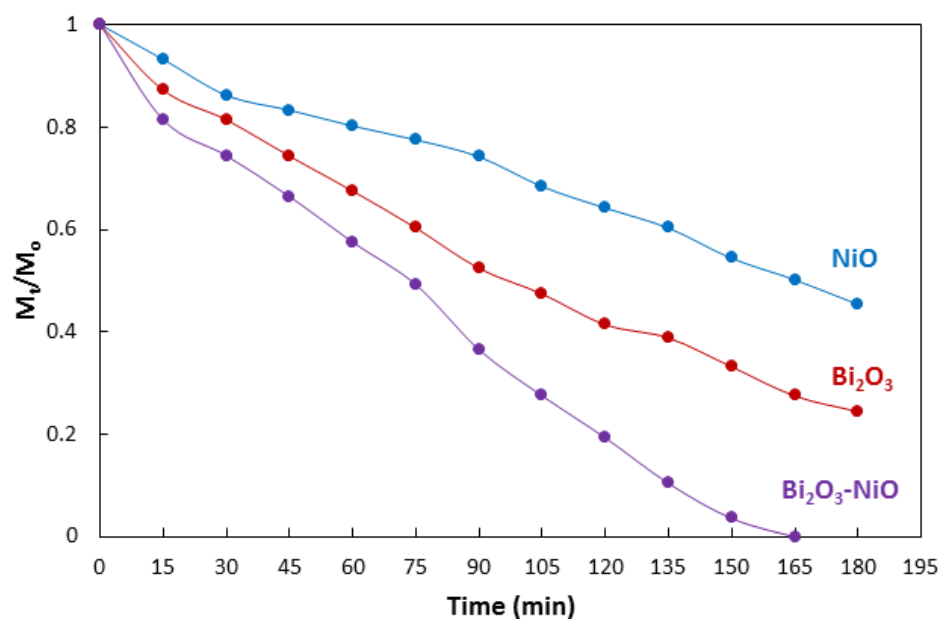


Figure 5. Comparison of photocatalytic activity of Bi_2O_3 , NiO, and $\text{Bi}_2\text{O}_3\text{-NiO}$ towards photodegradation of methyl orange.

The photocatalytic efficiency of a photocatalyst is significantly influenced by the separation, transfer, and recombination of the photo-induced positive holes and electrons. It is accepted that efficient separation and transfer of the positive holes and electrons suppresses the recombination of these charge carriers and ultimately causes a significant improvement in the photocatalytic activity of the photocatalyst. The higher photocatalytic activity of $\text{Bi}_2\text{O}_3\text{-NiO}$ than of its individual components in the present study is also attributed to the separation and transfer of the positive holes and electrons [48–50].

Methyl orange is a colored substance that absorbs light in the wavelength range of 340–530 nm of the UV-visible spectrum. Similarly, Bi_2O_3 , having a bandgap energy of 2.58 eV, also absorbs in the visible region at ~480 nm. Ordinary sunlight is composed of 10% ultraviolet light and 45% visible light in addition to other types of radiation. It shows that methyl orange masks the absorption region of Bi_2O_3 . Therefore, the available region of sunlight to be absorbed by the photocatalyst is only 300–340 nm. Hence, it is predicted that the photocatalytic degradation of methyl orange in the present study takes place via an indirect excitation process, unlike the normal photocatalytic process in which the photocatalytic degradation takes place via direct excitation. Furthermore, the inhibition of the recombination of positive holes and electrons through a synergic effect is also expected due to the composite photocatalyst. The irradiation of the reaction mixture (methyl orange and $\text{Bi}_2\text{O}_3\text{-NiO}$) causes excitation of the methyl orange due to the absorption of light. The excited methyl orange then transfers the absorbed energy to the Bi_2O_3 component of the photocatalyst. The transfer of this energy promotes the electrons from the VB to CB of Bi_2O_3 resulting in the formation of the exciton (a pair of positive holes and electrons). The positive holes are then diffused to the VB of NiO. As a result, the recombination of the positive hole and electron is inhibited. Similarly, the absorption of ultraviolet light causes the production of exciton in NiO as well. The photo-induced electron in the CB of NiO then flows to the CB of Bi_2O_3 [51–53]. The positive holes and electrons react with water and oxygen, respectively, and ultimately produce OH radicals. The OH radicals then take part in the degradation of methyl orange molecules. The formation of positive holes and hydroxyl radicals was verified experimentally using EDTA and t-BuOH as scavengers for charge carriers, respectively. It was noted that the photocatalytic activity of $\text{Bi}_2\text{O}_3\text{-NiO}$ decreased from 80% to 63 and 41% in the presence of EDTA and t-BuOH, respectively [54–56]. The proposed process can be summarized as follows (BN: $\text{Bi}_2\text{O}_3\text{-NiO}$). This is explained in Figure 6 as well.

