Formulation of Bismuth (Bi\(_2\)O\(_3\)) and Cerium Oxides (CeO\(_2\)) Nanosheets for Boosted Visible Light Degradation of Methyl Orange and Methylene Blue Dyes in Water

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Abstract: Annealing of periodic mesoporous organosilica supported with bismuth (Bi@PMOS) and cerium (Ce@PMOS) nanoparticles was carried out to derive bismuth oxide (Bi\(_2\)O\(_3\)) and cerium oxide (CeO\(_2\)) nanosheets. The hydrothermal sol-gel method was used to synthesize hexagonal Bi@PMOS and Ce@PMOS. These PMOS provided an opportunity for bismuth and cerium to retain a hexagonal configuration alongside their traditional crystalline phases (tetragonal and cubic) in Bi\(_2\)O\(_3\) and CeO\(_2\) nanosheets. All produced materials were found to be dynamic under sunlight irradiation for the degradation of methylene blue (MB) and methyl orange (MO). However, the Bi\(_2\)O\(_3\) and CeO\(_2\) nanosheets showed better potential and photo-catalytic performances than Bi@PMOS and Ce@PMOS due to the presence of the unique blend of crystalline phases. The synthesized Bi@PMOS, Ce@PMOS, Bi\(_2\)O\(_3\), and CeO\(_2\) were structurally characterized by FTIR and XRD techniques. These showed characteristic vibrations of successfully loaded bismuth and cerium with hexagonal symmetry. EDX results confirmed the elemental detection of bismuth and cerium, while SEM images revealed the nanosheets in the synthesized materials. The optical response and detection of reactive species were carried out by photoluminescence (PL) and showed emissions at 700 nm. The PL data were also used to calculate band gaps of 3.72, 3.70, 3.35, and 2.88 eV for Ce@PMOS, Bi@PMOS, CeO\(_2\), and Bi\(_2\)O\(_3\), respectively. A UV/visible spectrophotometer scanned the photocatalytic competences of the synthesized nanomaterials through the degradation of MB and MO dyes. Then, 10 mg of Bi@PMOS and Ce@PMOS degraded 15 mg and 8.4 mg of MB and 10.8 mg and 8 mg of MO, respectively, in 20 mg/L solutions. However, equivalent quantities of Bi\(_2\)O\(_3\) and CeO\(_2\) (10 mg of each) exhibited more efficient photocatalysis of the 20 mg/L solutions of MB and MO, degrading 18.4 mg and 15.4 mg, and 12.4 mg and 17 mg, respectively, in only 1 h. The Bi\(_2\)O\(_3\) and CeO\(_2\) photocatalysts were regenerated and their photodegradation results were also recovered. Bi\(_2\)O\(_3\) and CeO\(_2\) showed only 10% and 8% (for MB), and 8% and 10% (for MO) decline in catalytic efficiency, respectively, even after four consecutive recycles. These results demonstrate that these materials are dynamic, long-lasting photocatalysts for the rapid degradation of azo dyes in contaminated water.

Keywords: Bi\(_2\)O\(_3\) and CeO\(_2\) nanosheets; long-lasting photocatalysts; methylene blue; methyl orange; photodegradation; water decontamination
1. Introduction

The quality of freshwater reservoirs is falling due to the rising needs of residences and industry, harming aquatic life. A large number of investigations are now being reported to prepare suitable photocatalysts for treating textile industry effluents [1]. These effluents are considered more harmful than other toxins and must be removed using an advanced water purification treatment [2]. About half of the dye added for dyeing textile fibers does not adhere to the fibers and ends up as a contaminant in the aquatic environment [3]. Usage of unhygienic water resulted in a variety of ailments in human beings [4]. Undiagnosed and untreated dye pollutants in water not only damage the surface water, but they also block the transmission of light, which has negative consequences for water bodies [5]. Various techniques are used to remove these pollutants from water, such as the biodegradation of methylene blue (MB) by polyvinyl pyrrolidone supported with carboxymethyl starch [6], photodegradation [7], and adsorption [8]. Efficient removal of azo dyes cannot be achieved by traditional adsorption, enzymatic degradation, and ion exchange procedures because these methods produce secondary harmful emissions, such as poisonous fumes and sludge, which necessitate further processing [9,10].

