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Fe-doped BiVO₄/g-C₃N₄ Heterojunction for Efficient Tetracycline Degradation under Low-Intensity UV Irradiation

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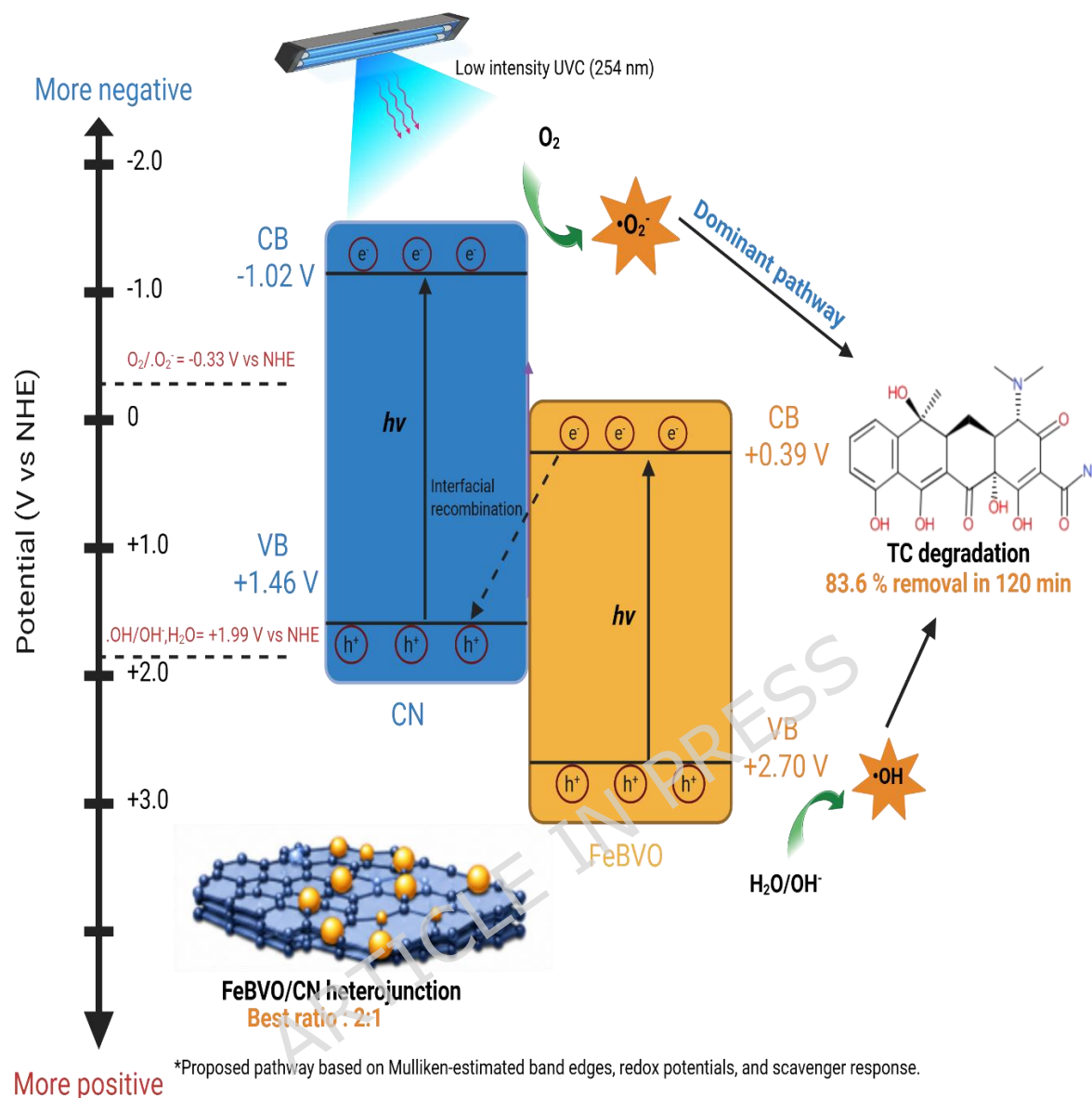
Abstract

Tetracycline (TC) contamination remains difficult to control because conventional wastewater treatment processes often do not fully remove persistent antibiotic residues. Although Fe doping and BiVO₄/g-C₃N₄ heterojunction formation have been reported previously, the composition-dependent behavior of Fe-doped BiVO₄/g-C₃N₄ photocatalysts under low-intensity UVC irradiation remains insufficiently clarified. This study developed 3% Fe-doped BiVO₄/g-C₃N₄ (FeBVO:CN) heterojunction photocatalysts with different FeBVO:CN mass ratios and evaluated their TC degradation performance, operating conditions, reactive-species involvement, reusability, and preliminary synthesis cost. The composites were synthesized by hydrothermal treatment followed by calcination and characterized using X-

ray diffraction, Fourier transform infrared spectroscopy, ultraviolet-visible spectroscopy, and field emission scanning electron microscopy. Among the prepared samples, FeBVO/CN-2:1 showed the highest photocatalytic activity, achieving 83.6% TC degradation within 120 min at 5 mg L⁻¹TC, 200 mg L⁻¹ catalyst loading, and pH 10. The degradation followed pseudo-first-order (PFO) kinetics with a rate constant of 0.01509 min⁻¹. Scavenger experiments suggested that •O₂⁻-related pathways exerted the strongest influence, while •OH and h⁺ also contributed to TC transformation. The catalyst retained 77.1% degradation efficiency after three cycles, indicating preliminary reusability under the tested conditions. This work advances the field by linking FeBVO:CN composition with structural features, optical response, photocatalytic activity, reactive-species behavior, and practical operating constraints under low-intensity UVC irradiation. Mulliken-estimated band-edge positions, together with the scavenger results, supported a proposed Z-scheme-like charge-transfer pathway as the more thermodynamically consistent interpretation of the observed reactive-species behaviour.

Keywords: Fe-doped BiVO₄; g-C₃N₄; heterojunction; tetracycline degradation; photocatalysis

Graphical Abstract



Highlights

- FeBVO:CN heterojunction was synthesized successfully.
- The 2:1 composite achieved 83.6% TC degradation under low-intensity UV.
- Scavenger tests showed $\bullet O_2^-$ as the main reactive species.

1.0 Introduction

The widespread occurrence of antibiotic residues in aquatic environments has become a serious environmental concern because these compounds are biologically active, chemically persistent, and continuously introduced through domestic, hospital, livestock, and aquaculture effluents [1–3]. Conventional wastewater treatment plants remain essential for reducing organic load and nutrients, yet they are not designed to fully remove many

antibiotics and other contaminants of emerging concern. As a result, residual concentrations often persist in treated effluents and enter rivers, lakes, and other receiving waters, where they can disrupt microbial balance and contribute to long-term ecological stress. Among these pollutants, tetracycline (TC) is an important target because it is one of the most widely used antibiotic classes in human therapy and animal production. Its frequent use, incomplete metabolism, and continuous discharge have led to repeated detection in aquatic systems [4]. TC is also difficult to manage because it exhibits high persistence, limited biodegradability, and strong interaction with environmental matrices, which prolongs its residence in water bodies. Beyond direct toxicity, the presence of TC residues promotes the spread of antibiotic-resistant bacteria and resistance genes, making its removal a matter of both environmental protection and public health control [5,6].

Photocatalysis is a promising advanced oxidation approach for antibiotic degradation because semiconductor photocatalysts generate reactive oxygen species (ROS) under light irradiation. These species, including superoxide radicals ($\bullet\text{O}_2^-$), hydroxyl radicals ($\bullet\text{OH}$), and photogenerated holes (h^+), attack organic pollutant structures and promote molecular transformation [7–9]. Compared with adsorption, photocatalysis offers pollutant degradation rather than only phase transfer. Compared with chemical oxidation, it operates under milder conditions and does not require continuous addition of strong oxidants. Recent studies have therefore focused on heterojunction photocatalysts that improve light absorption, reduce charge recombination, and enhance ROS-mediated degradation of TC and related organic pollutants [10–20]. Low-intensity ultraviolet C (UVC) irradiation is relevant for photocatalytic wastewater treatment because it offers a lower-power irradiation condition than high-intensity UV systems while still providing sufficient photon energy to activate suitable semiconductor photocatalysts. This operating mode is attractive for practical treatment because energy consumption remains one of the main barriers to photocatalytic scale-up. At the same time, low photon flux limits charge-carrier generation and reduces degradation efficiency. This creates a need for photocatalysts with stronger photon utilization, improved interfacial contact, and stable ROS generation under low-intensity irradiation.

Bismuth vanadate (BVO) is a promising photocatalyst because of its chemical stability, low toxicity, and suitable electronic structure for photoinduced redox reactions [21–23]. The monoclinic phase is widely regarded as the more photoactive form, yet pristine BVO often suffers from rapid electron-hole recombination, limited charge mobility, and modest surface reaction efficiency [24–26]. Metal doping has been used to address these shortcomings, and Fe incorporation has emerged as a useful strategy for tuning the electronic structure of BVO and improving charge separation behavior [24,25,27]. In our previous study, 3% FeBVO has shown improved photocatalytic performance with reduced band gap [28], but the gains

achieved by doping alone do not fully resolve the persistent limitations of carrier recombination, conductivity, and particle aggregation in aqueous systems. These remaining constraints indicate the need for a second component that can promote interfacial charge migration and improve catalyst utilization during pollutant degradation.

Graphitic carbon nitride (CN), is a suitable coupling partner for this purpose because it is a chemically stable, metal-free polymeric semiconductor with a layered structure and abundant surface nitrogen functionalities [29–31]. When integrated with BVO-based materials, CN can act as an electron-accepting and charge-transport platform, thereby suppressing recombination losses and improving the separation of photogenerated carriers [32–34]. Its layered morphology also supports better particle dispersion and provides additional surface contact for pollutant adsorption and interfacial reactions [35–37]. These features make BVO/CN heterojunctions attractive for antibiotic degradation, especially where efficient photocatalysis must be sustained under lower irradiation intensity rather than idealized high-energy conditions.

Various advanced treatment methods have been explored for antibiotic removal, including adsorption, membrane filtration, electrochemical oxidation, biodegradation, and advanced oxidation processes [38–40]. Among them, photocatalysis has attracted sustained attention because it enables the degradation of recalcitrant organic pollutants through in situ generation of reactive oxidative species (ROS) under relatively mild operating conditions [41–46]. In this context, low-intensity ultraviolet (UV) irradiation is of particular interest because it offers a more energy-conscious operating mode than high-power irradiation while still providing the photon input needed to activate suitable semiconductor photocatalysts. A photocatalyst that remains effective under low-intensity UV conditions would therefore be more attractive for practical water treatment, where energy demand and operating cost are key constraints [47].