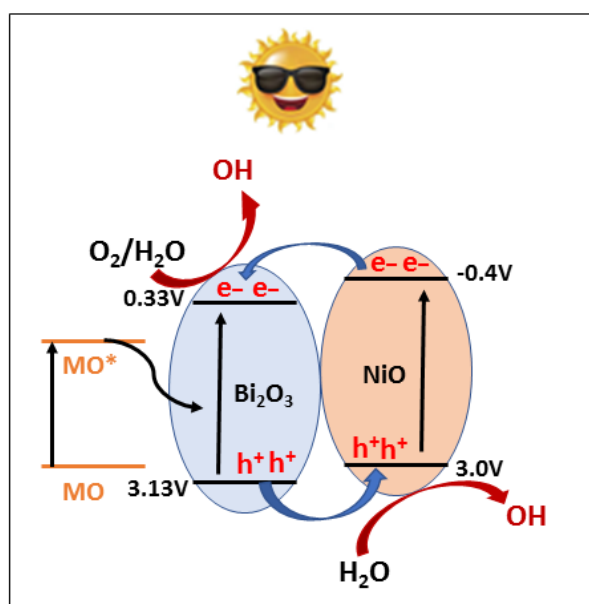
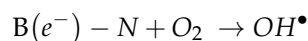
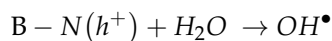
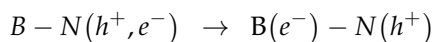
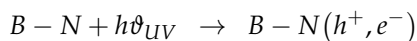
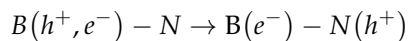
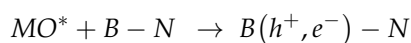


Figure 6. Mechanism of photocatalytic process.

It is necessary to mention that the excitation of NiO through the transfer of energy from methyl orange through indirect photocatalysis is less likely, as the energy absorbed by methyl orange is not sufficient to promote an electron from the VB to CB of the NiO counterpart of the composite photocatalyst. The higher photocatalytic activity of Bi₂O₃-NiO than of its individual components verifies the proposed assumptions.

Based on the proposed mechanism, the rate of photodegradation of MO in the present study is expressed in the following equation (M: concentration of methyl orange, θ_M : fraction of catalyst surface covered by methyl orange).

$$r = -\frac{d[M]}{dt} = k OH^\bullet \theta_M \quad (3)$$

As the amount of catalyst is constant and the reaction mixture is open to the atmosphere and continuously irradiated, the rate of reaction therefore does not depend on the concentration of OH radicals. Hence, the rate expression becomes

$$r = -\frac{d[M]}{dt} = k \theta_M \quad (4)$$

The surface of the catalyst covered by methyl orange is expressed in terms of Langmuir adsorption isotherm. Hence, the rate expression modifies as (K: Langmuir constant for adsorption of methyl orange, K_i : Langmuir constant for adsorption of degradation products, P_i : concentration of products):

$$r = -\frac{d[M]}{dt} = k \frac{KM}{1 + KM + \sum K_i P_i} \quad (5)$$

As

$$KM + \sum K_i [P]_i = M_0 = \text{Constant} \quad (6)$$

Therefore, the rate equation becomes

$$r = -\frac{d[M]}{dt} = k_{obs} M \quad (7)$$

where

$$k_{obs} = \frac{kK}{1 + M_0} \quad (8)$$

On integration of Equation (7)

$$\ln \frac{M_0}{M_t} = k_{obs} t \quad (9)$$

On re-arrangement of Equation (9), we obtain

$$M_t = M_0 e^{-k_{obs} t} \quad (10)$$

The photodegradation data of methyl orange over Bi_2O_3 , NiO , and $\text{Bi}_2\text{O}_3\text{-NiO}$ were analyzed according to kinetics Equation (10) using the non-linear method of analysis. The Microsoft Excel Solver add-in was used for non-linear analyses. Figure 7 shows the treatment of the degradation data according to kinetics Equation (10). The rate constants (k_{obs}) are given in Table 2. The rate constant for the photodegradation of MO in the presence of $\text{Bi}_2\text{O}_3\text{-NiO}$ was 3.2-fold and 1.7-fold of the rate constant with NiO and Bi_2O_3 , respectively.

Table 2. Rate constants determined by the non-linear method of analysis.

