



Green Synthesis of MnFe_2O_4 Spinel Nanoparticles Using *Camellia sinensis* Extract: Optical Bandgap Engineering and Sunlight-driven Photocatalytic Wastewater Remediation

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ABSTRACT

The conventional synthesis of spinel ferrite nanoparticles often involves hazardous chemicals and energy-intensive processes, thereby limiting environmental sustainability. In this study, manganese ferrite nanoparticles (MnFe_2O_4 NPs) were synthesized via a green co-precipitation route using *Camellia sinensis* extract (0.015–0.035 g/mL) as a reducing and stabilizing agent, followed by calcination at 540 °C for 3 h. XRD analysis confirmed the formation of single-phase cubic spinel MnFe_2O_4 with crystallite sizes ranging from 9.68 ± 1.01 nm to 9.41 ± 0.94 nm. Optical studies revealed a significant reduction in the bandgap from 2.33 eV to 1.11 eV with increasing extract concentration, indicating enhanced visible-light absorption. The synthesized nanoparticles exhibited effective photocatalytic degradation of methylene blue under natural sunlight, achieving degradation efficiencies of $43.0 \pm 2.0\%$, $44.0 \pm 1.8\%$, and $44.5 \pm 2.1\%$, with corresponding rate constants of 1.15×10^{-2} , 1.22×10^{-2} , and $1.45 \times 10^{-2} \text{ min}^{-1}$. Radical scavenging studies identified superoxide ($\text{O}_2^{\cdot-}$) and hydroxyl ($\cdot\text{OH}$) radicals as the dominant reactive species. Phytotoxicity assessment using *Vigna radiata* showed improved seed germination ($89.6 \pm 5.4\%$) in treated water compared with that in the untreated dye solution ($79.8 \pm 5.3\%$), confirming reduced toxicity. These findings highlight the potential of green-synthesized MnFe_2O_4 NPs as sustainable photocatalysts for wastewater remediation and the safe reuse of agricultural water.

Keywords: MnFe_2O_4 nanoparticles; Calcination; Photocatalytic degradation; Optical bandgap; Sustainability.

1. INTRODUCTION

Spinel ferrite nanoparticles, including CoFe_2O_4 , MnFe_2O_4 , CuFe_2O_4 , and NiFe_2O_4 , have attracted considerable interest due to their exceptional physicochemical features and many technological applications. MnFe_2O_4 was a p-type semiconductor characterized by a significantly broad optical bandgap. It demonstrates significant light absorption at ultraviolet and visible spectra, rendering it extremely appropriate for photocatalytic uses. Eco-friendly ZnO coatings synthesized by banana peel and green tea extracts were immobilized on alumina substrates using the doctor blade technique to prevent catalyst loss during photocatalysis. ZnO/GTE 30 (green tea extract-based coating) achieved the highest ciprofloxacin degradation efficiency (89.1%) under simulated solar light and showed good reusability in river water (Jovanović *et al.* 2026). A green-supported nanocatalyst (nSOD@CuO-NPs) derived from

aluminum sludge, rice husk waste, and green tea-mediated CuO nanoparticles was developed for large-scale degradation of crystal violet and methylene blue (Oviedo *et al.* 2025). Recycled glass-derived MCM-41 decorated with green tea-fabricated Co_3O_4 nanoparticles formed a visible-light-active photocatalyst for methyl parathion degradation, achieving complete removal within 60–90 min. The low bandgap (1.63 eV), hydroxyl radical-driven mechanism, formation of intermediate compounds, and five-cycle reusability confirmed its efficiency for pesticide remediation (Abdel *et al.* 2020). Detoxification was confirmed by improved germination index in *Vigna radiata*, demonstrating potential for wastewater treatment and biomedical applications (Li *et al.* 2021).

Fungus-mediated green synthesis of AgNPs using *Fusarium oxysporum* produced spherical nanoparticles (~40 nm) with confirmed antimicrobial and

photocatalytic activity against methylene blue (MB). Additionally, AgNPs enhanced the seed germination of *Vigna radiata* at optimized concentration (0.5 mg/mL), indicating agricultural and environmental applicability (Gupta *et al.* 2021). *Carissa macrocarpa* fruit extract-assisted AgNPs (CMFE@AgNPs) exhibited selective Hg²⁺ sensing (LOD 18.45 µM), strong antibacterial and antioxidant activity, and effective sunlight-driven degradation of methyl orange (77.3%) and methylene blue (88.9%). The stable spherical nanoparticles (~50 nm) demonstrated multifunctional potential in wastewater remediation and biomedical applications (Siddique *et al.* 2025). Green-synthesized CdS nanoparticles using *Dicliptera roxburghiana* extract exhibited spherical morphology (2.5–8 nm), high stability, and a bandgap of 3.31 eV. The nanoparticles achieved 87.12% MB degradation under solar irradiation within 120 min, confirming their efficient photocatalytic performance (Ullah *et al.* 2021). *Geranium wallichianum*-mediated Se-doped Au/ZnO/Fe₃O₄ nanocomposite (GW@Se-AZF) demonstrated multifunctional performance, including selective Hg²⁺ sensing with a limit of detection (LOD) of 92.7 nM and rapid sunlight-driven degradation of dyes (~89–91%) within 60 min. The crystalline, quasi-spherical nanocomposite (~22 nm) integrates sensing, photocatalytic, and magnetic properties for sustainable water remediation (Ullah *et al.* 2026). Green-synthesized lanthanum vanadate (LaVO₄) nanoparticles using *Colocasia esculenta* leaf extract achieved 98% MB degradation under visible light due to an optimal 2.8 eV bandgap and favorable surface properties (Mallikarjunaswamy *et al.* 2026).

Nigella sativa-assisted Ag/TiO₂ and gum Arabic-stabilized Ag/TiO₂/AG nanocomposites exhibited reduced particle size, narrowed bandgap (3.26–2.85 eV), and enhanced colloidal stability. The gum-stabilized composite showed antioxidant activity, moderate antibacterial effects, and 45.7% MB degradation, demonstrating eco-compatible multifunctionality (Alorfi *et al.* 2025). Green-synthesized ZnO nanoparticles from *Lupinus albus* and *Lupinus pilosus* extracts exhibited antibacterial properties and MB degradation efficiencies ranging from 47% to 61%. Characterization confirmed phytochemical-assisted nanoparticle formation, supporting their applicability in photocatalytic and antimicrobial applications (Mirza *et al.* 2023).

Bombax malabaricum gum-mediated Fe₃O₄ nanoparticles (~9 ± 2 nm) demonstrated strong antibacterial, antifungal, antioxidant, and photocatalytic activity, achieving 98.72% MB degradation optimized via response surface methodology (Awais *et al.* 2023). The green-synthesized Ag nanoparticles using *Endostemon viscosus* extract nanoparticles showed low zebrafish embryo toxicity below 500 µg/mL, highlighting biomedical and wastewater treatment

potential (Chinnasamy *et al.* 2023). *Muntingia calabura*-mediated cobalt oxide nanoparticles (CoO NPs, bandgap of 2.05 eV, and crystallite size of 27.6 nm) displayed mesoporosity and antiferromagnetic behavior with high surface activity. The CoO NPs achieved 88% MB degradation under sunlight following first-order kinetics ($k = 0.0065 \text{ min}^{-1}$) (Vinayagam *et al.* 2023). Green-synthesized CuO nanoparticles using *Camellia sinensis* and *Prunus africana* extracts showed strong antimicrobial activity against carbapenem-resistant bacteria (up to 30 mm inhibition zone) and 85.5% MB degradation (Ssekatawa *et al.* 2022).