Although Fe-doped BVO and BVO/CN heterojunctions have been widely explored, several limitations remain [48–50]. Many studies emphasize degradation efficiency under conventional irradiation conditions, but give less attention to how Fe incorporation, CN coupling, and mass ratio jointly regulate structure, optical response, morphology, photocatalytic activity, ROS involvement, and reusability. The behavior of Fe-doped BVO/CN photocatalysts under low-intensity UVC irradiation also remains insufficiently clarified. In addition, the component ratio is often treated as a simple optimization variable rather than as a structure-performance factor linked to crystallinity, interfacial contact, optical absorption, and degradation behavior. To address these gaps, this study develops 3% Fe-doped BVO/CN heterojunction photocatalysts with controlled Fe-doped BVO_4 :CN mass ratios and

evaluates their performance for TC degradation under low-intensity UVC irradiation. The originality of this work does not lie in Fe doping alone or BVO/CN heterojunction formation alone, since both strategies have been reported previously. Instead, the novelty lies in the integrated evaluation of composition-controlled Fe-doped BVO/CN photocatalysts by linking mass ratio with structural features, optical response, morphology, photocatalytic activity, kinetic behavior, reactive-species involvement, reusability, and preliminary synthesis cost. This structure-performance-operating condition analysis provides a clearer basis for assessing Fe-doped BVO/CN as a photocatalyst for antibiotic-contaminated water treatment.

2. Materials and Methods

2.1 Materials

Bismuth nitrate pentahydrate ($\text{Bi}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$), ammonium metavanadate (NH_4VO_3), ferric nitrate nonahydrate ($\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$), melamine, tetracycline hydrochloride (TC), nitric acid (HNO_3), and sodium hydroxide (NaOH) were used as chemical reagents. All reagents were of analytical grade and were used without further purification. Deionized water was used in all preparation and washing steps.

2.2 Synthesis of 3% Fe-doped BiVO_4

The 3% Fe-doped BiVO_4 was synthesized through a hydrothermal method, as previously reported [28]. Briefly, $\text{Bi}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$ was first dissolved in dilute HNO_3 under continuous stirring to form a clear precursor solution. In a separate beaker, NH_4VO_3 was dissolved in deionized water at elevated temperature to obtain a vanadium precursor solution. The two solutions were mixed under constant stirring to form the BiVO_4 precursor suspension. $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ was then added to the mixture in an amount corresponding to 3 wt% Fe relative to BiVO_4 . The suspension was stirred for 1 h for homogeneous dispersion, then sealed in a Teflon-lined stainless-steel autoclave and heated at 180 °C for 12 h. After natural cooling, the solid product was separated by centrifugation, washed repeatedly with deionized water and ethanol, dried overnight at 80 °C, and lightly ground to obtain the 3 wt% Fe-doped BiVO_4 photocatalyst, denoted as FeBVO.

2.3 Synthesis of CN

CN was prepared through thermal polymerization of melamine as reported previously [51,52]. Briefly, 10 g of melamine powder was placed in a covered alumina crucible and heated in a muffle furnace at 550 °C for 4 h at a heating rate of 5 °C/min. After natural cooling to room temperature, the yellow product was collected and ground into a fine powder. This sample was denoted as CN.

2.4. Synthesis of FeBVO/CN

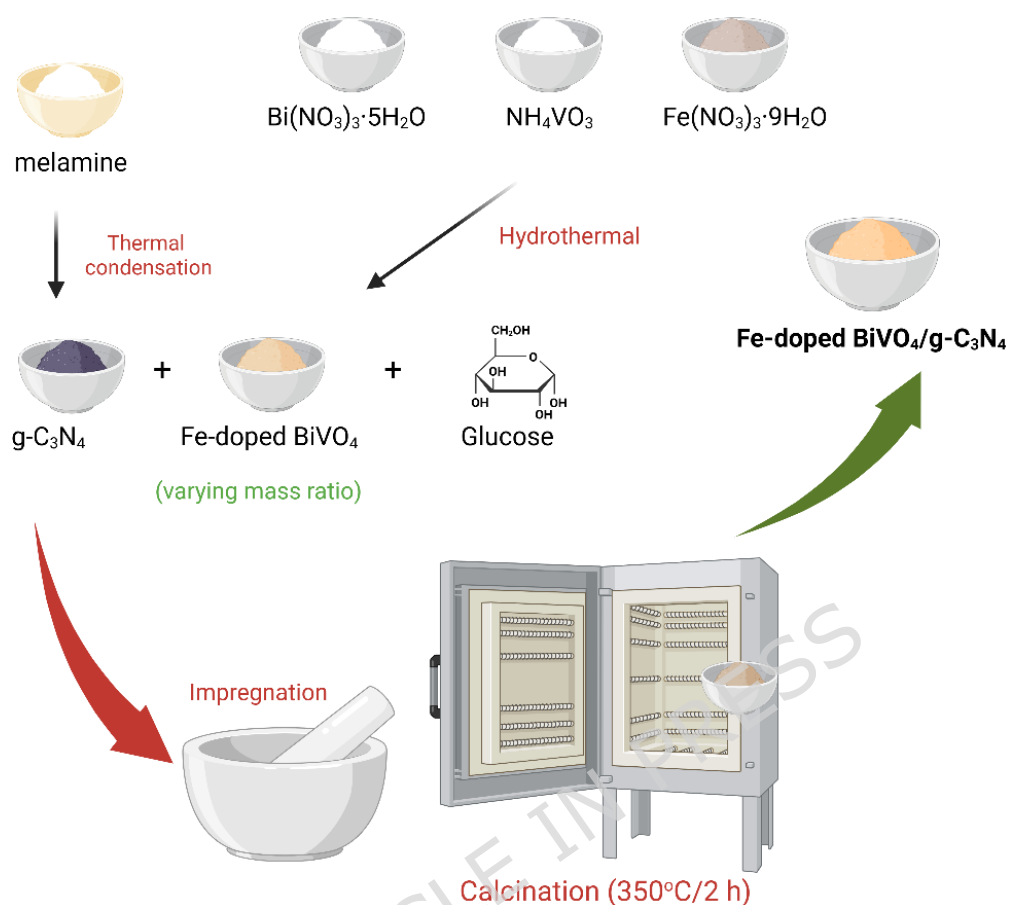


Fig. 1. Synthesis process of FeBVO/CN.

FeBVO/CN composites were prepared by combining FeBVO and CN at different mass ratios, as shown in Fig. 1. The FeBVO and CN powders were mixed according to FeBVO mass ratios of 1:1, 1:2, and 2:1. The resulting composite samples were denoted as FeBVO/CN-1:1, FeBVO/CN-1:2, and FeBVO/CN-2:1, respectively. Glucose (0.5 wt% of total FeBVO/CN mass) was added as a temporary organic binder and interfacial-contact promoter during composite preparation. During grinding, glucose helped improve physical contact between FeBVO particles and CN sheets. During calcination, partial decomposition of glucose may have assisted adhesion between the two phases and reduced physical separation of the components. At the low concentration used, its function was considered primarily contact-regulating rather than as a deliberate carbon precursor. Therefore, no contribution from glucose-derived carbon is assumed in the interpretation of the photocatalytic mechanism. Therefore, glucose is described here as a binder and morphology/contact-regulating additive rather than as a confirmed carbon source. The mixed powders were ground thoroughly in an agate mortar to obtain homogeneous mixtures. The mixtures were then transferred into crucibles and calcined at 350 °C for 2 h in a muffle

furnace to promote interfacial contact between FeBVO and CN. After cooling to room temperature, the composite powders were collected and stored for characterization and photocatalytic experiments.

2.5 Characterization

Fourier transform infrared (FTIR) spectroscopy was performed using an Agilent Cary 630 FTIR spectrometer to identify the functional groups and bonding characteristics of the prepared samples. X-ray diffraction (XRD) analysis was carried out using a Shimadzu XRD-6000 diffractometer equipped with Cu K α radiation ($\lambda = 1.54056 \text{ \AA}$) and operated at 40 kV and 30 mA to determine the crystalline structure and phase composition of the materials. Surface morphology and particle features were examined using field emission scanning electron (FESEM) microscopy on a JEOL JSM-6390 instrument operated at 5 kV, with magnifications ranging from 1000 $\times$ to 20,000 $\times$ to obtain clear visualization of surface topography and fine structural details while minimizing charging effects.

2.6 Photocatalytic degradation

The photocatalytic activity of the prepared catalysts was evaluated through degradation of TC in aqueous solution under low-intensity UVC irradiation [41,42]. Photocatalytic experiments were conducted in a batch system using 100 mL of TC solution in a 250 mL conical flask. Catalyst dosages of 5, 10, and 20 mg were used, corresponding to catalyst loadings of 50, 100, and 200 mg L⁻¹, respectively. The suspension was magnetically stirred at 400 rpm throughout the experiment to maintain uniform catalyst dispersion.

Before irradiation, the catalyst-TC suspension was stirred in the dark for 30 min to approach adsorption-desorption equilibrium. This dark equilibration period was selected because only a small change in C/C_0 was observed during the initial dark stage. However, an extended 120 min dark adsorption experiment was not performed in the present study; therefore, complete adsorption equilibrium cannot be conclusively confirmed.

After the dark equilibration period, the suspension was irradiated using a 13 W UVC sterilizer lamp operated at AC 220–240 V and 50 Hz. The lamp was positioned approximately 10 cm from the reaction suspension. The reaction was conducted at room temperature without active temperature control. The exact irradiance and photon flux at the reaction surface were not measured. At regular time intervals, approximately 5 mL of the reaction mixture was withdrawn and centrifuged to remove catalyst particles. The residual concentration of TC in the supernatant was determined using UV-visible spectrophotometry by monitoring the absorbance at 357 nm. The degradation efficiency was calculated from the change in TC concentration before and after irradiation. Control experiments were also performed under

identical conditions without photocatalyst to evaluate the extent of photolysis. The photocatalytic degradation efficiency was determined using the equation (Eq. 1) below;

$$\% \text{ Degradation efficiency (\%)} = \frac{C_0 - C_t}{C_0} \times 100\% \text{ (Eq. 1)}$$

where C_0 = TC initial concentration; C_t = TC concentration after irradiation at 't' minute.

The photocatalytic degradation kinetics were evaluated using the pseudo-first-order (PFO) model shown in Eq. (2):

$$-\ln\left(\frac{C}{C_0}\right) = kt \text{ (Eq. 2)}$$

where C_0 and C are the TC concentrations at irradiation time 0 and t , respectively, and k is the apparent PFO rate constant (min^{-1}). The rate constants were obtained from linear fitting of $-\ln(C/C_0)$ against irradiation time. The corresponding R^2 values were used to evaluate fitting reliability. All photocatalytic degradation experiments were performed in triplicate under identical conditions. The data are presented as mean $\pm$ standard deviation (SD). Error bars in the degradation profiles, optimization plots, scavenger experiments, and reusability tests represent the SD of three independent measurements ($n = 3$).