Alternative strategies, such as ultrasound-supported advanced oxidation and heterogeneous photocatalysis with micro semiconductors, have emerged as potential technologies for removing water contaminants [11]. Fenton-sonolysis under different conditions of catalysis can effectively reduce the dye molecules [12]. Moreover, doped semiconducting nanomaterials, such as layered metal oxides [13] and metal-assisted layered-double hydroxide (LDH) [14], provide several possible heterojunction sites for photocatalysis. The photocatalytic reaction is a progressive electrochemical reaction capable of completely degrading organic pollutants existing in textile wastewaters into CO₂ and H₂O by generating hydroxyl free radicals (·OH) and superoxides (O₂⁻) in situ [15].

Among several photo-catalysts, CeO₂ has been acknowledged as a particularly effective photocatalyst due to the redox potential of the Ce⁴⁺/Ce³⁺ pair, its non-toxicity, chemical stability, photo-corrosion resistance, and finely tunable band gap [16,17]. However, CeO₂’s high band gap confines its absorption largely to the UV region, but UV light represents only about 3–5% of incident radiation reaching the earth, while visible light accounts for roughly 45%. Consequently, extending CeO₂’s light absorption to the visible region would be extremely useful [18]. For this purpose, several approaches (coupling and doping) for shifting the absorption onset have been examined. Researchers have also identified bismuth oxide as another flexible photocatalyst [19] with excellent photocatalytic properties. Incorporating graphitic carbon nitride into bismuth and cerium oxide [20] nanoparticles (NPs) resulted in a material that showed enormous photodegradation [21], while cerium oxide NPs after doping with different metals demonstrated high photocatalysis [22]. This was due to an increase in Ce⁴⁺ to Ce³⁺ conversion, which resulted in more production of active species and increased photocatalytic activity [23]. Bismuth is a trivalent and non-toxic alternative to lead, for constructing “lead-free” pipes as required by the Safe Drinking Water Act [24]. Additionally, microspheres of blended porous BiO₂/ CeO₂ [25] and bismuth oxy-halide composites containing graphene oxide have also been found to have stronger photocatalytic activities [26].

Every effort has been made to maximize the use of solar energy by developing photo-catalysts that perform well in visible light. Photocatalytic degradation of azo dyes is an effective method for industrial wastewater decontamination [27]. Intense absorption of visible light, enhanced photo-generated methods, and improved catalytic material stability can all help to boost photocatalytic efficiency [28]. In azo dyes, methylene blue (MB) and methyl orange (MO), which are used as dyes in many industries, such as the textile and paper industries and agricultural sector, are the most frequently investigated contaminants used to analyze photocatalytic degradation [29].

Untreated, these dyes eventually end up in water bodies, posing serious health dangers to aquatic life and humans [30]. Hence, removing such recalcitrant pollutants from the water is strongly advised. For this purpose, in the present research work, hex-
agonally shaped solid Bi2O3 and CeO2 nanosheets were prepared from annealed Bi@PMOS and Ce@PMOS. Generally, Bi2O3 and CeO2 developed stable tetragonal and cubic crystalline phases, but, in this approach, the Bi@PMOS and Ce@PMOS provided hexagonal foundations to Bi2O3 and CeO2 alongside the traditional crystalline phases. These unique variations in structure enable Bi2O3 and CeO2 nanosheets to use direct sunlight for boosting the photodegradation of MB and MO during water purification on industrial and domestic levels.