Magnetic Fe–Mn spinel particle electrodes derived from red mud and shield slag enabled complete MB removal in a three-dimensional electrochemical reactor within 240 min. *In situ* MnFe₂O₄ formation promoted Fe/Mn redox cycling, hydroxyl radical generation, high recyclability (≥94.83% over 10 cycles), and cost-effective wastewater treatment (Li *et al.* 2026). Cobalt-doped Mn_xCo_{1-x}Fe₂O₄ nanoferrites synthesized by co-precipitation exhibited reduced bandgap (1.75 eV), enhanced magnetization (55.39 emu/g), and improved dielectric properties. The nanocomposites showed superior visible-light MB degradation, confirmed by Liquid Chromatography–Mass Spectrometry (LC–MS) analysis and excellent recyclability for catalytic and energy applications (Nanjappan *et al.* 2025). Green-synthesized MnFe₂O₄/TiO₂ nanocomposites (5.3–7.1 nm crystallites) exhibited superparamagnetic behavior and enhanced UV-driven photocatalytic performance, achieving 98% MB degradation within 2 h (Puspitarum *et al.* 2025). Hydrothermally synthesized MnFe₂O₄ NPs (23.98 nm, cubic spinel structure) demonstrated strong UV-induced photocatalytic activity with up to 99% MB degradation under optimized pH, catalyst dose, and exposure time. Structural parameters and magnetic properties confirmed their suitability for textile wastewater treatment (Ata *et al.* 2021).

Despite advances in the green synthesis of metal oxide nanoparticles, a few studies have focused on *Camellia sinensis*-mediated MnFe₂O₄ synthesis without chemical additives, particularly with controlled tuning of its optical and catalytic properties. Existing research largely emphasizes degradation efficiency, with limited insight into bandgap engineering, charge dynamics, and integrated evaluation, including recyclability and phytotoxicity under natural sunlight. This study addresses these gaps by presenting a fully green synthesis approach with a tunable bandgap (2.33–1.11 eV), demonstrating enhanced sunlight-driven photocatalytic performance with mechanistic insights, and validating environmental safety through *Vigna radiata* phytotoxicity assessment, thereby enabling sustainable wastewater remediation and agricultural water reuse.

2. MATERIALS AND METHODS

2.1 Extraction of *Camellia sinensis*

Camellia sinensis specimens, sourced from a local market in Erode, Tamilnadu, India, were utilized as reducing agents. The *C. sinensis* extract was made by crushing 5 g of dried plant material and combining it with 250 mL of deionized water while stirring continuously for 15 min (Thakur *et al.* 2025). The solution was then heated to 85 °C with continuous stirring until the volume decreased to 150 mL, after which the pH was adjusted to approximately 5. The resultant mixture was filtered twice using a vacuum-assisted filtration system to ensure the complete removal of residual plant particles.

2.2 Preparation of Reaction Solutions

The metal precursor solution was prepared to maintain a 2:1 molar ratio. Accordingly, 0.4 mmol of

manganese(II) nitrate tetrahydrate and 1 mmol of iron(III) nitrate nonahydrate were dissolved in 10 mL of distilled water under continuous stirring. The resulting solution exhibited a pH of approximately 5.

2.3 Synthesis of MnFe₂O₄ NPs

MnFe₂O₄ NPs were synthesized by gradually adding plant extract solution to metal precursor solution, maintaining continuous agitation for 4 min. The mixture was then heated to 80 °C for 90 min, reducing the solution volume to 10 mL and adjusting the pH to 1. The resulting solution was dried in an oven at 100 °C for 1 day, followed by calcination at 540 °C for 3 h. The nanoparticle-containing samples were labeled MF0.015, MF0.025, and MF0.035, corresponding to different concentrations (g/mL) of *C. sinensis* extract used. Fig. 1 illustrates the synthesis procedure for the MnFe₂O₄ NPs.