2.7 Economic assessment of catalyst synthesis

A preliminary economic assessment was conducted to estimate the laboratory-scale synthesis cost of the FeBVO/CN photocatalyst. The analysis considered the major reagents used in the optimized synthesis protocol, including $\text{Bi}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$, NH_4VO_3 , $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$, NaOH, melamine, ethanol, and deionized water. The quantities of each chemical were based on the amounts required to produce approximately 2.4 g of composite photocatalyst per batch.

Unit prices for the reagents were obtained from current supplier market prices and converted to Malaysian Ringgit (RM). The total material cost was calculated by multiplying the quantity of each reagent used per batch by its corresponding unit price. In addition to reagent costs, operational costs associated with the synthesis process were estimated by considering the electrical energy consumption of the hydrothermal reactor, calcination furnace, centrifugation step, and drying oven. Energy consumption was calculated using the rated power of each instrument and the operating duration during synthesis, while electricity costs were estimated based on the Sarawak Energy tariff rate.

Equipment depreciation costs were also included to provide a more realistic estimation of production expenses. These costs accounted for the proportional use of the Teflon-lined autoclave and centrifugation setup during each synthesis batch. The total synthesis cost was

then obtained by summing material costs, process energy costs, and equipment depreciation. Finally, the production cost per gram of photocatalyst was calculated by dividing the overall batch cost by the mass of composite obtained. This economic analysis should be interpreted as a preliminary laboratory-scale estimate rather than a complete techno-economic assessment. It does not fully account for scale-up factors such as bulk chemical pricing, continuous production, reactor design, labor, catalyst recovery, washing and regeneration, pH adjustment, post-treatment neutralization, waste handling, and real wastewater performance. A more complete techno-economic evaluation is required before large-scale wastewater treatment feasibility can be claimed.

3.0 Result and Discussion

3.1 Characterizations

3.1.1 FTIR analysis

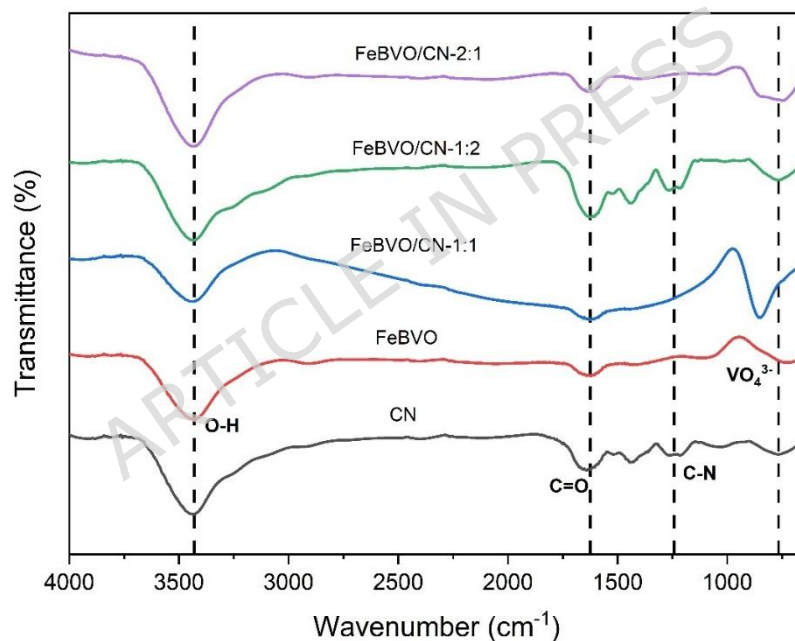


Fig. 2. FTIR spectra of CN, FeBVO, and FeBVO/CN composites with different FeBVO:CN mass ratios.

As shown in Fig.2, the FTIR spectra of CN, FeBVO, and the FeBVO/CN composite samples were recorded to characterize the surface functional groups, molecular bonding, and interfacial interactions. The CN spectrum displays characteristic absorption bands in the 1200–1650 cm^{-1} region, including peaks at approximately 1230, 1315, 1400, 1460, and 1630 cm^{-1} , which are attributed to stretching vibrations of C–N and C=N bonds in the tri-s-triazine rings and heptazine motifs of the polymeric structure [53]. A sharp peak at 805 cm^{-1} corresponds to the out of plane bending mode of the triazine ring, confirming the CN

framework. The broad band above 3000 cm^{-1} is assigned to N–H and O–H stretching vibrations from residual amino groups and adsorbed water molecules, respectively, which are typical of calcined CN and can serve as active sites for photocatalytic reactions [54,55].

For FeBVO, the broad band around 3430 cm^{-1} and the band near 1630 cm^{-1} are assigned to O–H stretching and H–O–H bending vibrations, respectively [56]. The bands between 700 and 900 cm^{-1} are associated with Bi–O and V–O vibrations in monoclinic BVO. In particular, the band around 830 cm^{-1} is attributed to V–O stretching in VO_4 tetrahedra, supporting the formation of the BVO framework [28]. No distinct Fe–O vibration is resolved in the 550 – 600 cm^{-1} region. This may be due to the low Fe content and possible overlap with the strong BVO lattice vibrations which is consistent with the low Fe content and spectral overlap with strong BVO lattice vibrations [57].

For the FeBVO/CN composites, the characteristic bands of both FeBVO and CN are retained, indicating successful integration of the two components. The O–H stretching (3430 cm^{-1}) and bending (1630 cm^{-1}) bands are consistently observed across all hybrids, often with slightly enhanced intensity compared with individual components, indicating that the composites maintain or even increase surface hydroxylation, which facilitates $\bullet\text{OH}$ generation during photocatalysis [58]. The C–N/C=N stretching bands and triazine ring vibration from CN remain visible, while the Bi–O and V–O vibrations from FeBVO become more pronounced as the FeBVO fraction increases. Minor intensity changes and peak shifts suggest interfacial interaction between FeBVO particles and CN sheets. The absence of new major bands or disappearance of characteristic peaks indicates that the composite formation preserved the main chemical features of both components without detectable phase decomposition. These results support the formation of FeBVO/CN composites, although more direct techniques such as XPS or elemental mapping would be required to confirm interfacial bonding and Fe distribution.

3.1.2 XRD analysis and microstructural parameters

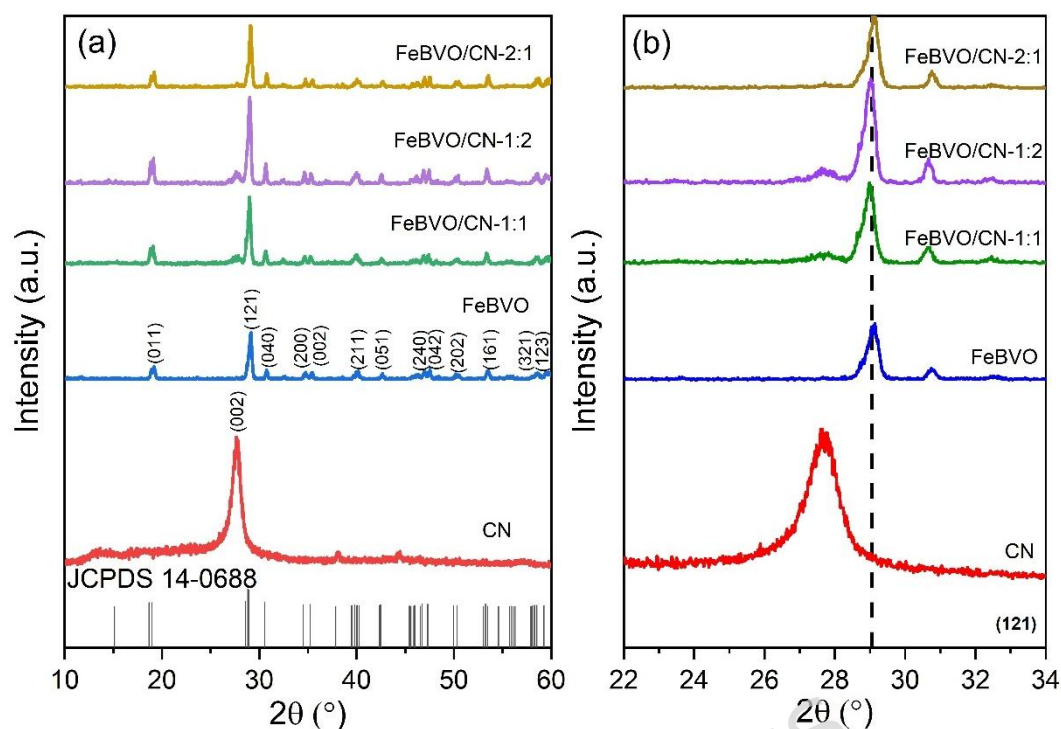


Fig. 3. XRD patterns of CN, FeBVO, and FeBVO/CN composites: (a) full XRD patterns; (b) enlarged view of the main BVO reflection.

Fig. 3(a) shows the XRD patterns of CN, FeBVO, and the FeBVO/CN composites, while Fig. 3(b) presents the enlarged region around the main BVO reflection. CN shows a broad diffraction peak at $2\theta \approx 27.4^\circ$, indexed to the (002) plane of graphitic carbon nitride, which corresponds to interlayer stacking of conjugated heptazine units [59,60]. FeBVO shows diffraction peaks at $2\theta \approx 18.7^\circ, 28.9^\circ, 30.6^\circ, 34.6^\circ, 35.2^\circ, 39.9^\circ, 42.4^\circ, 46.8^\circ, 50.2^\circ, 53.3^\circ, 55.9^\circ$, and 58.5° , corresponding to monoclinic scheelite BVO (JCPDS 14-0688) [28]. No secondary reflections attributable to iron oxide or other impurity phases are detected.

No distinct Fe-related diffraction peaks are observed in the FeBVO sample or the FeBVO/CN composites. This result is expected because the Fe loading is low, and Fe species incorporated into the BVO lattice or highly dispersed in the matrix may remain below the detection limit of XRD. No separate Fe_2O_3 , Fe_3O_4 , or other crystalline iron oxide reflections were detected [61,62]. At the low Fe loading used, the absence of distinct Fe-related peaks is consistent with Fe species being present at concentrations below the XRD detection threshold or being highly dispersed within the FeBVO matrix. Importantly, the monoclinic BVO diffraction pattern was retained, indicating that Fe addition did not produce a detectable secondary crystalline phase.

After coupling with CN, the FeBVO/CN composites retain the characteristic reflections of monoclinic BVO, indicating that composite formation does not destroy the FeBVO crystalline framework. The CN-related (002) feature near 27.4° becomes less distinct in the composites because it overlaps with the intense BVO reflection and decreases in relative intensity as the FeBVO fraction increases [63,64]. The coexistence of BVO and CN diffraction features, together with the absence of additional impurity peaks, supports the successful formation of FeBVO/CN heterostructures [65].