Catalyst	k_{obs}	R^2
NiO	0.0039	0.99
Bi_2O_3	0.0072	0.99
$\text{Bi}_2\text{O}_3\text{-NiO}$	0.0125	0.94

3.3. Dependence of Photocatalytic Activity on Catalyst Dosage

The optimization of catalyst dosage is also an important parameter in catalysis because the catalytic activity and rate of reaction significantly depend on catalyst dosage. Therefore, photocatalytic reactions were conducted with various catalyst dosages of $\text{Bi}_2\text{O}_3\text{-NiO}$ using a 50 mL (100 mg/L) solution of methyl orange. Figure 8 shows the photocatalytic activity of $\text{Bi}_2\text{O}_3\text{-NiO}$ towards the degradation of methyl orange after 120 min of reaction. It is evident that although the photodegradation increased with the catalyst dosage, the increase was not continuous. The catalytic activity initially significantly increased up to 0.08 g of the catalyst and then there was a slight increase with further increase in the catalyst dosage. On the other hand, the rate of reaction (degradation per gram of catalyst per minute) initially increased up to 0.08 g of the catalyst and then decreased with further additions. Hence, 0.08 g was chosen as an optimum catalyst dosage in this investigation. It is obvious that the number of active sites increases with catalyst dosage, therefore the catalytic activity also increases with catalyst dosage. However, a higher dosage of the catalyst creates a hindrance

to the penetration of photons due to scattering and turbidity, therefore, the catalytic activity decreased at the higher dosage [57,58].

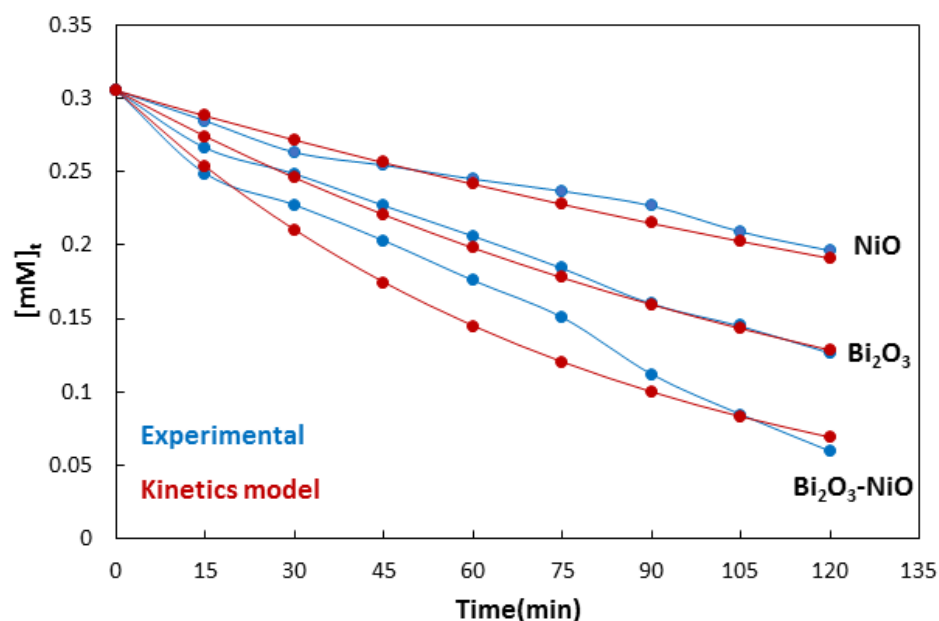


Figure 7. Kinetics analyses of degradation data of MO.