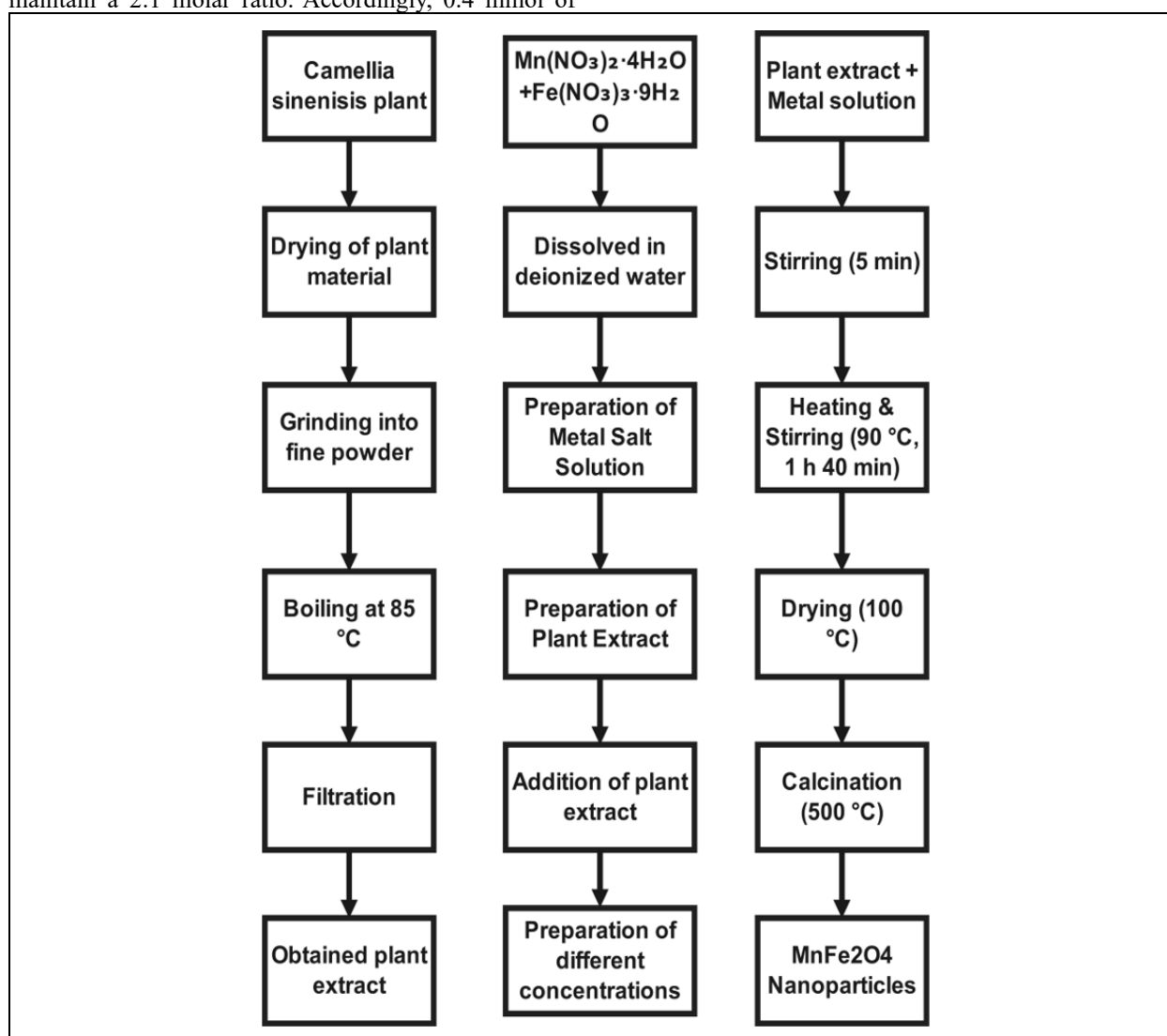


Fig. 1: Schematic illustration of the green synthesis process of MnFe₂O₄ NPs using *Camellia sinensis* extract

2.4 Characterization Methods

UV–Vis was used to determine phytochemical components in *C. sinensis* extract. The absorption spectra were measured using a Shimadzu UV-1800 UV–Vis spectrophotometer. Fourier transform infrared (FTIR) spectroscopy was employed to detect functional groups present in aqueous *C. sinensis* extract and confirm the formation of Fe–O and Mn–O bonds at synthesized MnFe₂O₄ NPs. FTIR spectra were recorded across a wavenumber range of 400–4000 cm⁻¹ using a Thermo Scientific Nicolet iS50 infrared spectrophotometer. The crystalline structure of the MnFe₂O₄ NPs was analyzed using X-ray diffraction (XRD). Diffraction patterns were collected using a Malvern Panalytical Empyrean X-ray diffractometer with CuK_α radiation ($\lambda = 1.54056 \text{ \AA}$), operating at 35 kV, over a 2θ scanning range of 10–80°. (Mahich *et al.* 2026). The morphological characteristics and elemental content of MnFe₂O₄ NPs were analyzed by a Zeiss Gemini field-emission scanning electron microscope. Selected area electron diffraction patterns were subsequently acquired to assess the phase composition and crystallinity of the synthesized samples. In addition, photoluminescence (PL) spectroscopy was performed to investigate the recombination dynamics. The PL spectra were recorded at ambient temperature using a Horiba Scientific Jobin Yvon Fluoromax-4 fluorometer with an excitation wavelength of 350 nm.

2.5 Phytochemical Analysis of *Camellia sinensis*

A preliminary qualitative analysis of the aqueous *C. sinensis* extract was conducted to identify the principal phytochemical elements present. Comprehensive descriptions of the experimental procedures and analytical methodologies are included in the supplemental material.

2.6 Photocatalytic Degradation Analysis Method

Photocatalytic degradation experiments were conducted utilizing 100 mL of 10 mg/L MB aqueous solution, with 10 mg of MnFe₂O₄ NPs serving as the catalyst. Before light irradiation, an adsorption–desorption equilibrium was achieved by dispersing the MnFe₂O₄ NPs in the MB solution and constantly stirring the suspension for 60 min in the dark at room temperature (Miu *et al.* 2024). Photocatalytic degradation experiments were performed in triplicate under direct sunlight for 120 min over a continuous 14-day duration. Solar irradiance and temperature were recorded daily at 12:00, 13:00, and 14:00 h, along with the overall fluctuations seen over the 14-day period. Throughout the trial duration, sunlight intensity fluctuated between 878 and 1022 W/m², and the room temperature ranged from 24.2 to 32.5 °C. The photocatalytic efficacy for the

breakdown of MB was assessed by extracting 0.5 mL samples at specified intervals. The residual dye concentration was measured with a UV–Vis spectrophotometer by assessing peak absorbance wavelength at 532 nm.

2.7 Stability and Reusability Assessment of Photocatalysts

The stability and recyclability of MnFe₂O₄ NPs were assessed over three consecutive photocatalytic degradation cycles. After each cycle, the photocatalysts were isolated from the reaction mixture by centrifugation at 4000 rpm for 12 min and washed several times with deionized water to remove residual dye. The recovered nanoparticles were then dried in an oven at 60 °C for 6 h to restore their photocatalytic activity before being reused under identical experimental conditions for the subsequent cycle. The degradation efficiency of MB was assessed after each cycle using UV–Vis spectroscopy. Additionally, potential structural and morphological alterations in the MnFe₂O₄ NPs were examined using FE-SEM and FTIR spectroscopy.

2.8 Scavenger Experiments for Reaction Analysis

The function of active species at the photocatalytic degradation of MB by MnFe₂O₄ NPs was examined by radical scavenging studies. This methodology used silver nitrate (AgNO₃) as an electron (e⁻) quencher, ethylenediaminetetraacetic acid disodium salt (EDTA-2Na) as a hole (h⁺) scavenger, p-benzoquinone (BQ) as a superoxide radical (O₂^{•-}) scavenger, and isopropyl alcohol (IPA) as a hydroxyl radical (•OH) scavenger. In each experiment, 10 mg of MnFe₂O₄ NPs and 1 mM were introduced into 100 mL of 10 mg/L MB. A resultant combination was subjected to natural sunlight for 100 min.

2.9 Phytotoxicity Assessment of Treated Water

Phytotoxicity evaluation was conducted using *Vigna radiata* seeds obtained from the local market in Erode, India. Before the experiment, the seeds were immersed in deionized water for 14 h to facilitate consistent germination. Subsequently, a total of 30 seeds were uniformly allocated on filter paper situated within three sterile Petri plates. Each dish was supplied with 10 mL of a specified solution: deionized water, photocatalytically treated water, or untreated dye-contaminated water. The Petri dishes were thereafter sealed and incubated at a temperature of 26.5 °C for 72 h to assess germination and growth responses (Shukla *et al.* 2025). All experiments were conducted in triplicate under identical conditions, and the results were reported as means to ensure reproducibility and reliability.

3. RESULTS AND DISCUSSION

3.1 UV–Vis and Phytochemical Evaluation Method

The findings of the phytochemical evaluation of aqueous *C. sinensis* extract are presented in Table 1. Qualitative investigation identified tannins, flavonoids, ethylenic chemicals, monosaccharides, and saponins in the extract, but alkaloids and sterols were absent.