2. Results and Discussion

2.1. Materials Synthesis and Characterization

Bi2O3 and CeO2 nanosheets were formed by annealing Bi@PMOS and Ce@PMOS at 550 °C, through which most of the organic content was decomposed. The percentage yield of the obtained Bi2O3 and CeO2 nanosheets were calculated from:

\[
\text{Yield of metal oxide (\%)} = \frac{\text{Weight of Final metal oxide}}{\text{Total weight of metal supported PMOS (Bi@PMOS and Ce@PMOS)}} \times 100 \tag{1}
\]

XRD and FTIR studies were used to examine the structure of the Bi@PMOS and Ce@PMOS, Bi2O3, and CeO2 nanosheets. Generally, tetragonal and cubic crystalline phases are present in more stable Bi2O3 and CeO2 (JCPDS no. 27-0050 and JCPDS no. 34-0394, respectively) [27,31] at low temperatures. These crystalline planes, corresponding to diffraction angles of 220, 221, and 400, were also found in the Bi2O3 and CeO2 nanosheets in this study (Figure 1a,b). However, the annealing of Bi@PMOS and Ce@PMOS at high temperatures triggered a phase change in Bi2O3 and CeO2 and retained their novel hexagonal base framework. Intense diffraction peaks were recorded corresponding to the (100), (200), and (220) hexagonal reflection planes [32]. The observed diffraction data showed that Bi@PMOS and Ce@PMOS provided a hexagonal foundation which is shown by the results of Bi2O3 and CeO2 nanosheets in Figure 1a,b. Scherer’s equation was used to determine the crystallite size of the most intense diffractions of Bi2O3 and CeO2, which were found to be 8.34 nm and 8.31 nm, respectively. These results showed a further reduction in particle size of Bi2O3 and CeO2 after annealing Bi@PMOS and Ce@PMOS (average crystallite size = 12.30 nm).

Figure 1. (a,b) XRD and (c,d) FTIR spectra of Bi@PMOS, Ce@PMOS, Bi2O3 and CeO2.
The FTIR spectrum of Bi$_2$O$_3$ showed characteristic peaks at 532, 705, 1002, 1363, and 3444 cm$^{-1}$, while the spectrum of CeO$_2$ has peaks at 472, 519, 793, 1011, 1370, and 3415 cm$^{-1}$ (Figure 1c,d). The FTIR spectra demonstrated the successful loading of bismuth and cerium oxides in Bi@PMOS and Ce@PMOS. Major vibrational peaks were recorded at 472, 519, and 532 cm$^{-1}$, demonstrating the presence of bismuth and cerium connections with oxygen in the material structures. Moreover, significant vibrational peaks at 705–793 cm$^{-1}$ and 1002–1011 cm$^{-1}$ signify the Si-O-Si stretching in hexagonal PMOS materials. However, the presence of these vibrations in the spectra of the annealed Bi$_2$O$_3$ and CeO$_2$ suggests an indication of silicon replacement by cerium and bismuth in the hexagonal silicon framework. However, C–H bending peaks at 1363 cm$^{-1}$, 1370 cm$^{-1}$ and stretching vibrations at 3415 cm$^{-1}$ and 3444 cm$^{-1}$ were also recorded [33]. All these spectral results were evidence of metal–oxygen interactions in the organo-silica framework [34,35].

SEM images exhibited membrane-like sheet organizations in Bi@PMOS and Ce@PMOS that were retained by the Bi$_2$O$_3$ and CeO$_2$:nanosheets, as shown in Figure 2. Moreover, EDX results (in Figure 3c,d) confirmed the loading of bismuth and cerium in Bi@PMOS and Ce@PMOS to formulate Bi$_2$O$_3$ and CeO$_2$:nanosheets for efficient photocatalytic degradation of MB and MO dyes.

Figure 2. SEM images of (a) Bi@PMOS, (b) Ce@PMOS, (c) Bi$_2$O$_3$, and (d) CeO$_2$. 
In addition, Tauc plots (in Figure 3a,b showed an energy difference between the valence band (VB) and conduction band (CB) in the Bi$_2$O$_3$ and CeO$_2$ materials. These band gaps were determined to be 2.88 eV and 3.35 eV for Bi$_2$O$_3$ and CeO$_2$, respectively. These pointed towards a ready availability of electrons during photodegradation.