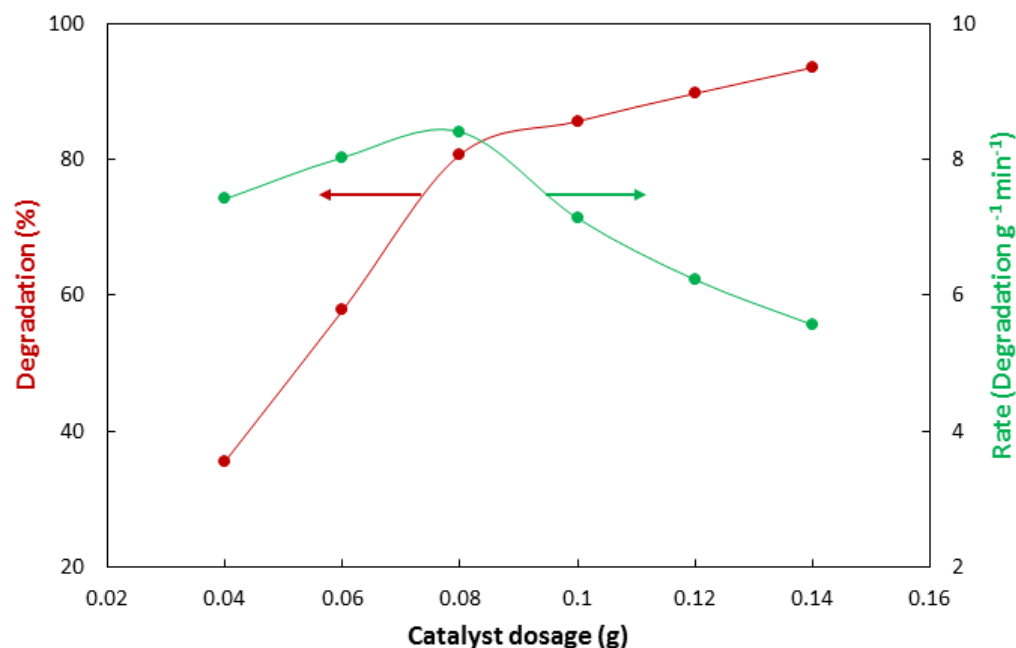


Figure 8. Dependence of photocatalytic activity on catalyst dosage.

3.4. Dependence of Photocatalytic Activity on Concentration of Methyl Orange

The initial concentration of dye significantly influences the efficiency of the photocatalytic process. Therefore, we investigated the dependence of the photocatalytic activity of $\text{Bi}_2\text{O}_3\text{-NiO}$ on the concentration of methyl orange. This investigation was accomplished by performing independent photodegradation experiments using 0.08 g $\text{Bi}_2\text{O}_3\text{-NiO}$ and 50 mL solution of methyl orange with concentrations of 50, 100, and 150 mg/L. The removal of dye due to adsorption was excluded by stirring the reaction mixture in the dark for 30 min. Figure 9 shows the obtained data. The data shows that the catalytic efficiency of $\text{Bi}_2\text{O}_3\text{-NiO}$ is inversely proportional to the concentration of methyl orange. The catalytic efficiency

of $\text{Bi}_2\text{O}_3\text{-NiO}$ towards the photodegradation of methyl orange was 100, 80, and 65% after 60 min of reaction with 50, 100, and 150 mg/L solutions of methyl orange, respectively. The obtained degradation data was analyzed for kinetics analysis according to kinetics Equation (10). The rate constants were found as 0.0227, 0.0125, and 0.0082 per minute for the photodegradation of 50, 100, and 150 mg/L solutions of methyl orange, respectively. An increase in the initial concentration of methyl orange decreased the photocatalytic performance. Generally, the rate of a reaction is directly proportional to the concentration of the reactant. However, in this study, we observed that the rate of reaction is inversely proportional to the concentration of methyl orange. It is because photons cannot penetrate the catalyst surface due to the intense color of the solution at higher concentrations. Furthermore, as the catalyst dose was kept fixed in all three experiments the concentration of hydroxyl radicals was therefore also expected to be the same in all experiments. The increase in the concentration of methyl orange causes a decrease in the number of hydroxyl radicals per molecule of methyl orange. Hence, the photocatalytic efficiency is inversely proportional to the initial concentration of methyl orange [59–62].