Table 1. Results of Phytochemical screening of *C. sinensis* extract

S. No.	Test	Phytochemical Compounds	Outcomes
1	Barfoed	Monosaccharides	+
2	Mayer	Alkaloids	-
3	FeCl ₃	Tannins	+
4	Shinoda	Flavonoids with benzopyrone groups	+
5	Foam	Saponins	+
6	KMnO ₄	Ethylene compounds	+
7	Liebermann Buchard	Sterols	-

Fig. 2 displays the UV–Vis absorption spectra of the aqueous extracts obtained at several concentrations. Two distinct and pronounced absorption peaks are evident in the spectra. The initial peak is observed at a range of 239–237 nm, whilst the subsequent peak is situated between 347 and 370 nm. The unique absorption bands are ascribed to flavonoid chemicals. The band in the lower wavelength area corresponds to the A-ring benzoyl system of flavonoids, generally linked to shikimate-derived metabolites, and accounts for the bright yellow hue of the extract (Perera *et al.* 2020). A weak absorption band detected in the 294–305 nm range may be ascribed to phenolic compounds. The spectrum features indicate that the aqueous extract has both flavonoids and other phenolic chemicals. A significant alteration in the absorption maxima is noted, namely a transition from 347 nm to 237 nm, which correlates with a concentration-dependent color change of the extract. Moreover, a progressive decline in absorbance intensity is apparent when the extract concentration diminishes, thus validating the dilution effect on optical density.

3.2 FTIR Analysis

Fig. 3 displays the FTIR spectrum of the aqueous *C. sinensis* extract alongside the spectra of MnFe₂O₄ NPs synthesized using varying extract

concentrations. The extracted spectrum has multiple distinctive vibrational characteristics. A wide absorption band detected in the 3200–3450 cm⁻¹ range is attributed to stretching of hydroxyl (–OH) groups from adsorbed water molecules or alcohols, along with –NH₂ groups of primary amines from phytochemicals acting as reducing agents. The peaks at 2920 and 2850 cm⁻¹ correspond to asymmetric and symmetric stretching vibrations of C–H bonds in –CH₂– groups, confirming organic capping. The bands at 1630 and 1650 cm⁻¹ correspond to C=O stretching vibrations of carbonyl groups and aromatic bonds. The absorption at 1380 cm⁻¹ corresponds to the vibration of –O–NO₂ groups seen in organic nitrate. Additionally, bands seen at 1150 and 1030 cm⁻¹ correspond to C–O and C–C stretching vibrations typical of carboxylic acid groups, confirming the presence of bioactive compounds. Weak absorption bands are found in the lower wavenumber range below 600 cm⁻¹, likely due to inorganic compounds such as K, Ca, and Cl in the plant extract. The locations of the above vibrational bands are comparable to those documented in the literature (Vijayakumar *et al.* 2021), where Ag and Au NPs were synthesized using extracts from the *C. sinensis* plant.

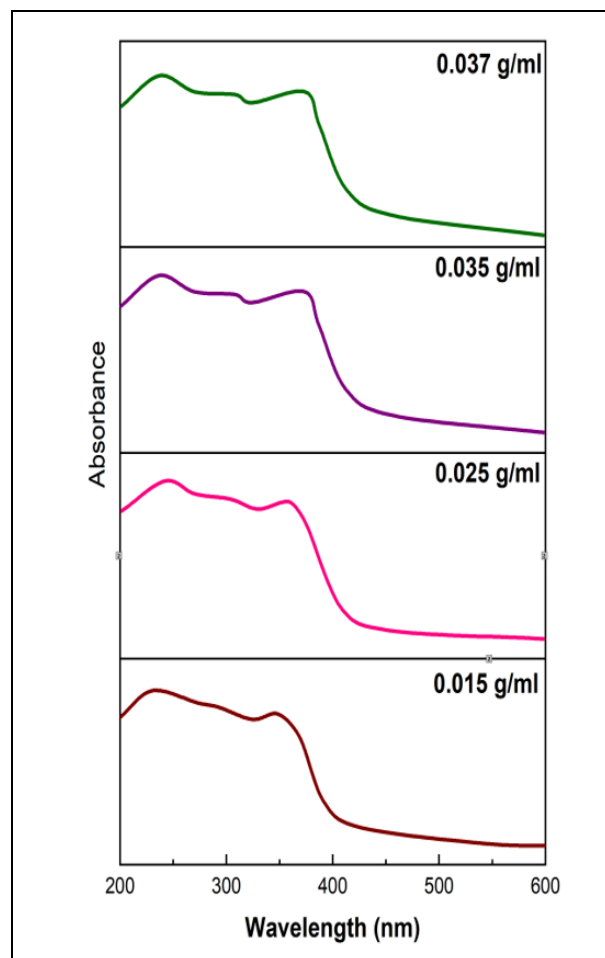


Fig. 2: Absorbance spectra of the aqueous extracts at various concentrations

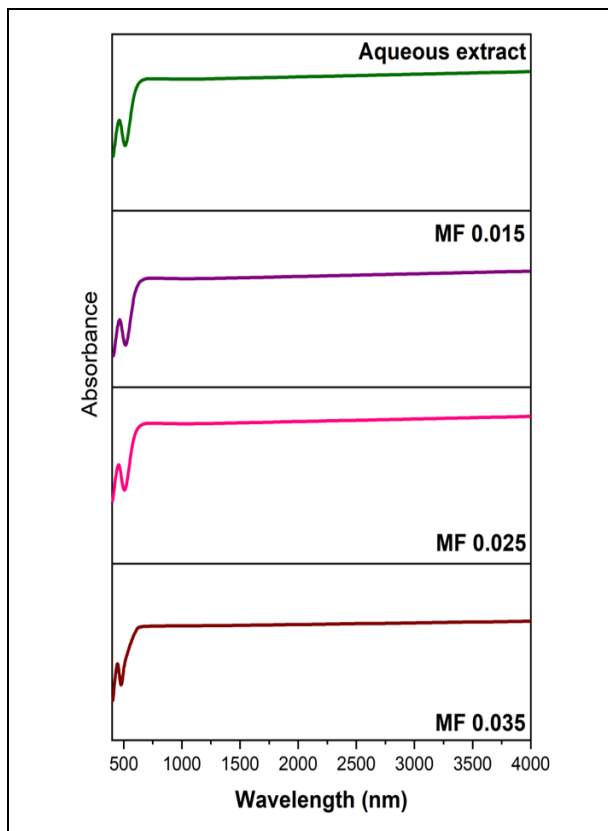


Fig. 3: FTIR spectra of the aqueous extract and MnFe_2O_4 NPs

The integrated results from phytochemical analysis, FTIR characterization, and UV–Vis spectroscopy indicate that tannins, flavonoids, monosaccharides, saponins, and ethylenic compounds are the principal bioactive components involved in the green synthesis of MnFe_2O_4 NPs. Flavonoids, tannins, and certain saponins possess phenolic functional groups that can reduce Mn^{2+} and Fe^{3+} ions, while also serving as stabilizing and capping agents to inhibit agglomeration of resultant nanoparticles.

Most importantly, the distinct bands at ~ 422 and $\sim 522 \text{ cm}^{-1}$ quantitatively confirm Mn–O and Fe–O vibrations at tetrahedral and octahedral sites of the spinel structure. The existence of these unique metal–oxygen vibrations verifies the establishment of the cubic spinel structure of MnFe_2O_4 . The lack of absorption bands above 600 cm^{-1} signifies that no leftover organic components from the plant extract were present at the nanoparticle surface.