During photocatalysis, Bi@PMOS, Ce@PMOS, Bi$_2$O$_3$, and CeO$_2$ showed 74.7, 41.4, 91.3, and 76.2% photoreduction of MB solutions and 53.2, 39.4, 61.8, and 85% photoreduction of MO solutions, respectively, as shown in Figure 4a,b. The results showed enhanced production of superoxides and hydroxyl reactive species by Bi$_2$O$_3$ and CeO$_2$, which enhanced their photocatalytic efficiency [36].
2.2. Kinetic Study

Linear fit photocatalytic models showed that the reaction was pseudo-first-order (Figure 4c,d). The rate constant ($k$) can be used as a measure of the degradation frequency of MB and MO by Bi@PMOS, Ce@PMOS, Bi$_2$O$_3$, and CeO$_2$. The observed rate constants values of Bi@PMOS and Ce@PMOS towards the photoreduction of MB and MO were 0.0203 and 0.0101, and 0.0079 and 0.0085 min$^{-1}$, respectively, which were increased in the case of Bi$_2$O$_3$ and CeO$_2$ and found to be 0.0375 and 0.0166, and 0.0234 and 0.0303 min$^{-1}$, respectively. The photodegradation activity of Bi$_2$O$_3$ and CeO$_2$ under visible light was enhanced after removing organic moieties from Bi@PMOS and Ce@PMOS by annealing. The comparative photocatalytic efficiencies of all the synthesized catalysts are given in Table 1 and demonstrate the effectiveness of the novel Bi$_2$O$_3$ and CeO$_2$ nanosheets under sunlight.

Furthermore, PL spectra acquired at an excitation wavelength of 400 nm, shown in Figure 5a,b, exhibited visible fluorescence at 700 nm. The more intense spectra of Bi$_2$O$_3$ and CeO$_2$ compared to those of Bi@PMOS and Ce@PMOS provide the reason for the outstanding catalytic performance, demonstrating the availability of a large number of active sites where charge can be transferred [37] efficiently during photodegradation.
Additionally, the observed PL results shown in Figure 5c,d revealed intense fluorescence in the Bi₂O₃ and CeO₂ trials in which a high generation of hydroxyl free radicals (OH) caused the formation of hydroxyl terephthalic acid (HOPTHA), a fluorescent chemical species [38] from PTHA. The Bi₂O₃ trial showed relatively high fluorescence, indicating the major role of these species in the production of reactive hydroxyl free radicals (OH).

Figure 5. (a,b) Photoluminescence of Bi@PMOS, Ce@PMOS, Bi₂O₃, and CeO₂. (c,d) Photoluminescence of HOPTHA produced by PTHA, Bi@PMOS, Ce@PMOS, Bi₂O₃, and CeO₂.
### Table 1. Comparison of photocatalytic efficiencies with the cited literature.

| Catalyst (Amount) | MB/MO (Amount) | % Reduction | Time (Hour) | [K] (Min⁻¹) | Reference |
|------------------|----------------|-------------|------------|-------------|-----------|
| PAM-TiO₂/Porous TiO₂ beads (5 mg/10 cm³) | (25 mg/L) | 16.2/32.8 | 01 | 0.00016/0.0024 | [36] |
| Ag-PMOS (10 mg/ 30 cm³) | (20 mg/L) | 81/48 | 01 | 0.025/0.0046 | [39] |
| ZnO-PMOS (10 mg/ 30 cm³) | (20 mg/L) | 47/57 | 01 | 0.0020/0.0062 | [39] |
| GF/TiO₂ NTA 1x4 Cm²-electrode | (5 mg/L) | 65.9 | 02 | 0.0088 | [40] |
| Bi@PMOS (10 mg/ 30 cm³) | (20 mg/L) | 74.7/53.2 | 01 | 0.0233/0.0101 | [41] |
| Ce@PMOS (10 mg/ 30 cm³) | (20 mg/L) | 41.4/39.4 | 01 | 0.0079/0.0085 | [41] |
| Bi₂O₃ (10 mg/ 30 cm³) | (20 mg/L) | 91.3/61 | 01 | 0.0375/0.0166 | Present study |
| CeO₂ (10 mg/ 30 cm³) | (20 mg/L) | 76.2/85 | 01 | 0.0234/0.0303 | Present study |