The microstructural parameters of the samples were estimated from the (121) reflection at $2\theta \approx 28.9^\circ$ using the Debye-Scherrer as shown in Eq. 3:

$$D = \frac{K\lambda}{\beta \cos \theta} \text{ (Eq. 3)}$$

where k is the shape factor (0.98), λ is the X-ray wavelength, β is the full width at half maximum (FWHM) of the (121) peak (in radians), and θ is the Bragg angle. Lattice strain (ε) and dislocation density (δ) were also evaluated to gain insight into microstructural distortions and defect densities using Eq. 4 and Eq. 5:

$$\varepsilon = \frac{\beta}{4 \tan \theta} \text{ (Eq. 4)}$$

$$\delta = \frac{1}{D^2} \text{ (Eq. 5)}$$

where β is the same FWHM parameter used in Scherrer analysis.

The crystallite size, lattice strain, and dislocation density were estimated from the BVO (121) reflection at $2\theta \approx 28.9^\circ$. The calculated values are summarized in Table 1. The crystallite size increased from 22.3 nm for FeBVO to 28.4 nm for FeBVO/CN-2:1. The FeBVO/CN-1:1 composite showed the highest lattice strain and dislocation density, suggesting greater structural distortion. In contrast, FeBVO/CN-2:1 showed lower strain and lower dislocation density than FeBVO/CN-1:1 and FeBVO/CN-1:2, indicating a more balanced microstructural state. This suggests that the 2:1 composition preserves the monoclinic BVO framework while limiting excessive defect formation.

The XRD-derived microstructural parameters indicate that photocatalytic performance should not be interpreted from crystallite size alone. Although smaller crystallite size may increase surface exposure, excessive strain and defect density may also introduce recombination centers. The higher activity of FeBVO/CN-2:1 is therefore more plausibly associated with preserved phase purity, suitable crystallinity, moderate interfacial strain, and effective heterojunction formation rather than crystallite-size reduction alone.

Table 1. Crystallite size (D), lattice strain (ϵ), and dislocation density (δ) calculated from the (121) diffraction peak of monoclinic BVO.

Photocatalyst	D (nm)	ϵ	δ
FeBVO	22.3	0.0056	0.0027
FeBVO/CN-1:1	22.4	0.0062	0.0040
FeBVO/CN-1:2	26.4	0.0057	0.0039
FeBVO/CN-2:1	28.4	0.0047	0.0020

3.1.3 Optical properties

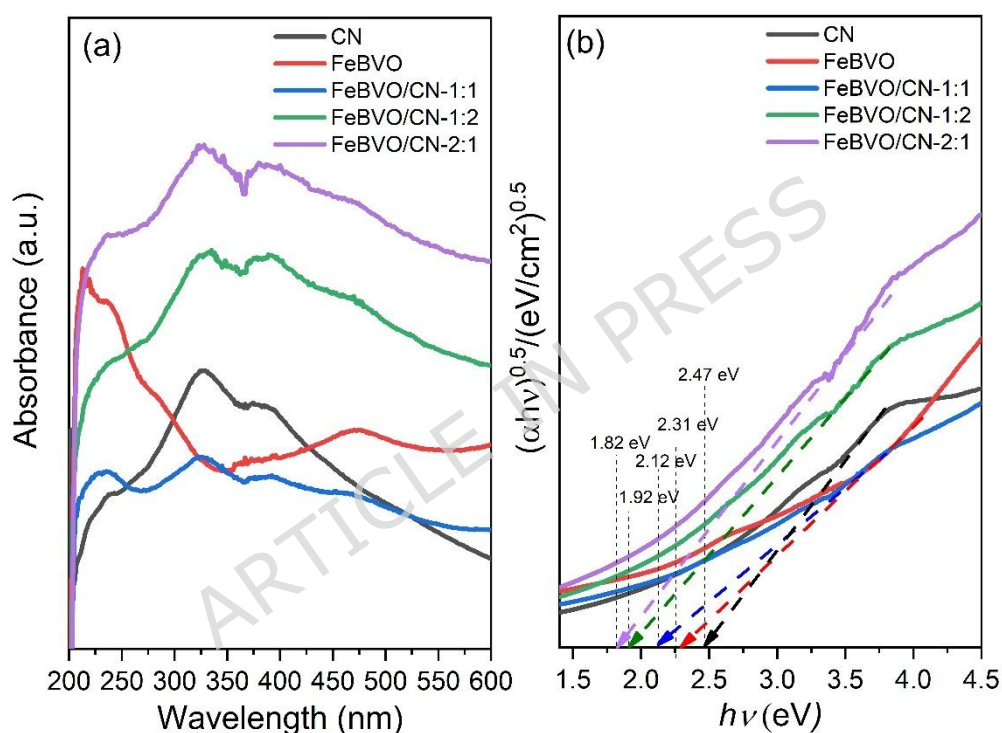


Fig. 4 Optical properties of CN, FeBVO, and FeBVO/CN composites: (a) UV-Vis absorption spectra; (b) corresponding Tauc plots.

Fig. 4(a) shows the UV-Vis absorption spectra of CN, FeBVO, and the FeBVO/CN composites, while Fig. 4(b) presents the corresponding Tauc plots. CN shows strong absorption in the ultraviolet region with an absorption edge extending into the visible range, which is consistent with the optical response of graphitic carbon nitride [66], while FeBVO showed broader absorption over the near-visible region, consistent with the intrinsic light-harvesting behavior of monoclinic BVO [67]. After coupling FeBVO with CN, the composite samples show broader optical absorption, especially in the 300–500 nm region. This indicates that heterojunction formation modifies the photoresponse of the system. Among the

composites, FeBVO/CN-2:1 and FeBVO/CN-1:2 show stronger absorption intensity than FeBVO/CN-1:1, suggesting that optical behavior is influenced by the FeBVO mass ratio rather than by simple physical mixing alone. This improved absorption is beneficial for photocatalysis because it increases the probability of photogenerated charge-carrier formation under UVC irradiation.

The apparent optical transition energies were estimated from Tauc plots. The band gap values were 2.47 eV for CN, 2.31 eV for FeBVO, 2.12 eV for FeBVO/CN-1:1, 1.92 eV for FeBVO/CN-1:2, and 1.82 eV for FeBVO/CN-2:1. The lower apparent transition energy of FeBVO/CN-2:1 suggests enhanced photoresponse after heterojunction formation. However, for composite photocatalysts, Tauc-derived values should be interpreted as apparent transition energies of the coupled system rather than as the intrinsic band gap of a single semiconductor phase [68]. Therefore, the higher photocatalytic activity of FeBVO/CN-2:1 should not be attributed to band gap narrowing alone. It is more reasonably associated with the combined effects of broader optical absorption, preserved BVO crystallinity, favorable FeBVO composition, and interfacial contact between the two semiconductor phases.

3.1.4 FESEM morphology and particle-size comparison

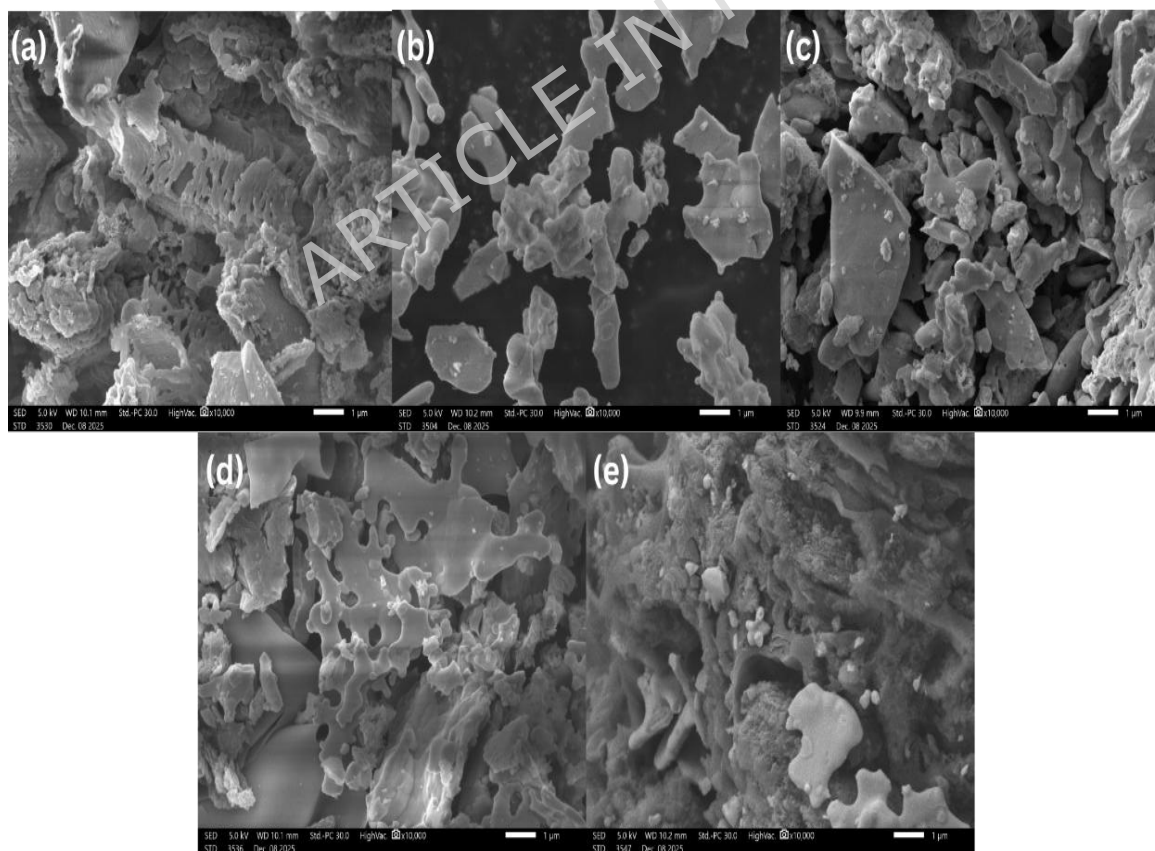


Fig. 5. FESEM micrographs of (a) CN, (b) FeBVO, (c) FeBVO/CN-1:1, (d) FeBVO/CN-1:2, and (e) FeBVO/CN-2:1.

Fig. 5 shows the FESEM micrographs of CN, FeBVO, and the FeBVO/CN composites. CN displays a layered and irregular sheet-like morphology (Fig. 5(a)), which is typical of graphitic carbon nitride prepared by thermal polymerization of melamine [69]. These wrinkled sheets provide a structural platform for anchoring oxide particles and improving interfacial contact during composite formation [70,71].