3.3 XRD Analysis

Fig. 4 displays the X-ray diffraction patterns of MnFe_2O_4 NPs produced with varying amounts of plant extract. The diffraction profiles exhibit distinct peaks at approximately 29.9° , 35.4° , 43.1° , 53.3° , 56.9° , 62.4° , and 74.1° (2θ), which correspond to lattice planes, thereby affirming the cubic spinel phase of MnFe_2O_4 .

Additionally, Fig. 4 depicts the enhanced XRD fitting results derived from the structural analysis.

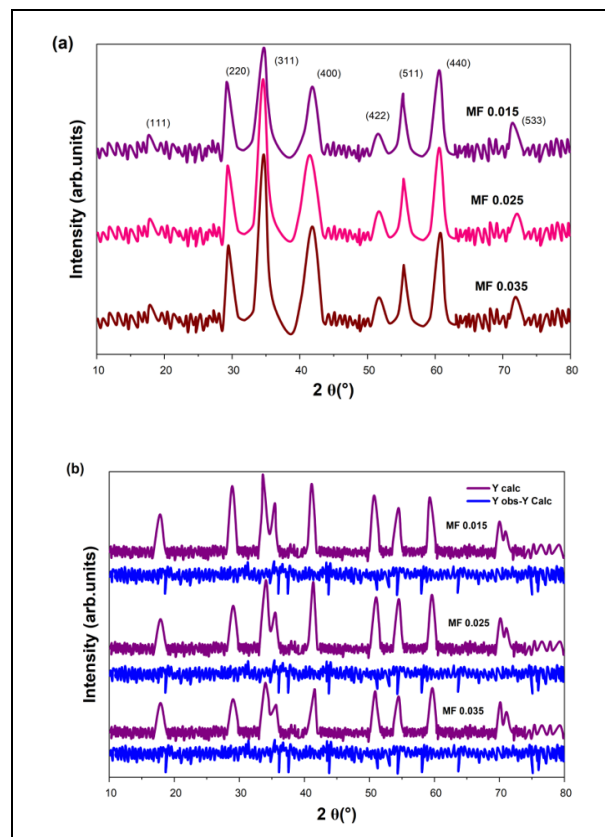


Fig. 4: (a) XRD patterns of MnFe_2O_4 NPs and (b) fitting results of XRD patterns

The crystallite size (CS) of the samples was ascertained by the Scherrer equation:

$$CS = \frac{K\lambda}{FWHM \cos \theta} \quad \dots (1)$$

λ signifies the X-ray wavelength, $FWHM$ refers to the full width at half maximum of the diffraction peak, K indicates the Scherrer constant or shape factor, and θ represents the Bragg diffraction angle. A lattice constant (a) of the cubic MnFe_2O_4 phase was determined using Bragg's law in conjunction with interplanar spacing (d) and the corresponding Miller indices:

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

The crystallite size calculated using the Scherrer equation yielded values of $9.68 \pm 1.01 \text{ nm}$ (MF0.015), $9.55 \pm 0.98 \text{ nm}$ (MF0.025), and $9.41 \pm 0.94 \text{ nm}$ (MF0.035). The calculated lattice constants (8.401 , 8.386 , and 8.372 Å) and unit cell volumes (592.98 , 589.62 , and 586.68 Å^3) show a slight decreasing trend with increasing extract concentration, indicating lattice contraction due to controlled nucleation. The reduction in crystallite size confirms enhanced capping efficiency of

phytochemicals, which limits crystal growth and improves structural uniformity.

3.4 SEM Analysis

Fig. 5 presents the FE-SEM micrographs (a) MF0.015, (b) MF0.025, and (c) MF0.035. SEM images distinctly illustrate the creation of agglomerated structures made up of tiny particles, primarily displaying an icosahedral morphology.

The nanoparticle morphology is uniform across samples; however, a steady reduction in particle size was

noted with increasing concentrations of plant extract. The size reduction can be primarily ascribed to improved steric hindrance effects. During the preliminary phase, phytochemical compounds in the extract facilitate the reduction of metal ions, resulting in the creation of tiny nucleation centers. Consequently, these bioactive molecules adhere to the surface of newly produced nuclei by their functional groups, resulting in a stabilizing organic capping layer. At elevated extract concentrations, augmented inclusion of voluminous organic compounds enhances the steric hindrance surrounding the developing crystals, thus limiting particle growth and leading to the formation of smaller nanoparticles.

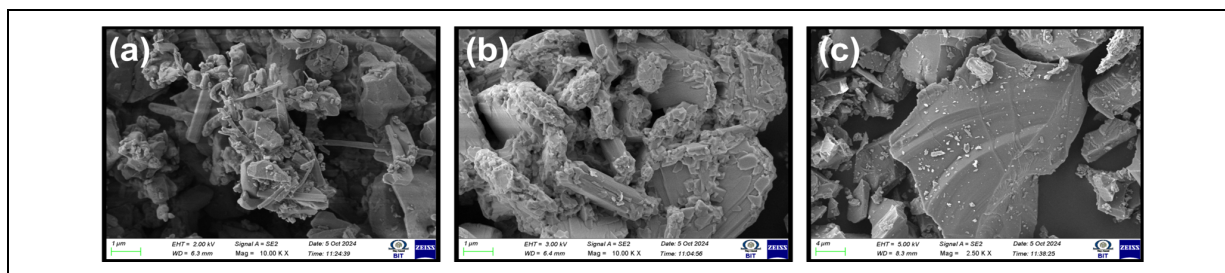


Fig. 5: FE-SEM images of (a) MF0.015, (b) MF0.025 and (c) MF0.035

The noted reduction in particle size aligns with the crystallite measurements established from XRD research. In contrast, the total agglomeration size escalates with an increase in extract concentration. This behavior can be ascribed to the increased concentration of phytochemical ingredients in the plant extract, which interact robustly with metal ions, thereby impeding their separation and altering the nucleation and growth mechanisms. Consequently, while smaller primary nanoparticles are initially generated, they tend to aggregate, resulting in the development of bigger agglomerated structures.

The lower extract concentrations and the restricted availability of phytochemicals limit selective surface adsorption, thereby reducing facet stability during particle formation. Consequently, the nucleation and growth processes become less regulated, promoting partial fusing of icosahedral nanoparticles at calcination

and resulting in bigger particles with diminished morphological uniformity.

SEM analysis revealed the formation of uniform nanoscale particles with predominantly icosahedral morphology, exhibiting varying degrees of agglomeration. The average particle size showed a decreasing trend with increasing extract concentration, consistent with XRD results, indicating effective size control through phytochemical-mediated stabilization. At lower extract concentrations, larger and less uniform particles were observed due to limited capping, whereas higher concentrations resulted in finer particles with improved uniformity due to enhanced steric hindrance effects. However, increased extract concentration also led to greater agglomeration size, attributed to strong interparticle interactions induced by organic residues. The correlation between SEM-observed particle size and XRD-derived crystallite size confirms structural refinement and validates the synthesis approach.