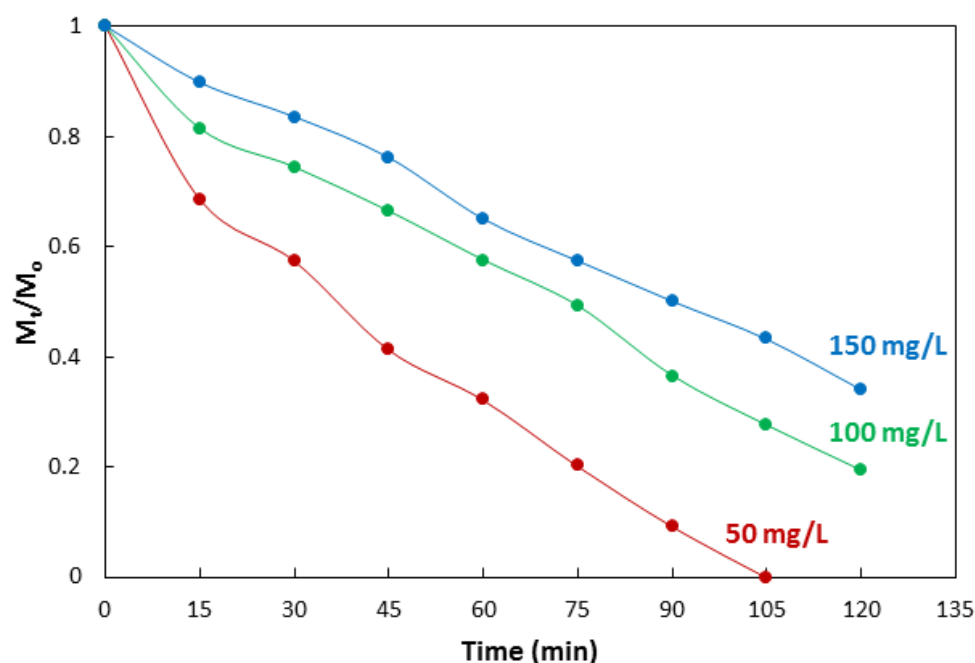


Figure 9. Dependence of photocatalytic activity on concentration of methyl orange.

3.5. Comparison of the Present Photocatalyst with Reported Photocatalysts

Various Bi_2O_3 -based photocatalysts have been reported for the photodegradation of dyes. Dhiman et al. [63] have reported the $\text{CoFe}_2\text{O}_4/\text{Bi}_2\text{O}_3/\text{NiO}$ photocatalyst for degradation of ofloxacin under visible light. They reported a 92% degradation of ofloxacin in 90 min using 100 mL of a 10 mg/L solution of ofloxacin. Shahzad et al. [64] have reported an organosilica-supported Bi_2O_3 photocatalyst for the degradation of methylene blue and methyl orange dyes. They obtained about 90% degradation of these dyes using 30 mL of a 20 mg/L solution of each dye. In another study, $\text{Co}_3\text{O}_4\text{-Bi}_2\text{O}_3$ was used as a catalyst for the photodegradation of rhodamine B dye with 92% degradation efficiency using a 100 mg/L solution of the dye [54]. Similarly, a 94% degradation of MB and MO over the $\text{NiO-Bi}_2\text{O}_3$ photocatalyst has been reported recently [65]. Banoth et al. [66] have prepared novel BiFeO_3 and $\text{BiFeO}_3\text{-Fe}_2\text{O}_3$ photocatalysts for the photodegradation of dyes. They tested their catalysts for the photodegradation of methylene blue dye. Almost complete degradation of methylene blue was obtained using 40 mg of catalyst and a 10 mg/L (100 mL) solution of methylene blue dye under visible light with a reaction duration of 70 min. Poorsajadi et al. [67] have synthesized a $\text{CuO-Bi}_2\text{O}_3$ nanocomposite via a hydrothermal method. After characterization, they evaluated the photodegradation of methylene blue

dye using the prepared nanocomposite as a photocatalyst. About 88% degradation of BM was obtained at an acidic pH using 0.2 g/L of CuO-Bi₂O₃ nanocomposite as a catalyst and a 10 mg/L solution of the dye. Zhang et al., [68] have studied the synthesis of Bi₂O₃@Zn-MOF composite via a hydrothermal method. They used the prepared composite for the degradation of rhodamine B dye. They found 97% degradation of the dye using a 50 mL (40 mL) solution of rhodamine B dye under visible light. In this study, we report Bi₂O₃-NiO as a photocatalyst for the degradation of methyl orange. The catalyst developed in this study is more effective for the degradation of dyes because the almost complete degradation of methyl orange was observed using a 100 mg/L (50 mL) solution of the dye under natural sunlight.

4. Conclusions

This study showed that photocatalytic activity can be improved with the formation of a heterojunction by coupling two suitable semiconductors. The enhanced photocatalytic performance of the heterojunction is attributed to an extended response in the wide region of the UV–visible spectrum. The heterojunction photocatalysts play an important role in the enhancement of drawbacks associated with conventional photocatalysts like poor light response and low efficiency. Hence, a 50 mL solution of methyl orange (100 mg/L) was completely degraded under sunlight irradiation in the presence of Bi₂O₃-NiO. In comparison, the photocatalytic activity of Bi₂O₃ and NiO was found to be 67 and 46%, respectively. The optimum catalyst dosage was found as 0.08g. The degradation data were analyzed according to first-order kinetics through the non-linear method of analysis using Solver software. The rate constant for the photodegradation of MO in the presence of Bi₂O₃-NiO was 3.2-fold and 1.7-fold of the rate constant with NiO and Bi₂O₃, respectively. The photocatalytic activity of Bi₂O₃-NiO was inversely related to the initial concentration of methyl orange. The catalytic efficiency of Bi₂O₃-NiO towards photodegradation of methyl orange was 100, 80, and 65% after 60 min of reaction with 50, 100, and 150 mg/L solutions of methyl orange, respectively.

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