### 2.3. Projected Photodegradation Mechanism

The formation of highly reactive free radicals required energy, which can be provided by the photons in sunlight [42–44]. The incident photons promoted electrons from the valence band (VB) into the conduction band (CB) and the resulting electron (e⁻) hole (h⁺) pairs played a vital role in the photocatalysis as shown below,

**Generation of electron–hole pairs by photon absorption**

\[
\text{Bi}_2\text{O}_3 + h\nu \rightarrow \text{Bi}_2\text{O}_3 (h^+) \text{ VB} + (e^-) \text{ CB} \quad (2)
\]

\[
\text{CeO}_2 + h\nu \rightarrow \text{CeO}_2 (h^+) \text{ VB} + (e^-) \text{ CB} \quad (4)
\]

**Generation of super radicals**

\[
(e^-) \text{ CB} + \text{O}_2 \rightarrow \text{O}_2^-. \quad (6)
\]

**Generation of hydrogen peroxide (H₂O₂)**

\[
\text{O}_2^- + \text{H}_2\text{O} \rightarrow \text{OOH} + \text{OH}^- \quad (7)
\]

\[
\text{OOH} + \text{e}^- \rightarrow \text{OOH}^- \quad (8)
\]

\[
\text{OOH} + \text{O}_2^- \rightarrow \text{OOH}^- + \text{O}_2 \quad (9)
\]

\[
\text{OOH}^- + \text{H}^+ \rightarrow \text{H}_2\text{O}_2 \quad (10)
\]

**Generation of hydroxyl free radicals (OH)**

\[
\text{H}_2\text{O}_2 + h\nu \rightarrow \text{OH} + \text{OH}^- \quad (11)
\]

\[
\text{H}_2\text{O}_2 + \text{O}_2^- \rightarrow \text{OH} + \text{OH}^- \quad (12)
\]
\[ \text{Bi}_2\text{O}_3 (\text{h}^+) + \text{CeO}_2 (\text{h}^+) + \text{OH}_2/\text{H}_2\text{O} \rightarrow \text{OH} + \text{Bi}_2\text{O}_3 + \text{CeO}_2 \]  

(Degradation of methylene blue and methyl orange dyes by reactive radicals)

MB + O\(_2\)  Principal degradation  
MB + OH  Secondary degradation  
MO + OH  Principal degradation  
MO + O\(_2\)  Secondary degradation  

Degraded Products

\(\text{Bi}_2\text{O}_3\) showed a substantial reduction in MB absorption [43,45], whereas \(\text{CeO}_2\) was responsible for a substantial reduction in MO absorption under visible light irradiation [25]. \(\text{Bi}_2\text{O}_3\) and \(\text{CeO}_2\) provided a longer time for the recombination of electron (e\(_{-}\)) hole (h\(^{+}\)) pairs, leading to higher degradation rates for MB and MO [46].

2.4. Recycling of Catalysts

The \(\text{Bi}_2\text{O}_3\) and \(\text{CeO}_2\) nanosheets were found to continue to be efficient photocatalysts when reused after their recovery as shown in Figure 6.

Figure 6. %Photoreduction in the absorption of MB and MO by recycled (a,b) \(\text{Bi}_2\text{O}_3\) and (c,d) \(\text{CeO}_2\) under visible light.

