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Comprehensive investigation of CuFe₂O₄/ZnO/RGO ternary nanocomposites for enhanced photocatalytic degradation of acid orange dye

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Abstract

A CuFe₂O₄/ZnO/RGO ternary nanocomposite and its pristine materials was synthesized and investigated for the photocatalytic degradation of acid orange. The synthesized materials were characterized using XRD, FTIR, SEM–EDX, elemental mapping, XPS, and UV–Vis spectroscopy. XRD confirmed the coexistence of CuFe₂O₄, ZnO, and RGO phases, with crystallite sizes of 19.23 nm, 26.42 nm, and 17.24 nm for CuFe₂O₄, ZnO, and CuFe₂O₄/ZnO/RGO, respectively. SEM–EDX and elemental mapping confirmed the good interfacial contact and homogeneous distribution of Cu, Fe, Zn, O, and C. XPS confirmed Cu²⁺, Fe²⁺/Fe³⁺, and Zn²⁺ chemical states, together with lattice and surface oxygen species and the RGO carbon framework. UV–Vis analysis revealed optical band gaps of 1.57 eV, 3.41 eV, 2.75 eV, and 1.41 eV for CuFe₂O₄, ZnO, RGO, and CuFe₂O₄/ZnO/RGO, respectively. Under sunlight irradiation, the ternary nanocomposite achieved 85% acid orange degradation under the initial photocatalytic comparison. At the optimized catalyst dosage of 30 mg, the degradation efficiency increased to 88%, with a rate constant of $2.155 \pm 0.176 \times 10^{-2} \text{ min}^{-1}$. The optimized acidic condition of pH 3 ± 0.1 further enhanced the degradation efficiency to 95%, with a rate constant of $2.953 \pm 0.306 \times 10^{-2} \text{ min}^{-1}$. Radical scavenger experiments

confirming the dominant contribution of $\cdot\text{OH}$, followed by h^+ and $\cdot\text{O}_2^-$, while e^- showed a comparatively lower contribution. The photocatalyst retained approximately 81% degradation efficiency after ten consecutive cycles.

Keywords: $\text{CuFe}_2\text{O}_4/\text{ZnO}/\text{RGO}$, Photocatalysis, Dye degradation, Radical analysis, Catalyst stability.

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1. Introduction

Industrial wastewater generated from textile, leather, paper, printing, cosmetic, and dye-manufacturing industries contains large amounts of synthetic dyes that pose serious environmental challenges. Among these, acid orange is one of the most widely used anionic azo dyes due to its excellent colour strength, high stability, and strong affinity toward natural and synthetic fibres [1, 2]. During dyeing processes, a significant fraction of acid orange remains unfixed and is discharged into wastewater, resulting in severe water pollution. The presence of acid orange in aquatic environments reduces light penetration, inhibits photosynthetic activity, decreases dissolved oxygen levels, and disrupts the ecological balance of aquatic ecosystems [3]. Moreover, the breakdown of acid orange may produce toxic aromatic amines, which are known to exhibit carcinogenic, mutagenic, and teratogenic effects, posing potential risks to both aquatic organisms and human health [4]. Conventional wastewater treatment methods, including adsorption, coagulation, membrane filtration, and biological treatment, often suffer from incomplete dye removal, high operational costs, and secondary waste generation [5]. Consequently, semiconductor photocatalysis has emerged as an efficient and environmentally sustainable advanced oxidation technology for the degradation of acid orange. Under light irradiation, photocatalysts generate reactive oxygen

species, particularly $\cdot\text{OH}$ and $\cdot\text{O}_2^-$ radicals, which effectively mineralize dye molecules into harmless products. However, rapid e^-/h^+ recombination limits photocatalytic performance, making heterojunction nanocomposites an attractive strategy for enhancing visible-light absorption, charge separation, and photocatalytic degradation efficiency.

Nanomaterials have attracted significant research interest because of their unique physicochemical properties, including high surface area, abundant active sites, tunable electronic structures, and excellent charge transport characteristics [6]. These features make them promising candidates for applications in environmental remediation, catalysis, energy conversion, sensing, and electronic devices. Among various nanomaterials, spinel ferrites have emerged as an important class of multifunctional semiconductors owing to their excellent chemical stability, magnetic behavior, low toxicity, and cost-effective synthesis [7]. Spinel ferrites, with the general formula MFe_2O_4 , where M represents a divalent metal ion, possess a well-defined spinel lattice in which metal cations are distributed between tetrahedral and octahedral sites [8]. The arrangement of these cations strongly influences their structural, optical, magnetic and electrical properties. Among them, CuFe_2O_4 has received considerable attention as a visible-light-responsive p-type semiconductor because of its narrow bandgap, excellent chemical stability, and magnetic separability after photocatalytic treatment [9]. ZnO is a widely investigated n-type semiconductor owing to its excellent chemical stability, non-toxicity, high electron mobility, and strong photocatalytic activity [10]. Despite these advantages, its relatively wide bandgap restricts light absorption mainly to the ultraviolet region, while the rapid recombination of photogenerated e^-/h^+ pairs and photocorrosion limit its photocatalytic efficiency under visible-light irradiation [11]. Furthermore, the incorporation of RGO significantly enhances photocatalytic performance because of its high electrical conductivity, large specific surface area, excellent electron-accepting capability, and abundant

active sites [12]. RGO serves as an efficient electron transport pathway, accelerating interfacial charge transfer and reducing charge carrier recombination [13].

Recently, various ternary nanocomposites, including Ag-CuFe₂O₄/rGO [14], CuFe₂O₄/g-C₃N₄/rGO [15], ZnO-CuFe₂O₄-CNTs [16], CoFe₂O₄/g-C₃N₄/rGO [17], BiVO₄/Cu₂O/grapheme [18], AgVO₃/rGO/CuFe₂O₄ [19], Ni-ZnO/g-C₃N₄ [20], NiO/ZnO/g-C₃N₄ [21], Chitosan/ZnO-Fe₃O₄ [22], have been investigated for photocatalytic dye degradation. The ternary nanocomposites provides enhanced photocatalytic activity and stability compared with the corresponding pristine materials through improved light utilization, charge separation, and interfacial interactions. However, many reported ternary photocatalysts involve relatively complex synthesis routes, such as chemical reduction-assisted ultrasonication, hydrothermal, and sonochemical methods, which may require multiple processing steps and extended preparation time.

In this present work, the present study focuses on developing a simple and less time-consuming ultrasonication-assisted approach for the preparation of CuFe₂O₄/ZnO/RGO ternary nanocomposites. To the best of our knowledge, the CuFe₂O₄/ZnO/RGO ternary nanocomposite prepared by this simple approach for Acid Orange degradation has not been comprehensively reported. The synergistic combination of p-type CuFe₂O₄, n-type ZnO, and conductive RGO is expected to promote interfacial charge separation and enhance photocatalytic activity. The synthesized nanocomposite was systematically characterized, and its photocatalytic performance was evaluated by investigating the effects of pH, catalyst dosage, reactive species, degradation kinetics, and recyclability. The results demonstrate the potential of CuFe₂O₄/ZnO/RGO as an efficient and reusable photocatalyst for wastewater treatment.

2. Experimental

2.1. Materials

Analytical grade of Copper nitrate trihydrate (Cu(NO₃)₂·3H₂O), ferric nitrate nonahydrate (Fe(NO₃)₃·9H₂O), zinc nitrate hexahydrate (Zn(NO₃)₂·6H₂O), urea

(NH_2CONH_2), citric acid monohydrate ($\text{C}_6\text{H}_8\text{O}_7 \cdot \text{H}_2\text{O}$), graphite powder, potassium permanganate (KMnO_4), sodium nitrate (NaNO_3), concentrated sulfuric acid (H_2SO_4), hydrochloric acid (HCl), and hydrogen peroxide (H_2O_2) were used as received without further purification. Deionized water was used throughout all the experimental procedures.

2.2. Preparation of CuFe_2O_4

Briefly, 0.1 M of $\text{Cu}(\text{NO}_3)_2 \cdot 3\text{H}_2\text{O}$, 0.2 M of $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$, and 0.4 M of $\text{C}_6\text{H}_8\text{O}_7 \cdot \text{H}_2\text{O}$ were separately dissolved in 100 mL of deionized water. The copper nitrate solution was slowly added to the ferric nitrate solution under continuous stirring, followed by the addition of citric acid to form a homogeneous metal–citrate sol. The resulting sol was subjected to microwave irradiation for 20 min, producing a porous precursor through rapid combustion. The obtained powder was then calcined at 800 °C for 3 h in a muffle furnace and finally ground to obtain fine CuFe_2O_4 nanoparticles.