FeBVO shows irregular block-like particles (Fig. 5(b)) with visible agglomeration, indicating the formation of discrete oxide particles [72]. After coupling with CN, the FeBVO/CN composites show clear morphological integration between the oxide particles and CN sheets. In FeBVO/CN-1:1 (Fig. 5(c)), FeBVO particles are distributed on and between the CN sheets, although partial aggregation remains visible. In FeBVO/CN-1:2 (Fig. 5(d)), the larger CN fraction provides broader sheet coverage, and the oxide particles appear more dispersed on the CN matrix. In FeBVO/CN-2:1 (Fig. 5(e)), the oxide phase is more dominant, but interfacial contact with CN remains evident. This morphology suggests that CN acts as a supporting scaffold that helps reduce excessive FeBVO agglomeration and increases contact between the two semiconductor phases. The FeBVO/CN-2:1 sample appears to provide a favorable balance between sufficient oxide loading and heterojunction contact. This balance is important because excessive CN may dilute the active oxide phase, while excessive oxide loading may reduce interfacial exposure.

The FESEM-estimated particle sizes were larger than the XRD-derived crystallite sizes, as summarized in Table 2. This difference indicates that the visible particles observed by FESEM consist of aggregated crystallites rather than single crystalline domains. FeBVO showed irregular particles of approximately 250–450 nm, with an estimated mean size of about 330 nm. FeBVO/CN-1:1 and FeBVO/CN-1:2 showed smaller visible oxide particles of approximately 180–350 nm and 150–300 nm, respectively, suggesting partial dispersion on the CN matrix. In contrast, FeBVO/CN-2:1 showed larger oxide-rich agglomerates of approximately 250–600 nm, with an estimated mean size of about 400 nm. Therefore, XRD crystallite size and FESEM particle size should be interpreted as complementary structural parameters. XRD estimates coherent crystalline domains, while FESEM shows external particle morphology and agglomeration. The improved activity of FeBVO/CN-2:1 is therefore not explained by smaller particle size alone, but by the combined influence of oxide loading, interfacial contact, optical response, and preserved crystallinity.

Table 2. Comparison between XRD-derived crystallite size and FESEM-estimated particle size of CN, FeBVO, and FeBVO/CN composites.

Sample	XRD	Approximate FESEM particle size
CN	Not applicable	sheet/agglomerate size, ~0.8–2.5 μm

FeBVO	22.3 nm	~250–450 nm, mean ~330 nm
FeBVO/CN-1:1	22.4 nm	~180–350 nm, mean ~260 nm
FeBVO/CN-1:2	26.4 nm	~150–300 nm, mean ~220 nm
FeBVO/CN-2:1	28.4 nm	~250–600 nm, mean ~400 nm

3.2 Photocatalytic degradation performance

3.2.1 Catalyst screening and kinetic analysis

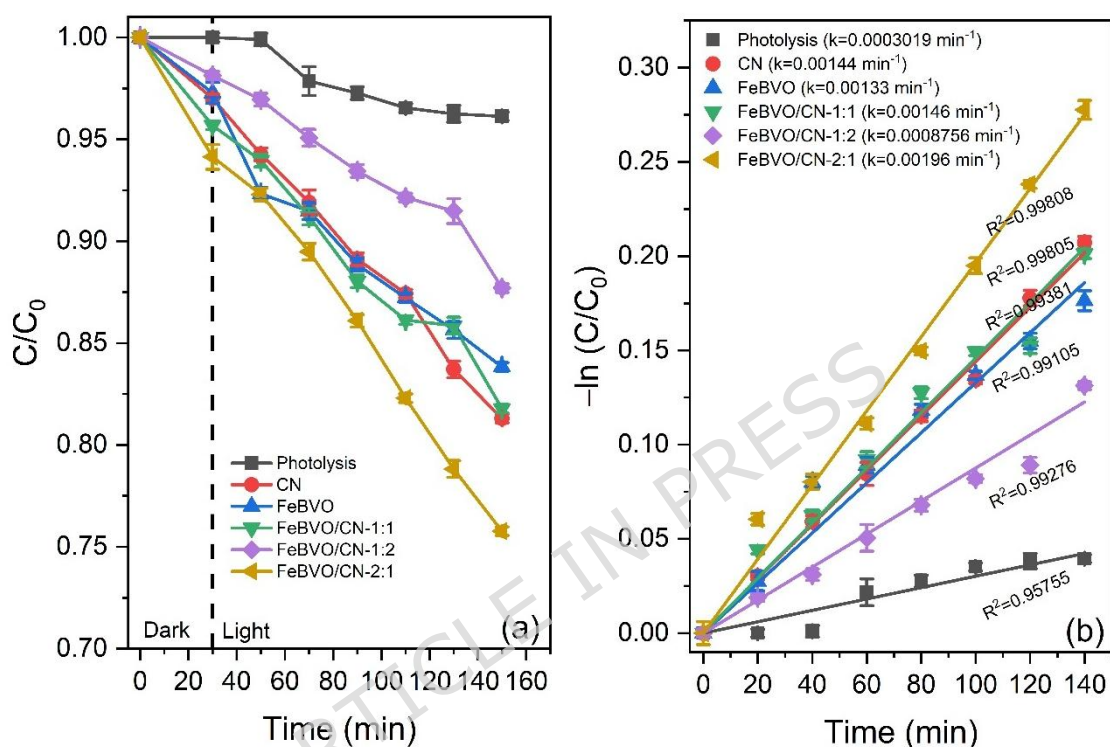


Fig. 6. Photocatalytic screening of CN, FeBVO, and FeBVO/CN composites under low-intensity UVC irradiation: (a) TC degradation profiles including the dark equilibration stage; (b) corresponding PFO kinetic plots of $-\ln(C/C_0)$ versus irradiation time. Experimental conditions: initial TC concentration = 10 mg L^{-1} , reaction volume = 100 mL, catalyst loading = 100 mg L^{-1} , pH = 4, irradiation time = 120 min. Data are presented as mean $\pm$ SD ($n = 3$).

The photocatalytic degradation of TC was first evaluated to compare CN, FeBVO, and the FeBVO/CN composite samples under low-intensity UVC irradiation. As shown in Fig. 6(a), only a small decrease in C/C_0 was observed during the initial dark equilibration stage, indicating limited TC adsorption under the tested conditions. The photolysis control conducted without catalyst showed negligible TC removal, confirming that direct UVC photolysis contributed minimally to TC degradation. Therefore, the greater decrease in TC concentration observed in the catalyst-containing systems after irradiation was mainly associated with photocatalytic activity [28].

Among the prepared materials, FeBVO/CN-2:1 showed the highest photocatalytic activity, reaching the lowest C/C_0 value after 120 min of irradiation. CN, FeBVO, and FeBVO/CN-1:1 showed intermediate degradation behavior, while FeBVO/CN-1:2 showed the weakest activity. This trend indicates that photocatalytic performance does not increase simply with either FeBVO or CN content alone. Instead, the activity depends on the FeBVO composition, which influences light absorption, interfacial contact, active-site accessibility, and charge utilization within the heterojunction.

The superior activity of FeBVO/CN-2:1 is plausibly related to its more favorable composition. At this ratio, FeBVO remains sufficiently abundant to provide the main photoactive oxide phase, while CN provides interfacial contact and a supporting matrix for charge migration. In contrast, the lower activity of FeBVO/CN-1:2 suggests that excessive CN may dilute the active oxide phase and reduce the number of effective photocatalytic sites, despite improving sheet-like dispersion. Therefore, the optimized activity of FeBVO/CN-2:1 is attributed to a balance between oxide loading and heterojunction contact rather than to simple compositional averaging.

The degradation data were fitted using the PFO kinetic model, as shown in Fig. 6(b). The apparent rate constants followed the order FeBVO/CN-2:1 ($k = 0.00196 \text{ min}^{-1}$) > CN ($k = 0.00144 \text{ min}^{-1}$) $\approx$ FeBVO/CN-1:1 ($k = 0.00144 \text{ min}^{-1}$) > FeBVO ($k = 0.00133 \text{ min}^{-1}$) > FeBVO/CN-1:2 ($k = 0.0008756 \text{ min}^{-1}$). This trend agrees with the degradation profiles and confirms that FeBVO/CN-2:1 provides the most favorable reaction environment for TC degradation under the tested conditions. The PFO fitting gave R^2 values of 0.99805, 0.99105, 0.99381, 0.99276, 0.99808 for CN, FeBVO, FeBVO/CN-1:1, FeBVO/CN-1:2, and FeBVO/CN-2:1, respectively. These values indicate that the PFO model reasonably describes TC degradation within the studied irradiation period. The kinetic enhancement observed for FeBVO/CN-2:1 should not be attributed to one factor alone. It is more likely associated with the combined effects of preserved BVO crystallinity, broader optical absorption, suitable microstructural disorder, and effective contact between FeBVO and CN.

The 30 min dark stirring period was used to approach adsorption-desorption equilibrium before irradiation. During this stage, only a small decrease in C/C_0 was observed, suggesting limited TC adsorption under the tested conditions. However, because a separate 120 min dark adsorption experiment was not performed, the possible contribution of slow adsorption beyond the initial equilibration period cannot be completely excluded. Therefore, TC removal under irradiation is interpreted mainly as photocatalytic degradation, while the possible contribution of continued adsorption is acknowledged as a limitation. Since

FeBVO/CN-2:1 showed the highest activity, it was selected for subsequent optimization studies.

3.2.2 Effect of catalyst loading

The effect of catalyst loading on TC degradation was investigated using the optimized FeBVO/CN-2:1 composite. As shown in Fig. S1(a), TC degradation increased as catalyst loading increased from 50 to 200 mg L⁻¹. The system containing 200 mg L⁻¹ catalyst showed the fastest decline in TC concentration, while the 50 mg L⁻¹ system showed the slowest degradation. This improvement can be explained by the greater number of available active sites and the increased amount of photoactive material in the reaction suspension [73–75]. Higher catalyst loading can enhance photon absorption and promote the formation of reactive species involved in TC degradation. The PFO kinetic plots in Fig. S1(b) further support this trend. The apparent rate constant increased from 0.00178 min⁻¹ at 50 mg L⁻¹ to 0.00197 min⁻¹ at 100 mg L⁻¹ and 0.00240 min⁻¹ at 200 mg L⁻¹. The corresponding R² values were 0.99505, 0.99659, and 0.98923, respectively, indicating acceptable fitting to the PFO model. Within the studied range, 200 mg L⁻¹ provided the most favorable catalyst loading. However, excessive catalyst loading may reduce photocatalytic efficiency in suspension systems because dense catalyst particles can scatter light, shield active surfaces, and limit photon penetration. Therefore, the selected loading of 200 mg L⁻¹ represents the best condition within the tested range rather than a universal optimum.

3.2.3 Effect of solution pH

The effect of initial solution pH was evaluated because pH influences TC speciation, catalyst surface charge, adsorption behavior, and ROS formation. As shown in Fig. S2(a), TC degradation was lowest at pH 4, increased at pH 7, and reached the highest level at pH 10. The kinetic analysis in Fig. S2(b) supports this trend, with the apparent rate constant increasing from 0.00241 min⁻¹ at pH 4 to 0.00484 min⁻¹ at pH 7 and 0.01055 min⁻¹ at pH 10. The corresponding R² values were 0.99360, 0.98845, and 0.96078, respectively. These values indicate acceptable PFO fitting, although the pH 10 system shows a more rapid initial degradation stage followed by slower removal at longer irradiation time.