2.4.1. UV/Visible Absorption Study

The used trial samples were centrifuged at 6000 rounds per minute (rpm) for 20 min. The samples were then collected from the centrifuge tubes, washed with distilled water, and kept at 100 °C in an oven for 12 h. Dry regenerated catalysts (\(\text{Bi}_2\text{O}_3\) and \(\text{CeO}_2\)) were used again, as described in previous photocatalytic experimental conditions and repeated over four cycles to observe their regenerated photodegradation competences. The absorption variations were examined using UV/vis spectrophotometry, as shown in Figure 6.
2.4.2. Photoreduction of Methylene Blue and Methyl Orange (%)

The regenerated Bi₂O₃ showed 90, 88, 85, and 81%, and 61, 59, 56, and 54% photo-degradations of MB and MO, respectively; while the figures for the regenerated CeO₂ were 76, 74, 72 and 69%, and 84, 81, 78, and 75% (Figure 6). These results indicated only a minor decline in the degradability of Bi₂O₃ and CeO₂ due to extended electron hole pair recombination time [26,47], and, hence, demonstrated their long life for utilization in photocatalysis.

3. Experimental Section

3.1. Materials

Bismuth oxide (Bi₂O₃) 99%, cerium oxide (CeO₂) 99%, sodium borohydride (NaBH₄) 99%, potassium chloride (KCl), sodium hydroxide (NaOH), analytical grade ethanol (C₂H₅OH), 85% methyl orange (MO), 82% methylene blue (MB), 98% terephthalic acid (PTHA), 37% hydrochloric acid (HCl), and 98% 3-methacryloxypropyl trimethoxysilane (C₁₀H₂₀O₅Si) were sourced from Sigma Aldrich. The 99% sodium silicate (Na₂SiO₃) and 95% polysorbate (C₆₄H₁₂₄O₂₆) were purchased from JEB.CHEM and Bio-World, respectively. OSAKA deionized water was used throughout the synthesis.

3.2. Synthesis of Bismuth (Bi₂O₃) and Cerium (CeO₂) Nanosheets

In the present research work, bismuth and cerium-supported periodic mesoporous organo-silicates (Bi@PMOS and Ce@PMOS) [41] were used to synthesize bismuth (Bi₂O₃) and cerium (CeO₂) nanosheets in powder form. First, periodic mesoporous organo-silicates (PMOS) were prepared through a sol-gel method. A micellar solution was created by mixing 0.6 g of HCl and 0.6 g of polysorbate in 120 g of water with constant stirring at room temperature. PMOS was then formed by adding 0.9 g of sodium silicate and 1.6 g of 3-methacryloxy propyl trimethoxysilane to the micellar solution and aging the solution at 95 °C for 24 h. The polysorbate network was then detached using 400 g of ethanol. The resulting colloidal solution was filtered, washed (with distilled water) and dried (at 95 °C for 12 h) to obtain the solid PMOS product. Next, 0.8 g of PMOS slurry was sonicated for 4 h and 0.1 g of bismuth and cerium oxides were separately added and adsorbed. The addition of 0.2 g of sodium borohydride facilitated the formation of bismuth (Bi₂O₃) and cerium (CeO₂) oxides from Bi@PMOS and Ce@PMOS [41]. The solid products (Bi@PMOS and Ce@PMOS) were obtained by filtering, washing (with distilled water), and drying (at 100 °C for 12 h). Finally, annealing of dried Bi@PMOS and Ce@PMOS in a furnace at 550 °C for 5 h obtained nanosheets of Bi₂O₃ and CeO₂. The complete synthetic scheme is shown in Figure 7.
3.3. Photocatalytic Efficiencies of Bi@PMOS, Ce@PMOS, Bi$_2$O$_3$, and CeO$_2$ for the Degradation of Methylene Blue and Methyl Orange Dyes

Photocatalytic experiments were carried out to examine the catalytic competences of Bi@PMOS, Ce@PMOS, Bi$_2$O$_3$, and CeO$_2$ under irradiation with sunlight using 30 cm$^3$ of 20 ppm solutions of MB and MO. After adjusting the pH of each solution to 2, 10 mg of the catalyst under test was added and the entire arrangement was placed in a dark box for 1 h to achieve adsorption–desorption equilibrium. The absorption performance in the dark was recorded using a UV/vis spectrophotometer. After that, the samples were irradiated by sunlight and scanned by the UV/vis spectrophotometer over regular time intervals of 0, 12, 24, 36, 48, and 60 min. The results showed significant reductions in the absorbance of MB and MO solutions, as shown in Figures S1 and S2.