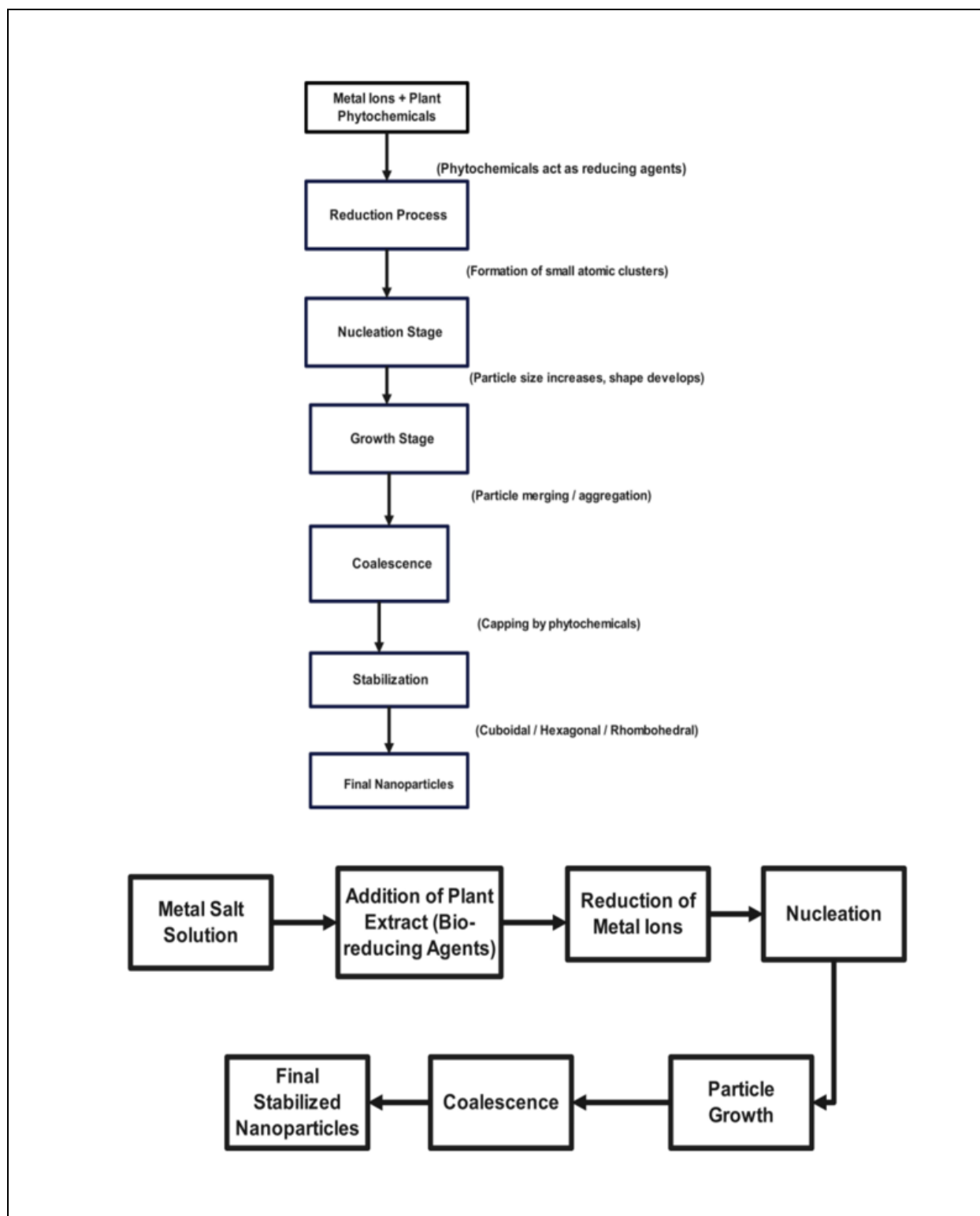


Fig. 6: Growth mechanisms of MF0.015, MF0.025, and MF0.035

3.5 Optical Properties

Fig. 7 displays UV–Vis absorption spectra of MnFe₂O₄ NPs. All produced samples exhibit substantial absorption in the visible spectra and ultraviolet, which enhances photocatalytic efficacy. The MF0.015 exhibits

two distinct absorption edges, located approximately in the ranges of 340–700 nm and 680–720 nm. The MF0.025 and MF0.035 samples have absorption edges at 350–650 nm and 650–700 nm, respectively. The absorption patterns are ascribed to excitonic transitions in the MnFe₂O₄ nanostructures.

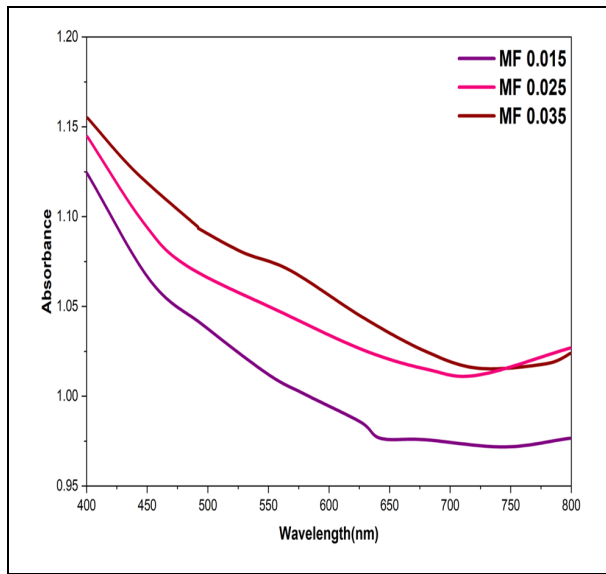


Fig. 7: Absorbance spectra of the MnFe₂O₄ NP samples

The MF0.035 sample, primarily consisting of evenly formed icosahedral nanoparticles, demonstrated the widest absorption range in the visible spectrum. The improved optical response is due to the symmetric structure of particles, which enables the most effective light interaction by greater scattering and absorption mechanisms. Conversely, MF0.015 and MF0.025 samples exhibit heterogeneous amalgamation of hexagonal, rhomboidal, icosahedral, and cuboidal morphologies, which engenders varied light-matter interaction pathways that collectively yield relatively diminished optical sensitivity and a constricted visible-light absorption spectrum.

The UV-Vis absorption data of MnFe₂O₄ NPs, along with the relevant theoretical relations, were utilized to ascertain several optical parameters (Table 2), including optical bandgap (E_g), absorption coefficient (α), extinction coefficient (k), refractive index (n), and the imaginary (ϵ_i) and real (ϵ_r) components of dielectric constant, as well as optical conductivity (σ). Except for the bandgap energy, the values of these optical constants were derived at a photon energy of 2 eV from absorption spectra.