2.3. Preparation of ZnO

A 0.2 M of $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ and a 0.4 M of NH_2CONH_2 were separately prepared in 100 mL of deionized water. The two solutions were then combined under continuous magnetic stirring to obtain a homogeneous precursor solution. Subsequently, the precursor solution was subjected to microwave irradiation for 20 min, resulting in the formation of a dry porous precursor. The obtained precursor was calcined in a muffle furnace at 800 °C for 3 h and then finely ground to obtain pure ZnO nanoparticles.

2.4. Preparation of RGO

Graphene oxide was synthesized using a modified Hummers' method. Briefly, 2 g of graphite powder was dispersed in 98 mL of concentrated H_2SO_4 under an ice bath and stirred for 1 h. Subsequently, 8 g of KMnO_4 and 4 g of NaNO_3 were gradually added under continuous stirring to initiate the oxidation process. The reaction mixture was maintained at 35 °C for 24 h, followed by the slow addition of H_2O_2 to terminate the reaction. The resulting GO was

repeatedly washed with dilute HCl and deionized water, sonicated, and dried at 80 °C. The dried GO was then thermally reduced at 350 °C for 30 min to obtain RGO without the use of chemical reducing agents.

2.5. Preparation of CuFe₂O₄/ZnO/RGO

The CuFe₂O₄/ZnO/RGO nanocomposite was synthesized using a simple ultrasonication method. Initially, 0.5 g of CuFe₂O₄, 0.5 g of ZnO, and 0.1 g of RGO were separately dispersed in 50 mL of deionized water and ultrasonicated for 30 min. The three suspensions were then combined and further ultrasonicated for 1 h. The resulting suspension was centrifuged, and the collected product was dried in a hot-air oven at 80 °C for 12 h to obtain the CuFe₂O₄/ZnO/RGO nanocomposite.

2.6. Characterization

The crystal structure and phase purity of the samples were analyzed by X-ray diffraction (XRD, PANalytical, Netherlands) using Cu K α radiation ($\lambda = 1.5406 \text{ \AA}$). Fourier transform infrared (FTIR) spectra were recorded using a Shimadzu IRTracer-100 (4000–400 cm⁻¹) to identify functional groups. The surface morphology, elemental composition, and elemental mapping were examined using a High-Resolution Scanning Electron Microscope (HR-SEM, Thermo Scientific Apreo S) equipped with energy-dispersive X-ray spectroscopy (EDS). The surface chemical composition and oxidation states were investigated using X-ray Photoelectron Spectroscopy (XPS, PHI VERSAPROBE III). Optical properties were measured using a Shimadzu UV-3600 Plus UV–Vis–NIR spectrophotometer in the wavelength range of 200–1200 nm.

2.7. Photocatalytic experiment

The photocatalytic performance of all prepared materials was evaluated for the degradation of Acid Orange (AO) dye under direct natural sunlight. A standard AO solution was prepared by dissolving 10 mg of AO dye in 1 L of distilled water. For each experiment,

10 mg of photocatalyst was dispersed in 100 mL of the dye solution and magnetically stirred in the dark for 30 min to establish adsorption–desorption equilibrium. During the dark adsorption period, the reaction vessel was completely covered with an opaque black sheet to eliminate ambient light exposure while maintaining continuous magnetic stirring. After the dark equilibration, the suspension was exposed to direct natural sunlight for 90 min under continuous stirring. The sunlight intensity was maintained within the range of 80,000–100,000 lux, as measured using a digital lux meter before each experiment. At 15 min intervals, 3 mL aliquots were withdrawn and centrifuged to separate the photocatalyst. The residual AO concentration was determined using a Bio-Spectrometer Fluorescence instrument. The photocatalytic degradation efficiency and reaction kinetics were calculated using Eqs. (1) and (2).

$$D\% = \left(\frac{A_0 - A_t}{A_0} \right) \times 100 \quad (1)$$

$$\ln \frac{A_0}{A_t} = k_{obs} t \quad (2)$$

where A_0 and A_t represent the initial and residual dye concentrations at time t , respectively, and k_{obs} (min^{-1}) is the pseudo-first-order rate constant. The pseudo-first-order model was employed since the catalyst concentration remained constant throughout the reaction. In addition, the effects of catalyst dosage and solution pH on the photocatalytic degradation efficiency were investigated under optimized reaction conditions. Furthermore, cycling stability and radical scavenger analyses were performed to evaluate the reusability of the photocatalyst and to identify the active reactive species involved in the degradation process.

3. Results and discussions

The XRD patterns of the prepared materials are shown in Fig. 1(a). For CuFe_2O_4 , the diffraction peaks observed at $2\theta = 18.24^\circ, 29.91^\circ, 34.54^\circ, 35.75^\circ, 37.09^\circ, 43.83^\circ, 53.87^\circ, 57.01^\circ, 57.97^\circ, 62.02^\circ$, and 63.93° correspond to the (101), (112), (103), (211), (202), (220),

(312), (303), (321), (224), and (400) planes, respectively, and are indexed to the tetragonal CuFe_2O_4 phase (JCPDS No. 00-034-0425) [23]. The weak diffraction peaks at $2\theta = 24.07^\circ$, 33.16° , and 49.40° can be assigned to the (012), (104), and (024) planes, respectively, corresponding to the rhombohedral $\alpha\text{-Fe}_2\text{O}_3$ phase (JCPDS No. 01-084-0306) [24]. In addition, the minor peak at $2\theta = 38.55^\circ$, corresponding to the (200) plane, is attributed to the monoclinic CuO phase (JCPDS No. 00-005-0661) [25]. For ZnO , the diffraction peaks at $2\theta = 31.74^\circ$, 34.38° , 36.17° , 47.52° , 56.47° , 62.75° , 66.24° , 67.81° , and 69.12° correspond to the (100), (002), (101), (102), (110), (103), (200), (112), and (201) planes, respectively, confirming the hexagonal structure of ZnO (JCPDS No. 03-065-0725) [16]. For RGO, the broad diffraction peak centered at $2\theta = 25.84^\circ$, corresponding to the (002) plane [12]. The XRD pattern of the $\text{CuFe}_2\text{O}_4/\text{ZnO}/\text{RGO}$ nanocomposite exhibits the characteristic diffraction peaks corresponding to the respective pristine CuFe_2O_4 and ZnO phases, with the RGO related peak also retained. No additional impurity or secondary-phase peaks are observed, confirming the successful formation of the $\text{CuFe}_2\text{O}_4/\text{ZnO}/\text{RGO}$ nanocomposite without any detectable new crystalline impurity phases.

The crystallite size (D) of the prepared materials was estimated using the Debye–Scherrer formula, as shown in Eq. (2).

$$D = \frac{K\lambda}{\beta \cos \theta} \quad (3)$$

where, λ is X-ray wavelength ($1.5406 \text{ \AA}$ for $\text{Cu K}\alpha$), β is the full width at half maximum (FWHM), θ is the Bragg angle and K is the shape factor. Accordingly, the calculated crystallite sizes were 19.23, 26.42, and 17.24 nm for CuFe_2O_4 , ZnO , and $\text{CuFe}_2\text{O}_4/\text{ZnO}/\text{RGO}$ nanocomposite, respectively. The relatively broad diffraction peak of RGO is associated with its turbostratic and disordered graphitic structure, making accurate crystallite-size estimation by the Debye–Scherrer method less reliable [26]. The smaller crystallite size of the $\text{CuFe}_2\text{O}_4/\text{ZnO}/\text{RGO}$ nanocomposite provides a higher accessible surface area and more active

sites, facilitating better interaction between the photocatalyst and pollutant molecules and thereby contributing to enhanced photocatalytic performance.