3.4. Examining the Generation of Hydroxyl Free Radicals (·OH)

Several reactive species are produced during the photodegradation process, with superoxides and hydroxyl free radicals being the most important. The production of a large number of hydroxyl free radicals by Bi$_2$O$_3$ and CeO$_2$ was monitored by performing a photoluminescence experiment study. The experiment was conducted using NaOH (0.01M), terephthalic acid (PTHA) (3 mM), and KCl (0.1 M) in a 4 cm$^3$ aqueous solution. A comparison test was performed by adding 2 mg of synthesized catalysts and irradiating with visible light with constant stirring for 30 min. The PL spectra of all the trials are shown in Figure 5c,d.

3.5. Characterizations

Fourier transform infrared (FTIR) spectroscopy was used for the identification of different functional moieties in the synthesized materials. X-ray diffractometry (XRD) revealed the structural characteristics of the Bi$_2$O$_3$ and CeO$_2$ nanosheets. Ultra-violet visible (UV/vis.) spectrophotometry and photoluminescence (PL) were used to investi-
gate the optical behavior, while EDX scanning electron microscopy (EDX-SEM) examined the elemental elucidation and surface texture of the Bi$_2$O$_3$ and CeO$_2$ nanosheets.

4. Conclusions

The present research work consisted of the revised study of Bi@PMOS and Ce@PMOS for the synthesis of Bi$_2$O$_3$ and CeO$_2$ nanosheets by annealing. The porous surfaces of Bi@PMOS and Ce@PMOS provided a foundation for the large surface area in Bi$_2$O$_3$ and CeO$_2$. The successful loading of bismuth and cerium in Bi@PMOS and Ce@PMOS was validated by XRD, PL, FTIR, and EDX-SEM. Furthermore, the photodegradation efficiencies of the annealed Bi$_2$O$_3$ and CeO$_2$ (91.3% and 76.2% toward MB, and 61.8% and 85% toward MO, respectively) demonstrated their improved capabilities in a short time of 1 h. In addition, the improved photodegradability of regenerated Bi$_2$O$_3$ (90, 88, 85, and 81% and 61, 59, 56, and 54% photoreduction of MB and MO, respectively) and CeO$_2$ (76, 74, 72, and 69%, and 84, 81, 78, and 75% photodegradation of MB and MO, respectively) were also demonstrated. This retained photocatalytic activity revealed the long-term durability of Bi$_2$O$_3$ and CeO$_2$ in photocatalysis. This novel approach to forming energetic Bi$_2$O$_3$ and CeO$_2$:nanosheets with enhanced photodegradation performance has created materials effective in the reduction of azo dyes water pollution. The present research work also offers the prospect of being effective for loading other useful chemical moietyes by PMOS, as well as for use in the medicinal and industrial fields.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/catal12101197/s1, Figure S1: Absorption reduction of (a,c) methylene blue and (b,d) methyl orange by (a,b) Bi@PMOS and (c,d) Bi$_2$O$_3$ under visible light; Figure S2: Absorption reduction of (a,c) methylene blue and (b,d) methyl orange by (a,b) Ce@PMOS and (c,d) CeO$_2$ under visible light.

Author Contributions: K.S. and N.A.K. performed all the experimental work. A.u.R. supervised this work and revised the manuscript. M.I.K. and A.S. polished the manuscript throughout. J.F.-G. contributed to the writing of the manuscript and co-supervised the research. All authors discussed the results and contributed to the final manuscript. All authors have read and agreed to the published version of the manuscript.

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