The enhanced degradation at pH 10 may be explained by the combined effects of TC speciation, catalyst surface behavior, and ROS formation. TC is an amphoteric compound with reported pK_a values of approximately 3.3, 7.7, and 9.7. At pH 4, TC mainly exists in cationic or zwitterionic forms, while near neutral pH it exists mainly as zwitterionic and partially deprotonated species [76]. At pH 10, TC becomes predominantly anionic due to more extensive deprotonation. These changes affect electrostatic interaction with the catalyst surface and the accessibility of TC molecules to reactive sites.

Although zeta potential and pH_{pzc} were not measured in this study, the surface charge of FeBVO/CN-2:1 is expected to vary with pH. At alkaline pH, increased OH^- concentration may modify the interfacial double layer and facilitate ROS formation, especially $\bullet\text{OH}$ generation through reactions involving photogenerated holes. The scavenger results also indicate that $\bullet\text{O}_2^-$ -related pathways remain the dominant contributor to TC degradation. Therefore, the higher activity at pH 10 should be attributed to the combined influence of TC deprotonation, surface-charge changes, oxygen activation, and hydroxyl-radical formation rather than pH alone. Although pH 10 produced the highest degradation efficiency, its practical application should be viewed cautiously. Real wastewater treatment would require alkaline adjustment and possible post-treatment neutralization, which would increase chemical use and operating cost. Therefore, pH 10 is identified as the most favorable laboratory condition in this study, while near-neutral performance remains important for practical wastewater applications.

3.2.4 Effect of initial TC concentration

The effect of initial TC concentration on photocatalytic degradation was evaluated using FeBVO/CN-2:1 under the optimized catalyst loading and pH conditions. As shown in Fig. S3(a), TC degradation decreased as the initial TC concentration increased from 5 to 20 mg L^{-1} . The 5 mg L^{-1} solution showed the fastest decrease in C/C_0 and reached the lowest residual TC concentration after 120 min of irradiation. In contrast, the 10 and 20 mg L^{-1} systems showed slower degradation and similar final C/C_0 values.

The apparent PFO kinetic analysis in Fig. S3(b) further supports this concentration-dependent trend. The apparent rate constant was highest at 5 mg L^{-1} , with $k = 0.01544 \text{ min}^{-1}$. The rate constants decreased to 0.01000 min^{-1} at 10 mg L^{-1} and 0.01040 min^{-1} at 20 mg L^{-1} . The corresponding R^2 values were 0.93188, 0.95524, and 0.96766, respectively. These values suggest that the PFO model provides an acceptable apparent description of TC degradation, although the 5 mg L^{-1} system shows faster early-stage degradation followed by slower removal at longer irradiation time.

The lower degradation rate at higher TC concentration may be explained by several factors. First, more TC molecules compete for a limited number of active sites on the catalyst surface, which restricts adsorption and interfacial oxidation [77–79]. Second, a more concentrated TC solution may absorb or screen part of the incident UVC light, reducing photon penetration to the catalyst surface and lowering the formation of photogenerated electron-hole pairs [65]. Third, higher pollutant loading may generate more intermediates during irradiation, which can occupy active sites and hinder further TC transformation. These factors collectively reduce the apparent degradation rate at elevated TC concentration. The

improved performance at 5 mg L^{-1} therefore reflects a more favorable balance between pollutant availability, active-site accessibility, and photon utilization. Based on these results, 5 mg L^{-1} was selected as the optimal initial TC concentration for subsequent scavenger and reusability experiments.

3.2.5 Reactive-species scavenger experiments

Reactive-species scavenger experiments were conducted to infer the possible reactive pathways involved in TC degradation over FeBVO/CN-2:1. Methanol (MeOH), ethylenediaminetetraacetic acid (EDTA), and ascorbic acid (AA) were used as scavengers for $\bullet\text{OH}$, h^+ , and $\bullet\text{O}_2^-$ -related pathways, respectively [41,42]. As shown in Fig. S4(a), the addition of scavengers reduced TC degradation to different extents compared with the system without scavenger, indicating that multiple reactive pathways contributed to TC transformation.

The strongest inhibition was observed in the presence of AA, suggesting that electron-mediated oxygen reduction and $\bullet\text{O}_2^-$ -related pathways exerted the strongest influence on TC degradation. The addition of EDTA also suppressed TC degradation, indicating that h^+ contributed to the oxidation process. Similarly, the decrease in activity after adding MeOH suggests the participation of $\bullet\text{OH}$ -related reactions. Therefore, TC degradation over FeBVO/CN-2:1 proceeded through the combined action of $\bullet\text{O}_2^-$ -related pathways, h^+ , and $\bullet\text{OH}$.

The apparent PFO kinetic analysis in Fig. S4(b) supports this interpretation. The apparent rate constant decreased from 0.01528 min^{-1} in the absence of scavenger to 0.00713 min^{-1} with MeOH, 0.00692 min^{-1} with EDTA, and 0.00628 min^{-1} with AA. The corresponding R^2 values were 0.93474, 0.90248, 0.96683, and 0.94665, respectively. These values indicate acceptable apparent PFO fitting under the tested conditions. The largest reduction in the rate constant after AA addition suggests that $\bullet\text{O}_2^-$ -related pathways played the most influential role, while h^+ and $\bullet\text{OH}$ also contributed meaningfully to TC degradation. Scavenger inhibition provides comparative evidence for the relative influence of reactive pathways rather than direct quantification of individual ROS. Within this basis, the greater decrease in k after AA addition supports a dominant electron-mediated O_2 reduction pathway, while the responses to EDTA and MeOH indicate additional contributions from h^+ - and $\bullet\text{OH}$ -related oxidation.

3.2.6 Reusability and post reaction stability

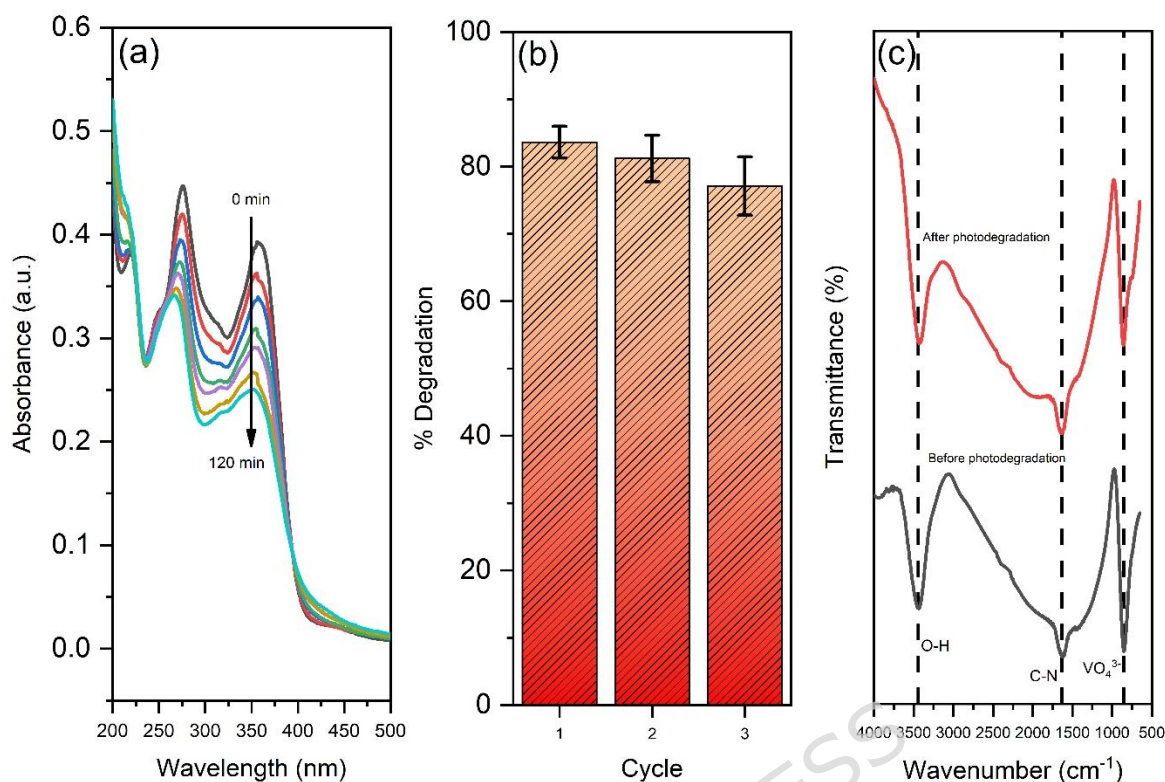


Fig. 7. (a) Temporal UV-Vis absorption spectra of TC during photocatalytic degradation over FeBVO/CN-2:1 under low-intensity UVC irradiation. (b) TC degradation efficiency over three consecutive photocatalytic cycles; (c) FTIR spectra of FeBVO/CN-2:1 before and after photocatalytic degradation. Experimental conditions: initial TC concentration = 5 mg L⁻¹, reaction volume = 100 mL, catalyst loading = 200 mg L⁻¹, pH = 10, irradiation time = 120 min. Degradation data are presented as mean $\pm$ SD from three independent experiments ($n = 3$).

The temporal evolution of the UV-Vis absorption spectra of TC during photocatalytic treatment with FeBVO/CN-2:1 was examined under the optimized reaction conditions. As shown in Fig. 7(a), the untreated TC solution exhibited a characteristic absorption band at approximately 357 nm. The intensity of this band progressively decreased with increasing UVC irradiation time from 0 to 120 min, indicating gradual transformation of the TC chromophoric structure. The continuous reduction in absorbance at 357 nm agrees with the concentration-dependent degradation profile and kinetic results obtained for FeBVO/CN-2:1. No recovery of the principal TC absorption band was observed during irradiation, supporting the progressive removal of the parent TC chromophore from the reaction system. The spectral changes therefore provide additional evidence for photocatalytic transformation of TC under low-intensity UVC irradiation.

The reusability of FeBVO/CN-2:1 was evaluated over three consecutive photocatalytic cycles under the optimized reaction conditions. As shown in Fig. 7(b), the catalyst

maintained most of its TC degradation ability after repeated use. The degradation efficiency decreased from 83.6% in the first cycle to 81.2% in the second cycle and 77.1% in the third cycle. This moderate decline indicates preliminary reusability of FeBVO/CN-2:1 under the tested conditions. The decrease in degradation efficiency may be associated with several factors. First, reaction intermediates formed during TC degradation may adsorb on the catalyst surface and partially block active sites [80,81]. Second, incomplete catalyst recovery during centrifugation, washing, and reuse may reduce the amount of active material in later cycles [82]. Third, minor surface changes during irradiation may affect the accessibility of reactive sites. Since catalyst mass recovery and metal leaching analysis were not performed, the exact contribution of catalyst loss or leaching cannot be confirmed.