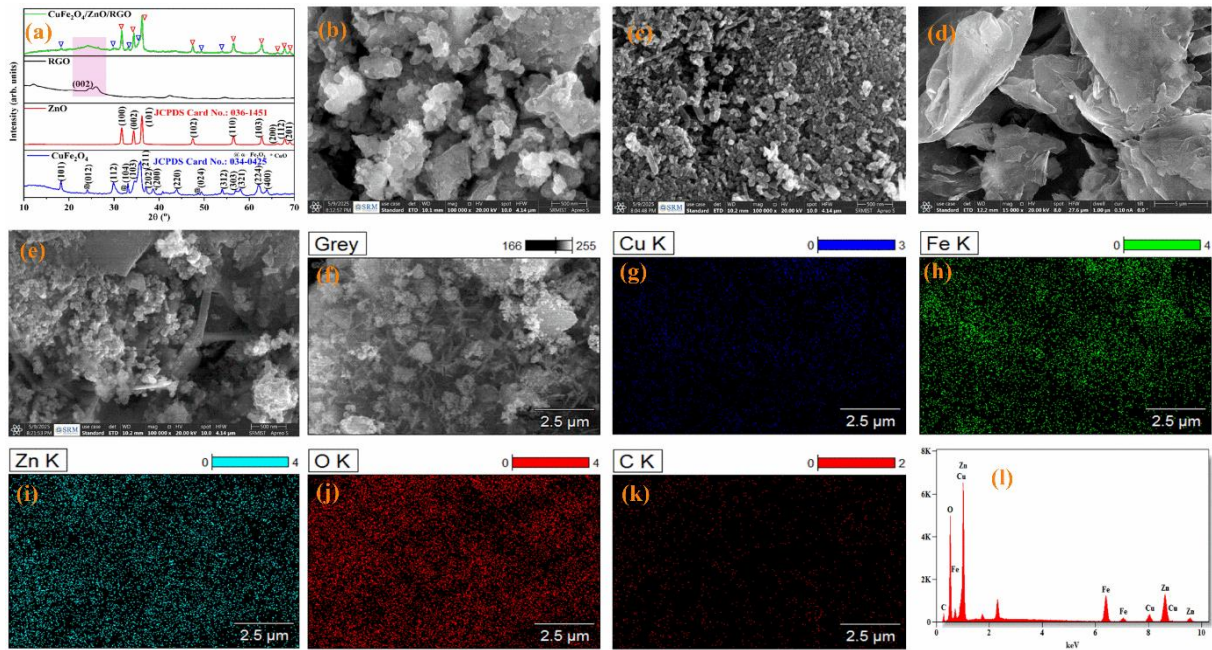


Fig. 1. (a) XRD patterns of the prepared materials; SEM images of (b) CuFe₂O₄, (c) ZnO, (d) RGO, and (e) CuFe₂O₄/ZnO/RGO; elemental mapping images of (f) Grey, (g) Cu K, (h) Fe K, (i) Zn K, (j) O K, and (k) C K; and (l) EDX spectrum of the CuFe₂O₄/ZnO/RGO nanocomposite.

Fig. 1(b-e) shows the SEM images of the prepared CuFe₂O₄, ZnO, RGO, and CuFe₂O₄/ZnO/RGO nanocomposite, respectively. Fig. 1(b) shows CuFe₂O₄ with an irregular, aggregated granular morphology, indicating the formation of closely packed nanoparticles [27]. Fig. 1(c) displays ZnO consisting of fine, densely distributed identical particles. Fig. 1(d) shows the characteristic bunch of sheet-like and layered morphology of RGO, with wrinkled and folded surfaces. Fig. 1(e) reveals the morphology of the CuFe₂O₄/ZnO/RGO nanocomposite, where the oxide nanoparticles are distributed over to the RGO sheets. The resulting rough and heterogeneous surface indicates good interfacial contact among the CuFe₂O₄, ZnO, and RGO components, which can provide numerous accessible surface sites [16]. Fig. 1(f-k) show the elemental mapping of the CuFe₂O₄/ZnO/RGO nanocomposite. Fig.

1(f) represents the corresponding grey-scale SEM image used for elemental mapping. Fig. 1(g–k) display the spatial distributions of Cu, Fe, Zn, O, and C, respectively. The relatively uniform distribution of all detected elements across the mapped area confirms the successful incorporation and homogeneous dispersion of the individual components within the ternary nanocomposite. Fig. 1(l) presents the EDX spectrum of the CuFe₂O₄/ZnO/RGO nanocomposite. The spectrum clearly shows characteristic signals corresponding to Cu, Fe, Zn, O, and C, confirming the elemental composition of the synthesized nanocomposite. Thus, the EDX results support the elemental mapping and further confirm the formation of the CuFe₂O₄/ZnO/RGO ternary nanocomposite.

As shown in Fig. 2(a), the FTIR spectra of CuFe₂O₄, ZnO, RGO, and the CuFe₂O₄/ZnO/RGO ternary nanocomposite provide information about the chemical bonding, surface functional groups, and metal–oxygen coordination in the prepared materials. For pristine CuFe₂O₄, the characteristic absorption bands observed around 420 cm⁻¹ and 530 cm⁻¹ is Cu–O and Fe–O stretching vibrations, respectively, confirming the formation of the spinel CuFe₂O₄ structure [14]. A broad absorption band around 3470 cm⁻¹ is attributed to the O–H stretching vibration of surface hydroxyl groups and adsorbed moisture, while the band near 1070 cm⁻¹ can be associated with C–O-related surface vibrations. For ZnO, the absorption band observed around 600 cm⁻¹ is characteristic of the Zn–O stretching vibration, confirming the presence of ZnO [16]. The band near 1040 cm⁻¹ is assigned to C–O-related vibrations, whereas the broad band around 3400 cm⁻¹ corresponds to O–H stretching vibrations arising from surface hydroxyl groups [20]. The FTIR spectrum of RGO exhibits characteristic bands around 1020 cm⁻¹ and 1470 cm⁻¹, which are associated with C–O and C–OH vibrations, respectively. The broad O–H band around 3400 cm⁻¹ is also observed, indicating the presence of residual oxygen-containing functional groups [15]. However, its relatively reduced intensity compared with oxygen-rich graphene oxide indicates the effective reduction of oxygenated

functional groups during the conversion to RGO [17]. The CuFe₂O₄/ZnO/RGO ternary nanocomposite retains the characteristic metal–oxygen vibrations of CuFe₂O₄ and ZnO together with the characteristic carbon–oxygen and hydroxyl-related bands of RGO. The coexistence of these characteristic vibrational features confirms the successful incorporation of CuFe₂O₄, ZnO, and RGO within the ternary nanocomposite [14]. The changes in the relative intensity and slight variation in the positions of some bands can be attributed to interfacial interactions between CuFe₂O₄, ZnO, and the RGO framework, suggesting the formation of strong interfacial coupling among the three components.

The XPS spectra of the prepared CuFe₂O₄/ZnO/RGO ternary nanocomposite are presented in Fig. 2(b–g). The wide-scan survey spectrum in Fig. 2(b) confirms the presence of Cu, Fe, Zn, O and C elements in the nanocomposite, demonstrating the successful incorporation of the CuFe₂O₄, ZnO and RGO components. The characteristic Cu 2p, Fe 2p, Zn 2p, O 1s and C 1s signals are clearly observed in the survey spectrum. The elemental signals observed in the survey spectrum are further examined through the corresponding high-resolution XPS spectra. The high-resolution Zn 2p spectrum in Fig. 2(c) exhibits two distinct peaks at binding energies of 1044.31 and 1021.33 eV, which are assigned to Zn 2p_{1/2} and Zn 2p_{3/2}, respectively. The energy separation between the Zn 2p_{3/2} and Zn 2p_{1/2} components is approximately 22.98 eV, which is characteristic of Zn²⁺ in ZnO. The absence of additional chemically shifted Zn components indicates that Zn predominantly exists in the Zn²⁺ oxidation state [21]. Thus, the Zn 2p spectrum confirms the formation and chemical state of the ZnO component in the ternary nanocomposite. The high-resolution Cu 2p spectrum in Fig. 2(d) consists of the characteristic Cu 2p_{3/2} and Cu 2p_{1/2} core-level components together with satellite features. The main Cu 2p_{3/2} peak is located at 933.05 eV, whereas the Cu 2p_{1/2} component appears at 953.15 eV. The corresponding spin–orbit separation is approximately 20.10 eV, which is consistent with Cu²⁺ species. In addition to the principal Cu 2p peaks, satellite features are observed at

approximately 940.02, 943.04 and 961.37 eV [19]. These satellite peaks are characteristic shake-up features associated with Cu^{2+} and arise from final-state effects during photoemission. The presence of these characteristic Cu^{2+} satellite structures, together with the main Cu 2p peaks, provides strong confirmation that Cu predominantly exists in the Cu^{2+} oxidation state in the CuFe_2O_4 phase.