The absorption coefficient (α) of the MnFe₂O₄ NPs was obtained from the UV-Vis absorbance data using the Beer-Lambert law.

$$\alpha = 2.303 \left(\frac{A}{l} \right) \quad \dots (3)$$

In this expression, A represents the observed absorbance, and l signifies the optical path length (sample thickness). Fig. 8 illustrates the absorption coefficient (α) spectra of the MnFe₂O₄ NPs. An elevation in α is noted for all samples within the photon energy spectrum of 1.5 to 4.5 eV. The MF0.035 demonstrates the

highest absorption coefficient, signifying its superior light-harvesting capabilities relative to the other compositions. Comparatively with the previous study, biochar derived from *Citrus limetta* peels embedded with green tea-synthesized Fe₂O₃ nanoparticles formed a BC@Fe₂O₃ nanohybrid for sunlight-driven degradation of endosulfan and ethion pesticides. High removal efficiency was attributed to enhanced surface area, favorable zeta potential (4.5 mV), low bandgap (1.6–1.8 eV), first-order kinetics, and strong reusability up to seven cycles (Rani *et al.* 2023).

Table 2. Optical constants of MnFe₂O₄ NP samples

Optical Constants	Samples		
	MF0.015	MF0.025	MF0.035
α (cm ⁻¹)	2.21	2.33	2.46
$E_{0.75}$ (eV)	1.25	1.18	1.12
E_{CB} (eV)	-0.18	-0.24	-0.30
E_g (eV)	2.3	2.18	2.05
n	1.29	1.25	1.21
k	1.08×10^{-5}	1.15×10^{-5}	1.23×10^{-5}
ϵ_r	1.66	1.57	1.47
ϵ_i	2.70×10^{-5}	2.85×10^{-5}	3.02×10^{-5}
σ (s ⁻¹)	6.75×10^9	6.98×10^9	7.22×10^9

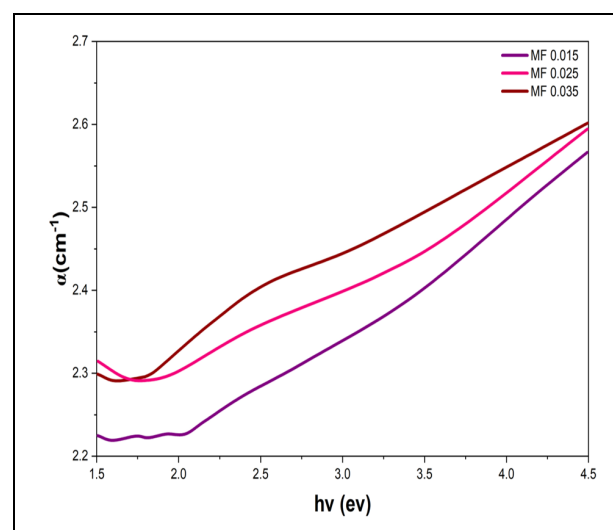


Fig. 8: Absorption coefficient (α) analysis

Optical bandgap (E_g) of MnFe_2O_4 NPs was determined utilizing Wood and Tauc connection, presuming allowed direct electronic transition and light absorption characteristics.

$$(ah\nu)^n \propto A(h\nu - E_g) \quad \dots (4)$$

where A denotes a proportionality constant, $h\nu$ signifies photon energy, and n represents an exponent contingent upon the characteristics of the electronic transition ($n = 2$).

Fig. 9 illustrates the graph of $(ah\nu)^2$ as a function of $h\nu$ for MnFe_2O_4 NPs. A progressive drop in bandgap energy (E_g) is noted with increasing concentrations of plant extract, potentially linked to the decrease in crystallite size, corroborated by morphological investigations and XRD.

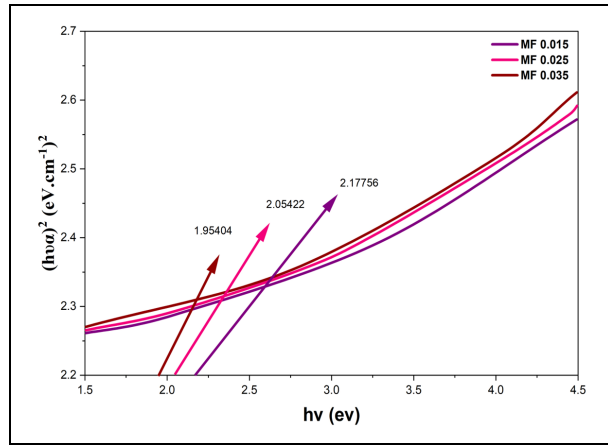


Fig. 9: Tauc plot of $(ah\nu)^2$ vs. photon energy ($h\nu$) for MnFe_2O_4 NPs, indicating optical bandgap variation

The reduction in bandgap enhances visible-light absorption and reduces electron-hole recombination, thereby improving photocatalytic efficiency. The electronic band structure of MnFe_2O_4 samples was assessed by determining the positions of the conduction band edge and valence band edge.

$$E_{VB} = \chi - E_e + 1/2E_{g\psi} \quad \dots (5)$$

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

In this context, χ denotes absolute electronegativity of the semiconductor, quantified as 5.03 eV for MnFe_2O_4 , whereas E_e signifies energy of free electrons, measured at 4.4 eV relative to Normal Hydrogen Electrode (NHE).

The refractive index (n) was calculated to understand light propagation characteristics within the material.

$$n = KE_g^C \quad \dots (7)$$

K and C were material-specific constants, valued at 3.3668 and -0.32234 , respectively. Fig. 10 displays the refractive index (n) spectra of the MnFe_2O_4 NPs. A progressive reduction in n is noted for all samples when energy escalates from 0 to 3.5 eV. Among the samples, MF0.015 demonstrates the highest refractive index, signifying reduced light propagation speed and increased optical deviation within the material, attributable to its comparatively larger particle size and elevated packing density. Moreover, the refractive index values acquired for all samples are inferior to those previously documented for MnFe_2O_4 thin films and nanoparticle systems.

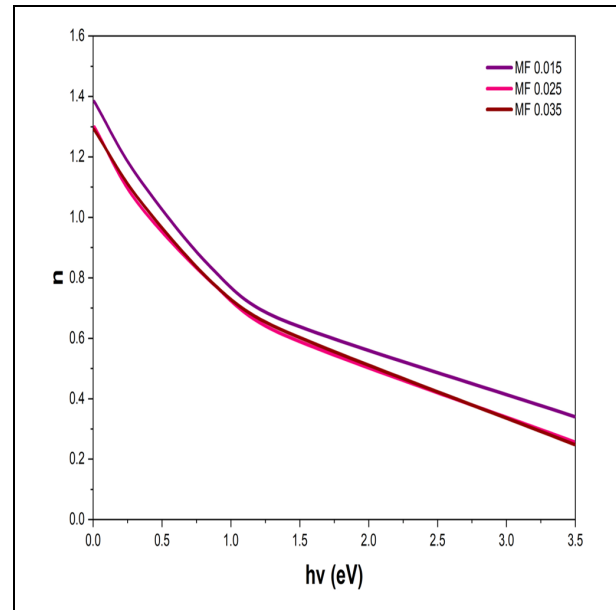


Fig. 10: Variation of refractive index (n) with photon energy for MnFe_2O_4 NP samples

The extinction coefficient (k), a dimensionless parameter, was determined to evaluate light attenuation within the nanoparticles. The k spectra of MnFe_2O_4 NPs were delineated using Eq. (8):

$$k = \frac{a\lambda}{4\pi} \quad \dots (8)$$

where λ denotes the wavelength. Fig. 11 illustrates k spectra of MnFe_2O_4 NPs. Each spectrum demonstrates a declining k trend as photon energy rises from 0 to 3.5 eV. MF0.035 exhibited the greatest k value, signifying its superior light absorption compared to the other samples.