Post-reaction FTIR analysis was conducted to examine changes in the surface functional groups of FeBVO/CN-2:1 after photocatalysis. As shown in Fig. 7(c), the main bands associated with O–H, C–N, and VO_4^{3-} vibrations were retained after TC degradation. This suggests that the major surface functional features of the composite were preserved after photocatalytic use. Minor intensity changes may be related to adsorbed intermediates, surface hydroxyl variation, or residual species remaining after reaction. However, FTIR provides only surface-level evidence and cannot confirm crystallographic stability, phase preservation, metal leaching behavior, or long-term photocorrosion resistance. Therefore, the stability result should be interpreted as preliminary.

3.3 Proposed degradation mechanism of TC

Based on the optical properties, Mulliken-estimated band-edge positions, photocatalytic performance, and scavenger response, a plausible charge-transfer and TC degradation mechanism over FeBVO/CN-2:1 is proposed in Fig. 8. The enhanced photocatalytic activity of FeBVO/CN-2:1 is attributed to the combined effects of preserved monoclinic BiVO_4 structure, broader optical response, interfacial contact between FeBVO and CN, and preservation of charge carriers with sufficient redox ability for reactive-species generation. To further assess the energetically feasible charge-transfer pathways in the FeBVO/CN heterojunction [83], the conduction-band (CB) and valence-band (VB) edge potentials of CN and FeBVO were estimated using the Mulliken electronegativity method according to Eq. (6) and (7):

$$E_{VB} = \chi - E^0 + 0.5E_g \quad (\text{Eq. 6})$$

$$E_{CB} = E_{VB} - E_g \quad (\text{Eq. 7})$$

where E_{CB} and E_{VB} are the CB and VB edge potentials, respectively, χ is the absolute electronegativity of the semiconductor, E^0 is the energy of free electrons on the hydrogen

scale, approximately 4.5 eV, and E_g is the experimentally determined optical transition energy. Based on the experimentally determined optical transition energies of 2.47 eV for CN and 2.31 eV for FeBVO, the estimated CB and VB potentials were -1.02 and $+1.46$ V for CN and $+0.39$ and $+2.70$ V for FeBVO, respectively, versus NHE. These estimated band positions provide a thermodynamic basis for comparing conventional Type-II and Z-scheme-like charge-transfer pathways.

Under a conventional Type-II configuration, photogenerated electrons would migrate from the CN CB to the FeBVO CB, while holes would migrate from the FeBVO VB to the CN VB. Consequently, the accumulated electrons would reside on the FeBVO CB at $+0.39$ V, and the holes would reside on the CN VB at $+1.46$ V. Since the FeBVO CB is more positive than the $O_2/\bullet O_2^-$ redox potential of approximately -0.33 V versus NHE, these electrons would have insufficient reduction potential for thermodynamically favorable O_2 reduction to $\bullet O_2^-$. Similarly, the oxidation ability of holes retained on the CN VB would be weakened. Therefore, a conventional Type-II pathway is less consistent with the strong influence of $\bullet O_2^-$ -related reactions observed experimentally.

In contrast, the estimated band alignment is more consistent with a Z-scheme-like charge-transfer pathway. Under UVC irradiation, both CN and FeBVO are photoexcited, generating electrons in their respective CBs and holes in their VBs. At the heterointerface, electrons associated with the FeBVO CB are proposed to recombine with holes associated with the CN VB. This interfacial recombination pathway retains strongly reducing electrons on the CN CB at -1.02 V and strongly oxidizing holes on the FeBVO VB at $+2.70$ V. The CN CB potential is sufficiently negative relative to the $O_2/\bullet O_2^-$ potential of -0.33 V to support reduction of dissolved O_2 and formation of $\bullet O_2^-$ -related reactive species [84]. Meanwhile, the FeBVO VB potential is more positive than the $OH^-/\bullet OH$ oxidation potential of approximately $+1.99$ V versus NHE, supporting $\bullet OH$ generation and strong oxidative reactions involving photogenerated holes [85].

The scavenger experiments are consistent with this proposed redox configuration. AA caused the greatest reduction in the apparent degradation rate constant, indicating that electron-mediated O_2 reduction and $\bullet O_2^-$ -related pathways exerted the strongest influence on TC degradation. The inhibitory effects of EDTA and MeOH further indicate contributions from h^+ - and $\bullet OH$ -related oxidation, respectively. Thus, TC degradation over FeBVO/CN-2:1 is proposed to proceed through the combined action of $\bullet O_2^-$ -related pathways, h^+ , and $\bullet OH$, with the $\bullet O_2^-$ -related pathway showing the greatest contribution under the tested conditions. Further Mott-Schottky analysis, photoluminescence spectroscopy, electrochemical impedance spectroscopy, photocurrent response, XPS, and ESR/EPR spin-trapping analysis

are required to confirm the band alignment, charge separation behavior, interfacial charge-transfer route, oxidation states, and ROS formation more directly [86–89].

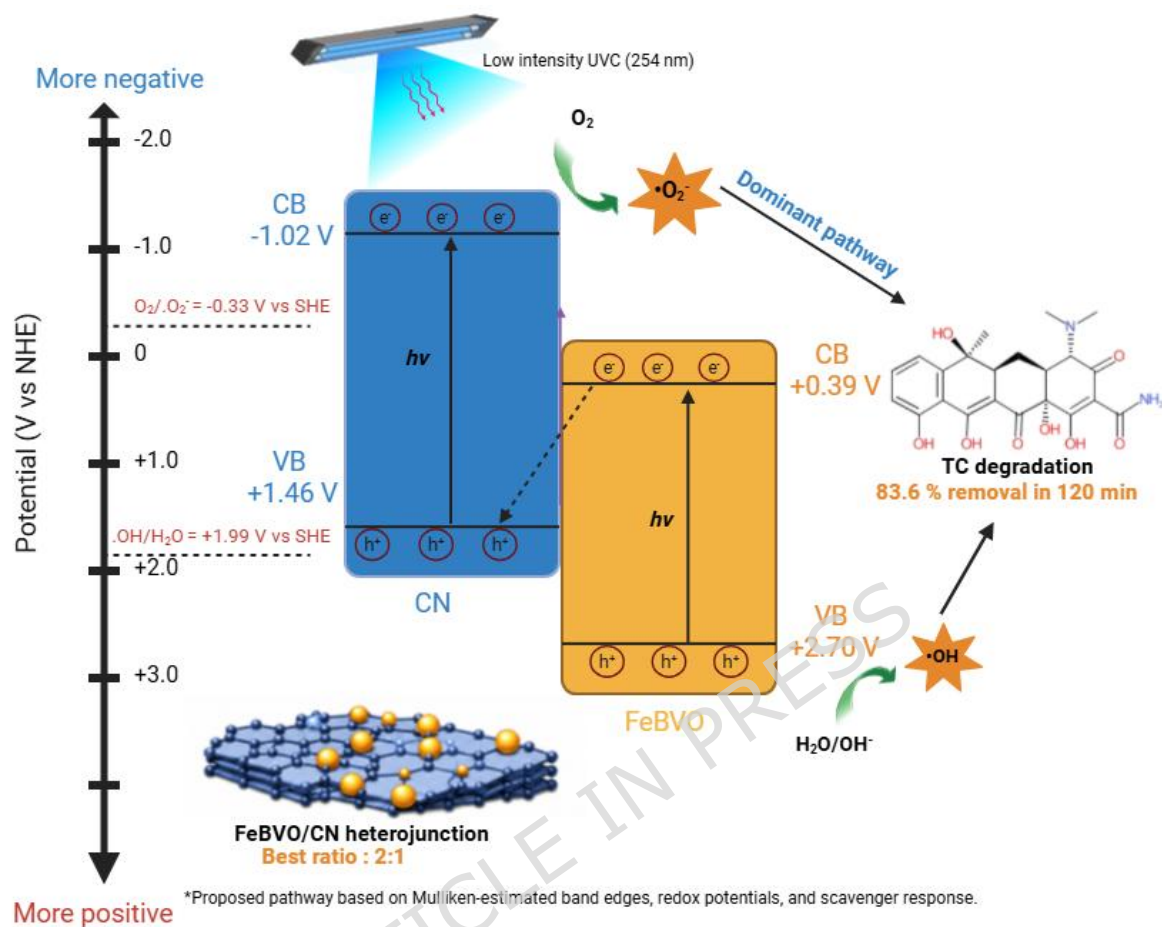


Fig. 8. Proposed Z-scheme-assisted charge-transfer and TC degradation pathway over FeBVO/CN-2:1 under low-intensity UVC irradiation.

3.4 Comparison with previous studies

To place the performance of FeBVO/CN-2:1 in context, Table 3 compares the present photocatalyst with recent BVO/CN-based systems reported for TC degradation. The comparison includes photocatalyst composition, synthesis method, irradiation source, pollutant concentration, degradation efficiency, reaction time, and apparent rate constant where available. The FeBVO/CN-2:1 photocatalyst achieved 83.6% TC degradation within 120 min under low-intensity UVC irradiation, with an apparent PFO rate constant of 0.01509 min^{-1} . This performance is comparable with several reported BVO/CN-based photocatalysts, although some systems show higher degradation efficiency or faster kinetics. These higher-performing systems often involve more complex architectures, such as ternary composites [90], doped carbon nitride [91], or quantum dots [92]. In contrast, the present system uses a relatively simple Fe-doped BVO/CN heterojunction prepared through hydrothermal treatment

and calcination. In addition, several literature systems relied on more complex architectures, such as ternary composites [90], doped carbon nitride [91], or quantum dots [92], while the present work used a simpler FeBVO/CN heterojunction design.

Direct comparison across studies should be made cautiously because photocatalytic performance is strongly influenced by experimental conditions, including pollutant concentration, catalyst loading, solution pH, irradiation type, lamp intensity, reaction volume, and reaction duration. Some studies used visible light, simulated sunlight, xenon lamps, or higher-power irradiation sources, whereas the present work was conducted under a 13 W UVC sterilizer lamp. Therefore, the main contribution of this study is not to claim universal superiority, but to show that composition-controlled FeBVO/CN photocatalysts can achieve effective TC degradation under low-power UVC irradiation when the FeBVO ratio and operating conditions are optimized.

The comparison also highlights the need for more standardized reporting in photocatalytic TC degradation studies. Catalyst loading, irradiation intensity, photon flux, pollutant concentration, pH, dark adsorption control, and kinetic fitting quality should be reported consistently to allow more reliable benchmarking. In this study, catalyst loading, reaction volume, pH, replicate number, error bars, PFO rate constants, and R^2 values are reported to improve comparability.

Table 3. Comparison of FeBVO/CN-2:1 with recent BVO/CN-based photocatalysts reported for TC degradation.