The Fe 2p spectrum in Fig. 2(e) exhibits the characteristic spin–orbit-split Fe $2p_{3/2}$ and Fe $2p_{1/2}$ regions. The fitted components indicate the coexistence of Fe^{2+} and Fe^{3+} species. In the Fe $2p_{3/2}$ region, components are observed at approximately 709.09 and 710.37 eV, which are assigned to Fe^{2+} and Fe^{3+} related chemical states, respectively, according to the assignments shown in the spectrum. The higher-binding-energy features around 713.24 eV and 718.94 eV are associated with satellite contributions. In the Fe $2p_{1/2}$ region, components are observed at approximately 722.35 eV and 724.58 eV, corresponding to Fe^{2+} and Fe^{3+} species, respectively. An additional satellite feature is observed around 734.57 eV [15]. The separation between the principal Fe $2p_{3/2}$ and Fe $2p_{1/2}$ components is approximately 14.2 eV, consistent with the spin–orbit splitting of Fe 2p states. The O 1s spectrum in Fig. 2(f) were deconvoluted into two components located at 529.55 and 531.12 eV. The lower-binding-energy component at 529.55 eV is corresponding to lattice oxygen (O^{2-}) associated with metal–oxygen bonds in CuFe_2O_4 and ZnO, particularly the Cu–O, Fe–O and Zn–O environments. The higher-binding-energy component at 531.12 eV can be assigned to surface-related oxygen species, such as hydroxyl groups, defect-associated oxygen and/or chemisorbed oxygen species [19]. The presence of both lattice and surface oxygen components indicates that oxygen exists in different chemical environments within the ternary nanocomposite.

The C 1s spectrum in Fig. 2(g) provides information about carbon framework and residual oxygen-containing functional groups of RGO. The dominant peak at 284.03 eV is assigned to sp^2 -hybridized carbon associated with C=C bonds in RGO. The peaks appeared at

286.12 eV is attributed to C–O-containing groups, such as hydroxyl/epoxy functionalities. The peak at 288.13 eV is associated with carboxyl-type oxygenated carbon species (O–C=O) [17]. A weak feature at 292.38 eV is assigned to a shake-up satellite associated with the $\pi-\pi^*$ transition of the carbon structure [28]. The predominance of the sp^2 C=C component at 284.03 eV, together with the relatively weaker oxygen-containing carbon components, indicates that the oxygenated functional groups were substantially reduced during the conversion of graphene oxide to RGO, while a small amount of residual oxygen functionality remained on the RGO surface.

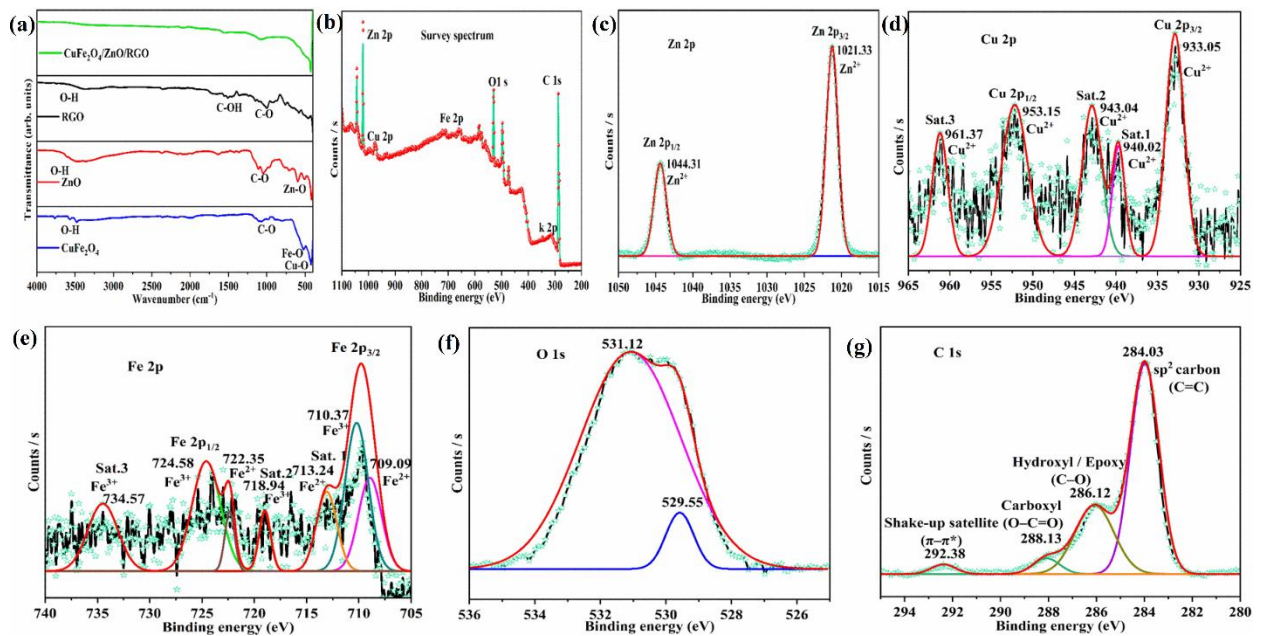


Fig. 2. (a) FTIR analysis of all prepared materials; XPS analysis of CuFe₂O₄/ZnO/RGO nanocomposite: (b) survey spectrum, (c) Zn 2p, (d) Cu 2p, (e) Fe 2p, (f) O 1s and (g) C 1s

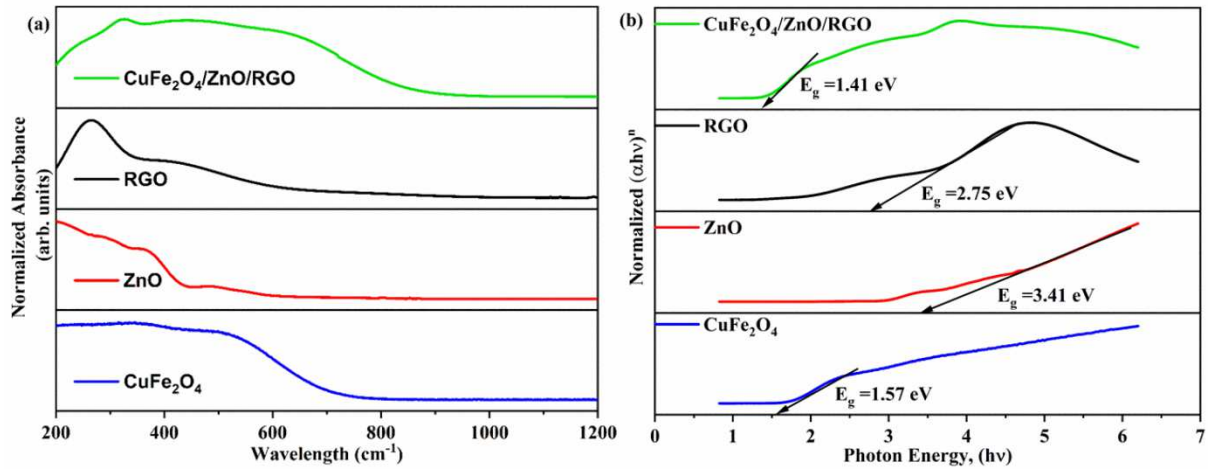


Fig. 3. (a) UV-Vis absorbance spectra (b) Tauc plot for all prepared materials

Fig. 3. (a) The UV–Vis absorption spectra of the prepared CuFe₂O₄, ZnO, RGO, and CuFe₂O₄/ZnO/RGO demonstrate their distinct optical absorption characteristics. CuFe₂O₄ exhibits a broad absorption edge extending toward the visible region, whereas ZnO and RGO shows a relatively stronger absorption mainly in the UV region [14]. Notably, the CuFe₂O₄/ZnO/RGO ternary nanocomposite exhibits enhanced and broadened optical absorption, indicating improved light-harvesting capability resulting from the interfacial interaction among the constituent materials. The optical bandgap was calculated by using Eq. 4 [16].

$$(\alpha h\nu)^n = A(h\nu - E_g) \quad (4)$$

where α is the absorption coefficient, $h\nu$ is the photon energy, A is a proportionality constant, and E_g is the optical band-gap energy. The exponent n depends on the type of electronic transition ($n = 2$ is direct and $n = 1/2$ is indirect transition). Thus, the optical band gap is determined by plotting $(\alpha h\nu)^n$ versus $h\nu$ and extrapolating the linear portion of the curve to $(\alpha h\nu)^n = 0$ as shown in Fig. 3(b). The obtained band gaps are 1.57 eV for CuFe₂O₄, 3.41 eV for ZnO, 2.75 eV for RGO, and 1.41 eV for CuFe₂O₄/ZnO/RGO. The reduced band gap of the ternary nanocomposite indicates strong interfacial coupling, promoting efficient charge generation and separation.

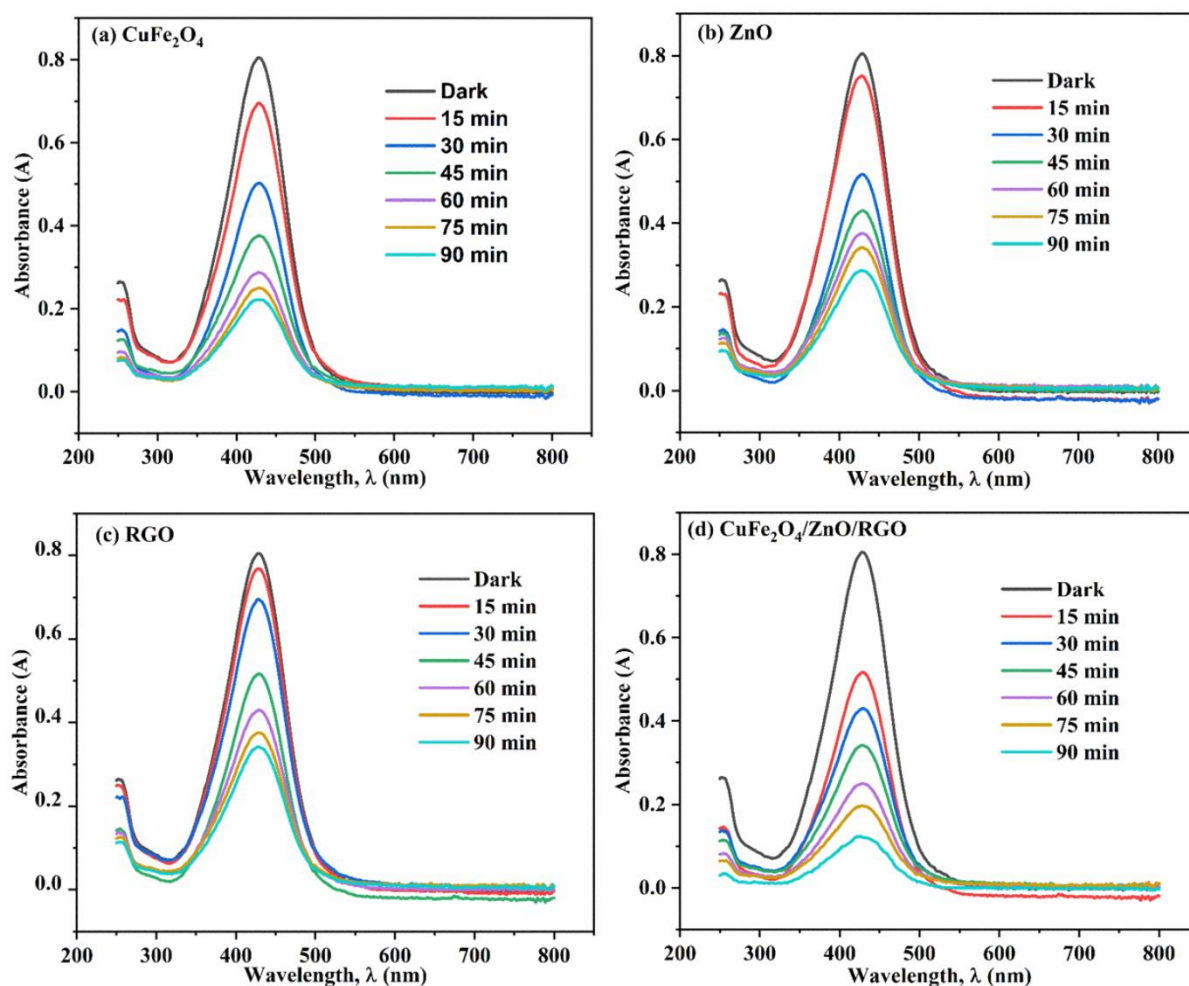


Fig. 4. Time-dependent absorbance spectra for the photocatalytic degradation of acid orange using (a) CuFe_2O_4 , (b) ZnO , (c) RGO , and (d) $\text{CuFe}_2\text{O}_4/\text{ZnO}/\text{RGO}$ photocatalysts.

The photocatalytic degradation of acid orange by the prepared photocatalysts was evaluated from the time-dependent absorbance spectra shown in Fig. 4(a–d). The characteristic absorption peak of acid orange progressively decreased with increasing irradiation time, confirming the continuous degradation of the dye. The corresponding dynamic degradation profiles are presented in Fig. 5(a). As shown in Fig. 5(b), the degradation efficiencies after 90 min were 72%, 65%, 57%, and 85% for CuFe_2O_4 , ZnO , RGO , and $\text{CuFe}_2\text{O}_4/\text{ZnO}/\text{RGO}$, respectively. The enhanced performance of the ternary nanocomposite indicates improved photocatalytic activity due to effective interfacial interactions among the pristine materials. The kinetic profiles in Fig. 5(c) were obtained using the pseudo-first-order kinetic model. The

observed rate constants (k_{obs}) were $1.954 \pm 0.107 \times 10^{-2} \text{ min}^{-1}$ for CuFe_2O_4 , $1.203 \pm 0.103 \times 10^{-2} \text{ min}^{-1}$ for ZnO , $1.073 \pm 0.089 \times 10^{-2} \text{ min}^{-1}$ for RGO, and $1.544 \pm 0.109 \times 10^{-2} \text{ min}^{-1}$ for $\text{CuFe}_2\text{O}_4/\text{ZnO}/\text{RGO}$. The corresponding linear fitting demonstrates the kinetic behavior of acid orange degradation.

Further, the ternary $\text{CuFe}_2\text{O}_4/\text{ZnO}/\text{RGO}$ nanocomposite exhibited enhanced photocatalytic activity compared with the individual photocatalysts. The effect of catalyst dosage on acid orange degradation was investigated by varying the catalyst amount from 20 to 40 mg, and the corresponding time-dependent absorbance spectra are presented in Fig. 6(a–c). The progressive decrease in the characteristic absorption intensity confirms the continuous degradation of acid orange with irradiation time. The corresponding dynamic degradation profiles and degradation efficiencies are shown in Fig. 6(d) and (e). The degradation efficiencies after 90 min were 86%, 88%, and 68% for 20 mg, 30 mg, and 40 mg of catalyst, respectively. The kinetic profiles shown in Fig. 6(f) indicate k_{obs} of $2.074 \pm 0.096 \times 10^{-2} \text{ min}^{-1}$, $2.155 \pm 0.176 \times 10^{-2} \text{ min}^{-1}$, and $1.958 \pm 0.147 \times 10^{-2} \text{ min}^{-1}$ for 20, 30, and 40 mg, respectively. The degradation efficiency increased from 20 to 30 mg because the higher catalyst dosage provides a greater number of available active sites for dye adsorption and photocatalytic reactions [29]. However, a further increase to 40 mg resulted in decreased degradation efficiency, due to particle aggregation, increased light scattering, and reduced light penetration through the catalyst suspension, thereby limiting the effective generation of photogenerated charge carriers [30]. Thus, 30 mg was identified as the optimum catalyst dosage under the investigated conditions.

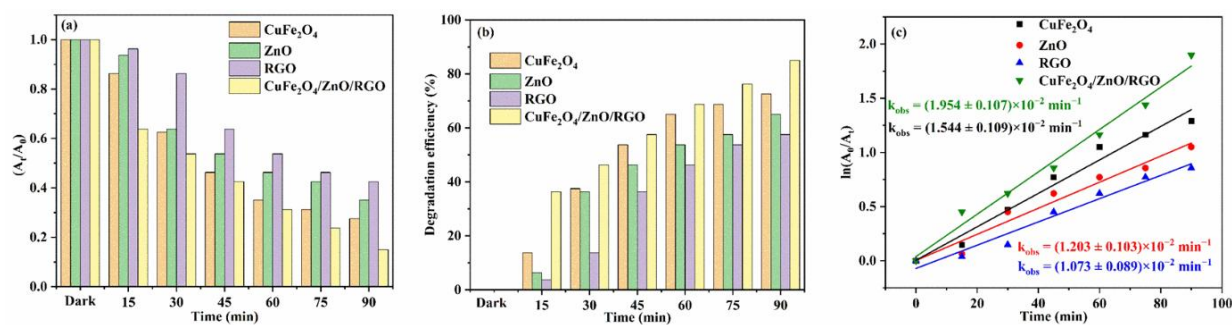


Fig. 5. (a) Dynamic degradation profile, (b) degradation efficiency, and (c) kinetic profile of acid orange photocatalytic degradation using the prepared photocatalysts.