No.	Photocatalyst	Synthetic Method	Light source	Pollutant	Degradation efficiency (%)/ time (min)	Degradation rate constant (min ⁻¹)
1	g-C ₃ N ₄ /BiVO ₄	Solvothermal	Visible	TC	89%/150min	0.0117
2	rGO@g-C ₃ N ₄ /BiVO ₄	Microwave irradiation	Visible	TC	92%/50min	0.098
3	g-C ₃ N ₄ /BiVO ₄	Microwave-hydrothermal	Visible	TC	87%/240 min	0.0117
4	g-C ₃ N ₄ QDs/BiVO ₄	Calcination + hydrothermal	Xenon	TC	73.8%/60 min	0.0109
5	BiVO ₄ /rGO/g-C ₃ N ₄	Hydrothermal	Simulated sunlight	TC	81.10%/60 min	0.1384
6	rGO@g-C ₃ N ₄ /BiVO ₄	One-step microwave	Visible	TC	92%/50 min	0.098

irradiation						
7	BiVO ₄ /g-C ₃ N ₄	Hydrothermal		Simulated visible light	TC	90.7%/60 min 0.0141
8	Bi ₂ S ₃ /BiVO ₄ /g-C ₃ N ₄	Hydrothermal		Visible	TC	80.7%/90 min N
9	BiVO ₄ /LHSCN (BiVO ₄ on hollow sponge g-C ₃ N ₄)	Calcination + hydrothermal		Simulated sunlight	TC	94.9%/180 min 0.0623
10	VN-g-C ₃ N ₄ /BiVO ₄	Thermal polymerization + heterojunction assembly		Visible	TC	76.89%/100 min N
11	LaFeO ₃ /BiVO ₄ /g-C ₃ N ₄	Hydrothermal		Visible	TC	84.5%/120 min N
12	BiVO ₄ /g-C ₃ N ₄ /UiO-66-NH ₂	Hybrid calcination		Visible	TC	93.2%/120 min N
13	BiVO ₄ /g-C ₃ N ₄ (BCN21)	Ultrasonication-assisted synthesis		Visible (26 W lamp)	TC	78.2%/60 min N
14	C-g-C ₃ N ₄ /BiVO ₄ /PVDF photocatalytic membrane	Electrostatic self-assembly + electrospinning		Xenon lamp (>420 nm)	TC	83.33% /NR N
15	Fe-doped BiVO ₄ /g-C ₃ N ₄	Hydrothermal		Low intensity UV lamp (13W)	TC	83.6%/120 min 0.0150

NR: not reported. Direct comparison should be interpreted cautiously because reaction conditions differ across studies.

3.5 Preliminary cost analysis

A preliminary cost analysis was conducted to estimate the laboratory-scale synthesis cost of the FeBVO/CN photocatalyst. The calculation included reagent consumption, electricity use, and selected equipment-related costs for a synthesis batch producing approximately 2.4 g of composite photocatalyst. The main reagents considered were Bi(NO₃)₃·5H₂O, NH₄VO₃, Fe(NO₃)₃·9H₂O, NaOH, melamine, ethanol, and deionized water. As summarized in Table 4, the estimated total synthesis cost was RM 41.07 per batch, corresponding to approximately RM 17.11 per gram of catalyst. The cost breakdown shows that raw materials account for

the largest fraction of the overall expense, representing about 63% of the total cost. This contribution is mainly associated with the relatively high unit prices of bismuth and vanadium precursors as well as the stoichiometric requirements needed to obtain the desired BVO phase. At larger production scales, the cost of raw materials is expected to decrease further through bulk chemical purchasing and process optimization. In comparison, process energy costs from hydrothermal treatment, calcination, centrifugation, and drying contributed a smaller fraction of the total estimate.

The use of a 13 W UVC lamp during photocatalytic testing suggests lower irradiation power than high-intensity UV or xenon lamp systems. However, the present study did not measure irradiance, photon flux, continuous-operation energy demand, catalyst recovery cost, pH adjustment cost, or post-treatment neutralization cost. Therefore, the cost result should not be interpreted as proof of large-scale economic feasibility.

This analysis provides only an initial laboratory-scale cost estimate. A complete techno-economic assessment would need to include bulk chemical pricing, synthesis scale-up, reactor design, labor, catalyst recovery, catalyst regeneration, pH adjustment, neutralization, waste handling, metal leaching control, real wastewater performance, and long-term cycling stability. Future work should benchmark FeBVO/CN against commercial photocatalysts, conventional advanced oxidation processes, and other BVO- or CN-based systems under comparable treatment conditions.

3.6 Study scope and limitations

The findings should be interpreted within the experimental scope of the study. Photocatalytic performance was evaluated using synthetic TC solutions under a 13 W UVC source and optimized alkaline conditions; therefore, matrix effects and chemical conditioning requirements in real wastewater remain unresolved. UV-Vis monitoring demonstrates transformation of the TC chromophore but does not establish mineralization or transformation-product toxicity. The Mulliken band-edge calculations and scavenger experiments provide thermodynamic and reaction-based support for the proposed charge-transfer pathway, although they do not directly track interfacial carrier migration or individual ROS. Finally, the three-cycle test and post-reaction FTIR analysis support short-term reusability and surface chemical preservation, while long-term structural stability and metal release require separate evaluation. These boundaries define the present study as a composition-performance assessment of FeBVO/CN under low-intensity UVC irradiation rather than a full-scale treatment or definitive mechanistic validation study.

Table 4. Estimated material, process energy, and equipment costs for laboratory-scale synthesis of FeBVO/CN photocatalyst.

Category	Chemical/Material	Quantity/batch	Unit Cost (RM)	Total Cost (RM)	Purpose	
Raw Material	$\text{Bi}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$	3.3954 g	2.38/g	8.08	Main source	Bi
	NH_4VO_3	0.8189 g	1.25/ g	1.02	Main source	V
	$\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$	2.430 g	2.25/g	5.47	Fe source for ferrite formation	
	NaOH pellet	4.012 g	0.74/g	2.97	Precipitating agent	
	$\text{C}_2\text{H}_5\text{OH}$	30 mL	0.11/mL	3.30	Washing solvent	
	Deionized water	300 mL	0.01/mL	3.00	Reaction medium and washing	
	Melamine	6.360 g	0.30/g	1.90	Main source	g- C_3N_4
Total Material Cost (RM)				25.74		
Synthesis Process	Hydrothermal heating (180°C)	1.5 kWh X 15 h	RM 0.24 per kWh*	5.40	Energy cost for autoclave operation	
	Calcination (500 °C, 2 h)	3.0 kW X 4 h		2.88	Crystallization and phase stabilization	
	Filtration and washing (centrifugation)	0.13 kW x 1 h		0.03	Separation of solid product and solvent removal	
	Drying (oven at 80 °C, 12 h)	1.5 kW X 12 h		4.32	Post-treatment to remove residual moisture	
Total Process Cost (RM)				12.63		

Equipment	Teflon-lined autoclave usage	1.40	
Depreciation			
	Centrifuge/filtration setup	1.30	
Total Equipment Cost (RM)		2.70	
Overall Cost (RM)		41.07	2.4 g /batch

Notes: *refer to the Sarawak Energy tariff rate.

**Cost estimate is based on laboratory-scale synthesis and does not include labor, catalyst recovery, regeneration, pH adjustment, post-treatment neutralization, waste handling, real wastewater testing, or long-term operation

4.0 Conclusion

This study developed composition-controlled FeBVO/CN heterojunction photocatalysts for TC degradation under low-intensity UVC irradiation. The main contribution of this work is not Fe doping alone or BVO/CN heterojunction formation alone, since both strategies have been reported previously. Instead, this study advances the field by showing how the FeBVO mass ratio influences the structural, optical, morphological, kinetic, and operational behavior of the photocatalyst under a lower-power irradiation condition. Among the synthesized materials, FeBVO/CN-2:1 showed the highest photocatalytic activity, achieving 83.6% TC degradation within 120 min under optimized conditions. This performance was plausibly associated with preserved monoclinic BVO structure, broader optical response, suitable FeBVO composition, and effective interfacial contact between FeBVO and CN. Operational optimization showed that catalyst loading, solution pH, and initial TC concentration strongly affected TC degradation. The most favorable condition was obtained at 200 mg L⁻¹ catalyst loading, pH 10, and 5 mg L⁻¹ TC. The pH result also highlights an important practical constraint because alkaline adjustment may increase treatment cost and require post-treatment neutralization.

Scavenger experiments indicated that electron-mediated O₂ reduction and •O₂⁻-related pathways exerted the strongest influence on TC degradation, while h⁺- and •OH-related oxidation also contributed. Mulliken electronegativity calculations estimated the CB/VB potentials of CN at -1.02/+1.46 V and FeBVO at +0.39/+2.70 V versus NHE. These band positions are less consistent with a conventional Type-II pathway, in which electrons would accumulate on the FeBVO CB with insufficient reduction potential for O₂ reduction to •O₂⁻. In contrast, the proposed Z-scheme-like pathway retains strongly reducing electrons on the CN CB and strongly oxidizing holes on the FeBVO VB, providing a thermodynamic basis for the reactive-species behavior observed in the scavenger experiments.

FeBVO/CN-2:1 retained 77.1% degradation efficiency after three photocatalytic cycles, while the principal post-reaction FTIR bands remained detectable, supporting short-term reusability and preservation of the major surface chemical features. The preliminary laboratory-scale synthesis cost was estimated at RM 41.07 per 2.4 g batch, or approximately RM 17.11 g⁻¹, with raw materials accounting for the largest cost contribution. Overall, the findings identify the FeBVO:CN mass ratio as a key structure-performance variable and show that FeBVO/CN-2:1 supports efficient TC transformation under low-intensity UVC irradiation. The alkaline operating condition remains an important treatment constraint, while the mechanistic assignment should be regarded as a thermodynamically supported Z-scheme-like interpretation rather than direct evidence of interfacial carrier migration.

Compliance with Ethical Standards

Competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Ethics approval

Not applicable.

Consent to participate

Not applicable.

Consent for publication

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Generative AI and AI-assisted technologies

During the preparation of this manuscript the author(s) used ChatGPT by Open AI in order to refine the language and improve the overall readability. After using this tool/service, the author(s) reviewed and edited the content as needed and take(s) full responsibility for the content of the publication.

Author Contributions: Melissa Alexander Maran performed the experiments, analyzed the data, and prepared the original draft of the manuscript. Alvin Lim Teik Zheng supervised the

study, critically reviewed the manuscript, and oversaw the final editing. Yiizamy Suffian and Nor Hisam Zamakshshari performed FESEM and XRD analysis. Tan Kar Ban and Supakorn Boonyuen contributed to investigation of degradation mechanism and manuscript review. Hisham N. Farrag, Eric Lim Teik Chung, and Yoshito Andou contributed to manuscript review and final approval. All authors read and approved the final manuscript.

Data Availability

All data generated or analysed during this study are included in this published article.

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