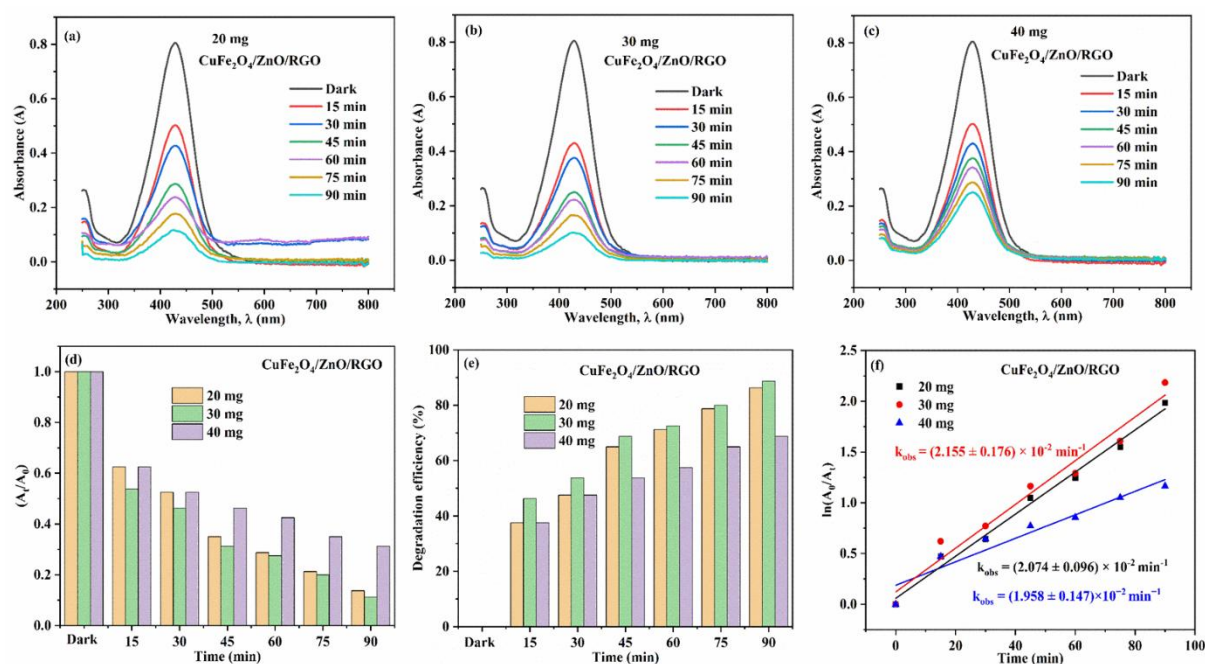


Fig. 6. Photocatalytic degradation of Acid Orange using CuFe₂O₄/ZnO/RGO nanocomposite at different catalyst dosages: (a–c) time-dependent absorbance spectra for catalyst dosages of 20–40 mg, (d) dynamic degradation profile, (e) degradation efficiency, and (f) pseudo-first-order kinetic profile.

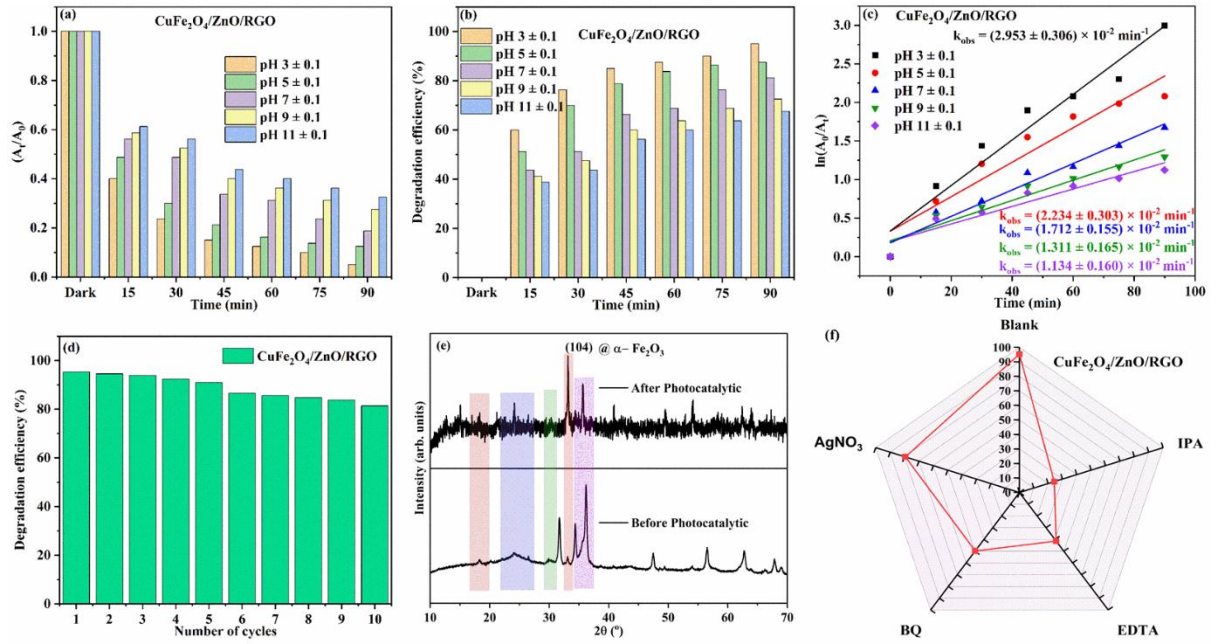


Fig. 7. Photocatalytic performance of the $\text{CuFe}_2\text{O}_4/\text{ZnO}/\text{RGO}$ nanocomposite toward acid orange degradation (a) dynamic degradation profiles at different pH values ($\text{pH } 3\text{--}11 \pm 0.1$), (b) corresponding degradation efficiencies, (c) pseudo-first-order kinetic plots at different pH values, (d) cycling stability of the $\text{CuFe}_2\text{O}_4/\text{ZnO}/\text{RGO}$ nanocomposite at the optimized pH of 3 ± 0.1 , (e) comparative XRD patterns of the $\text{CuFe}_2\text{O}_4/\text{ZnO}/\text{RGO}$ photocatalyst before and after the photocatalytic cycling experiment, and (f) radical scavenger analysis.

The effect of pH on the photocatalytic degradation of acid orange using an optimized catalyst dosage of 30 mg was analyzed for further pH-dependent photocatalytic degradation. The dynamic profile for pH-dependent acid orange degradation in the range of 3 ± 0.1 to 11 ± 0.1 is shown in Fig. 7(a). It is observed that, with increasing pH from 3 to 11 ± 0.1 , the degradation efficiency gradually decreased from 95% to 67% as shown in Fig. 7(b). The degradation efficiencies were 95%, 87%, 81%, 72%, and 67% at $\text{pH } 3 \pm 0.1$, 5 ± 0.1 , 7 ± 0.1 , 9 ± 0.1 , and 11 ± 0.1 , respectively. The corresponding rate constants obtained from the kinetic plots shown in Fig. 7(c) were $2.953 \pm 0.306 \times 10^{-2} \text{ min}^{-1}$, $2.234 \pm 0.303 \times 10^{-2} \text{ min}^{-1}$, $1.712 \pm 0.155 \times 10^{-2} \text{ min}^{-1}$, $1.311 \pm 0.165 \times 10^{-2} \text{ min}^{-1}$, and $1.134 \pm 0.160 \times 10^{-2} \text{ min}^{-1}$ for $\text{pH } 3 \pm 0.1$, 5

± 0.1 , 7 ± 0.1 , 9 ± 0.1 , and 11 ± 0.1 , respectively. The progressive decrease in the rate constant with increasing pH confirms the reduction in the photocatalytic reaction rate under alkaline conditions.

The enhanced degradation under acidic conditions due to the surface-charge characteristics of the $\text{CuFe}_2\text{O}_4/\text{ZnO}/\text{RGO}$ photocatalyst and the ionic nature of acid orange. At lower pH, enhanced electrostatic interaction between the catalyst surface and dye molecules can promote adsorption and facilitate the transfer of photogenerated charge carriers to the catalyst–solution interface [31]. Furthermore, the acidic environment can favour the generation of reactive oxygen species, which participate in the oxidation of the dye molecules. As the pH increases, the surface charge and adsorption behaviour of the catalyst change, which can reduce the interaction between acid orange and the photocatalyst and consequently suppress the formation and utilization of reactive species [32]. Therefore, the photocatalytic degradation efficiency decreases with increasing pH.

Further, the cycling stability analysis was carried out, as shown in Fig. 7(d). After each photocatalytic cycle, the catalyst was separated by centrifugation, thoroughly washed several times with ethanol and deionized water to remove the residual dye and degradation intermediates, and dried at 70°C before being reused for the subsequent cycle [20]. The catalyst maintained high photocatalytic activity during repeated use, demonstrating good structural and operational stability. After ten cycles, the degradation efficiency remained around 81%, indicating that the $\text{CuFe}_2\text{O}_4/\text{ZnO}/\text{RGO}$ photocatalyst possesses excellent cycling stability and can be effectively reused for acid orange degradation. After the photocatalytic cycles, the reused catalyst was subjected to XRD analysis, as shown in Fig. 7(e). The XRD pattern of the reused photocatalyst retained the characteristic diffraction features of the catalyst. However, a prominent diffraction peak at 33.13° corresponding to the (104) plane of $\alpha\text{-Fe}_2\text{O}_3$ [24], was observed after photocatalytic treatment. The appearance or enhancement of this peak may be

attributed to photocorrosion or partial phase modification of the photocatalyst during prolonged photocatalytic irradiation. Nevertheless, the major diffraction features were retained, indicating that the $\text{CuFe}_2\text{O}_4/\text{ZnO}/\text{RGO}$ composite maintains considerable structural stability after repeated photocatalytic operation.

Further, radical scavenger analysis was carried out to identify the major reactive species involved in Acid Orange degradation, as shown in Fig. 7(f). IPA, EDTA, BQ, and AgNO_3 were employed as scavengers for hydroxyl radicals ($\cdot\text{OH}$), photogenerated holes (h^+), superoxide radicals ($\cdot\text{O}_2^-$), and photogenerated electrons (e^-), respectively [15, 16]. The degradation efficiency decreased significantly to 24% in the presence of IPA, indicating that $\cdot\text{OH}$ radicals play a dominant role in the photocatalytic degradation process. Similarly, the addition of EDTA reduced the degradation efficiency to 41%, confirming the significant contribution of photogenerated h^+ . In the presence of BQ, the degradation efficiency decreased to 49%, demonstrating the substantial involvement of $\cdot\text{O}_2^-$. In contrast, the addition of AgNO_3 resulted in a degradation efficiency of 79%, indicating that photogenerated e^- have a comparatively lesser direct contribution to Acid Orange degradation.

These results indicate that $\cdot\text{OH}$, h^+ , and $\cdot\text{O}_2^-$ collectively play major roles in the photocatalytic degradation of acid orange, while photogenerated electrons contribute to a lesser extent. The overall contribution of the reactive species can be considered in the order $\cdot\text{OH} > h^+ > \cdot\text{O}_2^- > e^-$. Under irradiation, the $\text{CuFe}_2\text{O}_4/\text{ZnO}/\text{RGO}$ photocatalyst generates photogenerated e^-/h^+ pairs. The photogenerated holes participate in the oxidation of surface-adsorbed species and promote the formation of highly reactive $\cdot\text{OH}$ radicals, while the photogenerated electrons can interact with dissolved oxygen to generate $\cdot\text{O}_2^-$ radicals [33]. These reactive species subsequently attack the acid orange molecules, leading to their degradation into smaller intermediate products and ultimately contributing to the mineralization of the dye. Thus, the

scavenger results confirm the synergistic involvement of hydroxyl radicals, photogenerated holes, and superoxide radicals in the acid orange photocatalytic degradation process.

The proposed photocatalytic mechanism of the CuFe₂O₄/ZnO/RGO nanocomposite is illustrated in Fig. 8. The band-edge positions were estimated using the Mulliken electronegativity approach. The conduction-band potential (E_{CB}) and valence-band potential (E_{VB}) were calculated using the following Eqs. (5) and (6) [21].

$$E_{VB} = \chi - E^e + 0.5E_g \quad (5)$$

$$E_{CB} = E_{VB} - E_g \quad (6)$$

where χ is the absolute electronegativity of the semiconductor, E^e is the energy of free electrons on the hydrogen scale (~ 4.5 eV), and E_g is the optical band gap. Accordingly, the calculated E_{CB}/E_{VB} values are 0.60/2.17 eV for CuFe₂O₄, -0.41/2.99 eV for ZnO, and -1.12/1.62 eV for RGO, respectively were the values are summarized in Table 1.

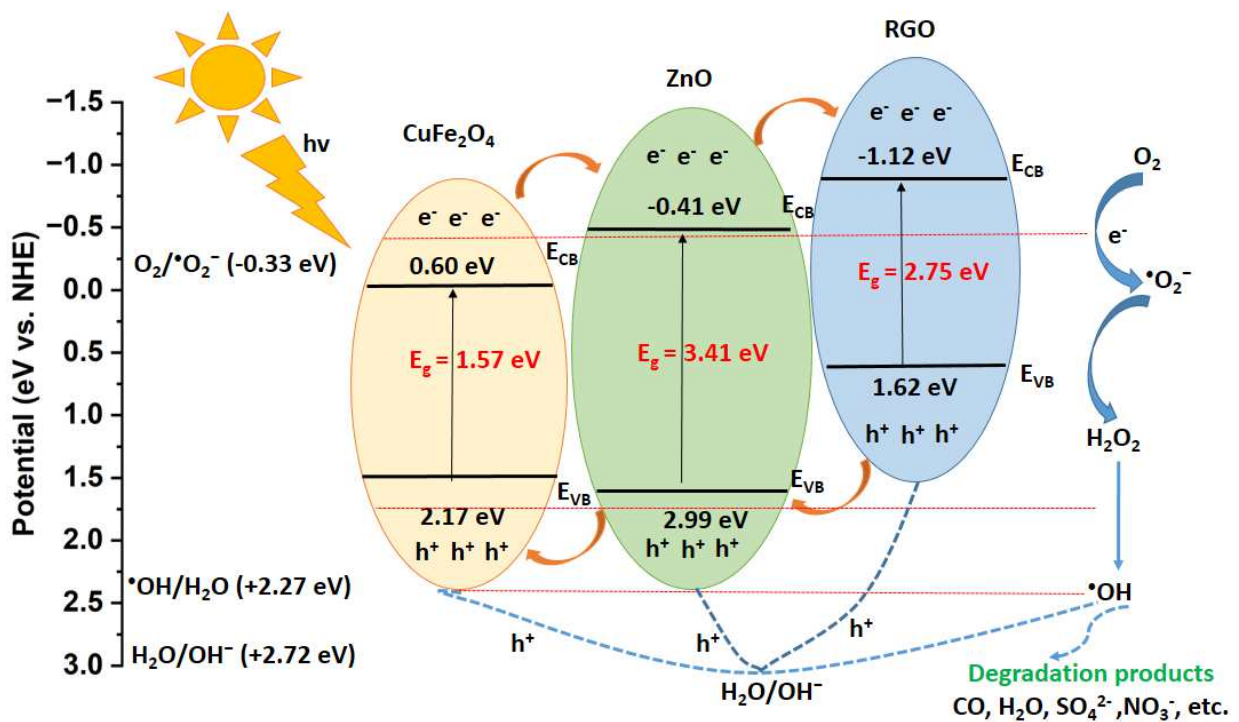
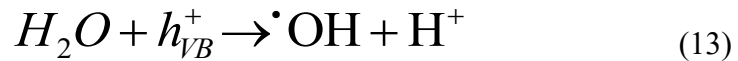
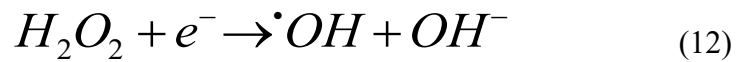
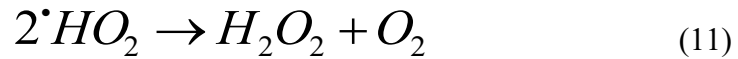
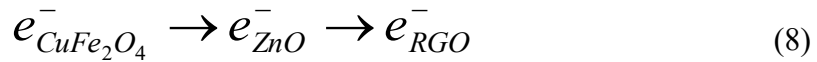
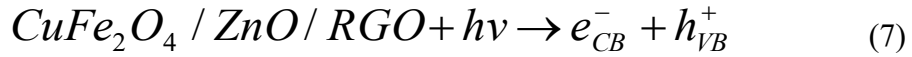


Fig. 8. Schematic illustration of the proposed photocatalytic degradation mechanism of acid orange over the CuFe₂O₄/ZnO/RGO nanocomposite.

Under solar irradiation, the CuFe_2O_4 , ZnO , and RGO components are photoexcited, generating e^-/h^+ pairs. Owing to the favourable interfacial band alignment, photogenerated electrons can migrate from CuFe_2O_4 toward ZnO and subsequently to the RGO , where RGO acts as an effective electron-accepting and charge-transport component. The highly negative E_{CB} of RGO (-1.12 eV) is sufficiently negative to reduce dissolved O_2 to superoxide radicals ($\cdot\text{O}_2^-$; $\text{O}_2/\cdot\text{O}_2^- = -0.33$ eV). The photogenerated holes retained in the semiconductor phases participate in oxidation reactions, with the relatively positive VB of ZnO ($+2.99$ eV) favouring the generation of highly reactive hydroxyl species through surface hydroxyl/water oxidation pathways. The generated $\cdot\text{O}_2^-$, $\cdot\text{OH}$, and h^+ species subsequently attack the acid orange molecules, leading to their decomposition and eventual formation of smaller degradation products such as CO_2 , H_2O , SO_4^{2-} , and NO_3^- . Thus, the $\text{CuFe}_2\text{O}_4/\text{ZnO}/\text{RGO}$ heterostructure promotes efficient charge separation and transfer, suppresses e^-h^+ recombination, and enhances photocatalytic degradation. The overall reaction mechanism as shown in Eqs. (7)–(14).



Photocatalyst + Acid Orange + $\cdot\text{OH}/\cdot\text{O}_2^- / h^+ \rightarrow \text{Degradation intermediates}$ (14)

Table 1. Calculated band gap, and band-edge potentials of CuFe₂O₄, ZnO, and RGO.

Material	χ (eV)	E _g (eV)	E _{CB} (eV)	E _{VB} (eV)
CuFe ₂ O ₄	5.89	1.57	0.60	2.17
ZnO	5.79	3.41	−0.41	2.99
RGO	4.75	2.75	−1.12	1.62

Table 2. Comparison of the photocatalytic performance of CuFe₂O₄/ZnO/RGO with previously reported photocatalysts.

Materials	Synthesis method	Pollutant	Light source	Degradation (%)	Time (min)	Rate constant (min ^{−1})	Catalyst reusability and stability	Ref.
Ag-CuFe ₂ O ₄ /rGO	Chemical reduction-ultrasonication	Rhodamine-B	500 W Xe lamp ($\lambda > 420$ nm)	96.27	210	−0.0068	90.89% after 5 cycles	[14]
CuFe ₂ O ₄ /g-C ₃ N ₄ /rGO	Hydrothermal	Reactive Black 5	UVA with intensity 12.50 W m ²	88.5	150	0.1159	Stable efficiency over 5 cycles	[15]
ZnO-CuFe ₂ O ₄ -CNTs	Ultrasonication	Congo red	Sunlight	82	120	0.013	-	[16]
CoFe ₂ O ₄ /g-C ₃ N ₄ /rGO	Hydrothermal	Indigo Carmine	UVA with intensity 1250 $\mu\text{W cm}^2$	96.99	120	0.0235	negligible variation over 4 cycles	[17]
BiVO ₄ /Cu ₂ O/graphene	Hydrothermal	Rhodamine-B	300 W light source (>420 nm filter)	96	120	-	Stable efficiency over 5 cycles	[18]
AgVO ₃ /rGO/CuFe ₂ O ₄	Hydrothermal	4-Nitrophenol	Xe lamp (300 W; ~100 mW/cm ²)	94.7	240	0.01841	85.3% after 6 cycles	[19]
Ni-ZnO/g-C ₃ N ₄	Hydrothermal	Reactive Black 5	Solar irradiation (68–73 Klux)	89.7	120	-	No obvious loss after 5 cycles	[20]
NiO/ZnO/g-C ₃ N ₄	Hydrothermal	Methylene blue	Tungsten lamp (500 W/420 nm cutoff filter)	~95	90	0.02396	~90% after 3 cycles	[21]
Chitosan/ZnO-Fe ₃ O ₄	Sonochemical	Methyl orange	Visible light	99.49	150	0.031	81.63% retained after 4 cycles	[22]
CuFe ₂ O ₄ /ZnO/RGO	Ultrasonication	Acid orange	Sunlight	95	90	2.953×10^{-2}	81% after 10 cycles	Present work

The comparison of the photocatalytic performance of CuFe₂O₄/ZnO/RGO with previously reported photocatalysts are summarized in Table 2. Pavasupree et al. [14] reported an Ag-CuFe₂O₄/rGO heterojunction that achieved 96.27% Rhodamine-B degradation within 210 min under a 500 W Xe lamp, with 90.89% efficiency retained after five cycles. Rajendran et al. [15] reported CuFe₂O₄/g-C₃N₄/rGO for Reactive Black 5 degradation, achieving about 88.5% degradation within 150 min under UVA irradiation, with good recyclability. Afaq et al. [16] developed ZnO-CuFe₂O₄-CNTs, which showed 82% Congo red degradation within 120 min under sunlight. Leelert et al. [17] reported CoFe₂O₄/g-C₃N₄/rGO for Indigo Carmine degradation, achieving 96.99% degradation within 120 min with good cycling stability. Liaquat et al. [18] reported BiVO₄/Cu₂O/graphene, which achieved 96% Rhodamine-B degradation within 120 min and maintained stable activity over five cycles. Wei et al. [19] demonstrated 94.7% 4-nitrophenol degradation using AgVO₃/rGO/CuFe₂O₄ within 240 min, retaining 85.3% activity after six cycles. Kavitha et al. [20] reported Ni-ZnO/g-C₃N₄, which achieved 89.7% Reactive Black 5 degradation within 120 min with negligible activity loss after five cycles. Jayaprakash et al. [21] reported approximately 95% methylene blue degradation using NiO/ZnO/g-C₃N₄ within 90 min, with approximately 90% activity retained after three cycles. Quy et al. [22] reported 99.49% methyl orange degradation using Chitosan/ZnO-Fe₃O₄ within 150 min, retaining 81.63% activity after four cycles. In comparison, the present CuFe₂O₄/ZnO/RGO nanocomposite achieved 95% acid orange degradation within only 90 min under sunlight, with a rate constant of $2.953 \times 10^{-2} \text{ min}^{-1}$ and 81% degradation efficiency retained after ten cycles, demonstrating its excellent photocatalytic activity and recyclability.

The enhanced photocatalytic activity of the CuFe₂O₄/ZnO/RGO ternary nanocomposite is strongly correlated with its physicochemical properties. XRD confirms the successful coexistence of CuFe₂O₄, ZnO, and RGO with a reduced crystallite size of 17.24 nm, while SEM-EDX and elemental mapping reveal good interfacial contact and homogeneous

distribution of Cu, Fe, Zn, O, and C. FTIR and XPS further confirm the characteristic metal–oxygen bonds and the corresponding Cu^{2+} , $\text{Fe}^{2+}/\text{Fe}^{3+}$, Zn^{2+} , lattice oxygen, and RGO chemical states, supporting strong interfacial coupling. UV–Vis analysis shows broadened light absorption and a reduced band gap of 1.41 eV, while the calculated band-edge positions favour charge separation and electron transfer through RGO. Consequently, these combined structural, morphological, chemical, and optical characteristics promote reactive-species generation and contribute to the enhanced acid orange degradation performance of the ternary nanocomposite.

4. Conclusions

In this study, a $\text{CuFe}_2\text{O}_4/\text{ZnO}/\text{RGO}$ ternary nanocomposite was successfully synthesized through a simple ultrasonication-assisted approach and systematically investigated for the photocatalytic degradation of acid orange under natural sunlight. XRD confirmed the coexistence of CuFe_2O_4 , ZnO, and RGO phases, while SEM–EDX and elemental mapping revealed good interfacial contact and homogeneous distribution of the constituent elements. FTIR and XPS analyses confirmed the characteristic chemical bonds and oxidation states of the ternary system. UV–Vis analysis demonstrated enhanced light absorption with a reduced optical band gap of 1.41 eV, while the calculated band-edge positions indicated favourable charge-transfer pathways. The optimized catalyst dosage of 30 mg achieved 88% acid orange degradation within 90 min, and acidic conditions significantly enhanced the photocatalytic activity, reaching 95% degradation at $\text{pH } 3 \pm 0.1$ with a rate constant of $2.953 \times 10^{-2} \text{ min}^{-1}$. Radical-scavenger experiments confirmed that $\cdot\text{OH}$, h^+ , and $\cdot\text{O}_2^-$ were the major reactive species involved in the degradation process, with $\cdot\text{OH}$ showing the dominant contribution. The photocatalyst also exhibited good recyclability, retaining approximately 81% degradation efficiency after ten cycles.

Elumalai Arulkumar: Formal analysis, Data curation, Investigation, Writing – original draft, Writing – review & editing.

Gopinath Dhamodaran: Conceptualization, Methodology, Formal analysis, Writing – review & editing

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