The dielectric constants (ϵ_r and ϵ_i) were derived from n and k to describe the polarization and energy loss behavior of the material:

$$\epsilon_r = n^2 - k^2 \quad \dots (9)$$

$$\epsilon_i = 2n \quad \dots (10)$$

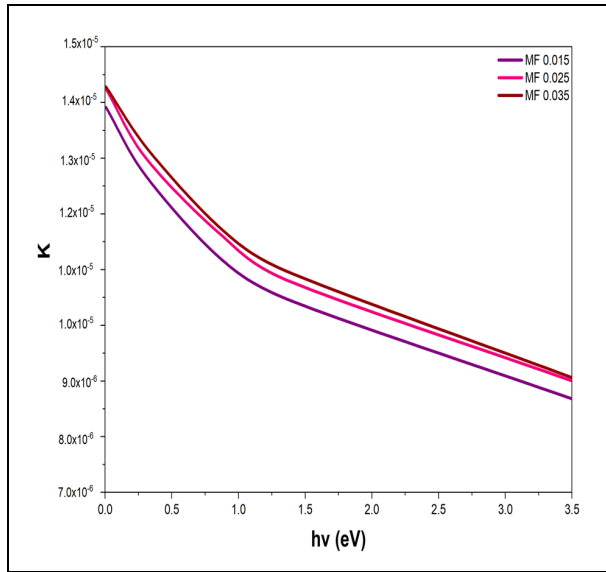


Fig. 11: Variation of extinction coefficient (k) with photon energy ($h\nu$) for MnFe_2O_4 NP samples

Figs. 12 (a–b) illustrate the spectra of ϵ_r and ϵ_i dielectric constants of MnFe_2O_4 NPs. ϵ_r is associated with the refractive index n , while ϵ_i pertains to the extinction coefficient k . The ϵ_r spectra have a trend analogous to that of n . Conversely, ϵ_i spectra exhibit a pattern analogous to that of k . The MF0.015 had an elevated ϵ_r value, signifying enhanced polarizability. Conversely, the MF0.035 sample demonstrated an elevated ϵ_i value, attributed to improved light absorption.

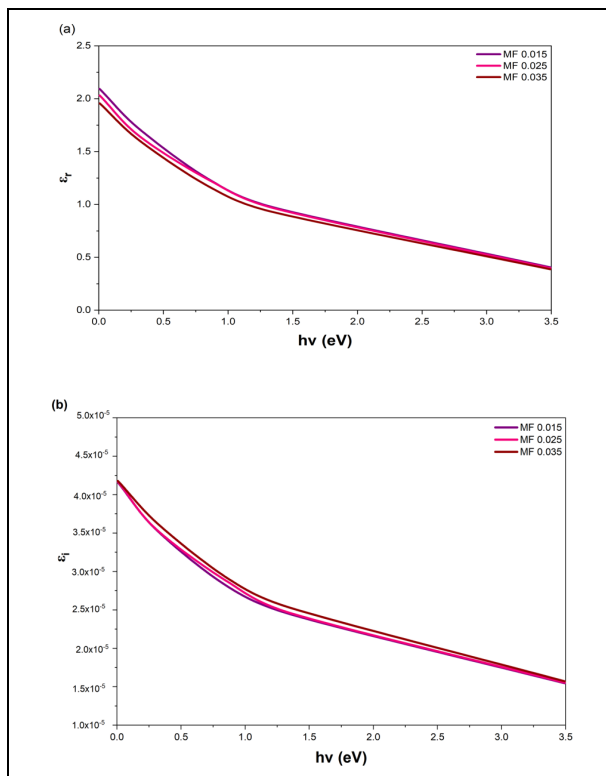


Fig. 12: (a) Real (ϵ_r) and (b) imaginary (ϵ_i) constants

The optical conductivity (σ) was calculated to assess the interaction of the material with electromagnetic radiation:

$$\sigma = \frac{anc}{4\pi} \quad \dots (11)$$

where c is the speed of light. Fig. 13 illustrates σ spectra of MnFe_2O_4 NPs. A reduction in σ was noted across each spectrum, varying from 0 to 3.5 eV. A maximum σ value was documented at sample MF0.035.

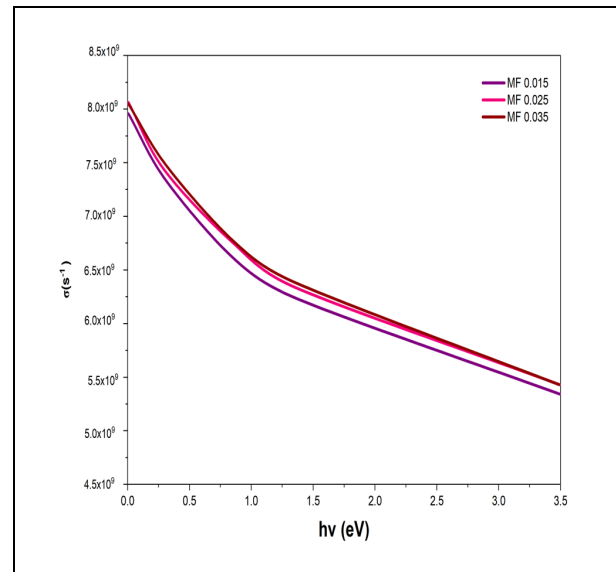


Fig. 13: Optical conductivity analysis

Fig. 14 illustrates photoluminescence spectra of MnFe_2O_4 NPs. Two primary shoulder and emission peaks were detected. The initial peak is observed between 400 and 600 nm, whereas the subsequent peak is found between 470 and 480 nm. The shoulder is observed within a wavelength range of 500–510 nm. The initial peak is linked to violet emission, while the subsequent peak pertains to blue emission associated with intrinsic area, including zinc interstitial deficiencies. The shoulder was a blue-green emission resulting from oxygen vacancies. The MF0.015 sample has the highest photoluminescence emission intensity, signifying an elevated electron-hole recombination. Conversely, the MF0.035 sample exhibits the lowest PL intensity, indicating more efficient inhibition of electron-hole recombination.

Among all optical parameters, the bandgap and absorption behavior were found to be the most significant factors governing photocatalytic performance.

3.6 Photocatalytic Activity

Fig. 15 illustrates alterations at UV–Vis absorption spectra of MB because of photocatalytic degradation at solar irradiation, (a) MF0.015, (b) MF0.025, and (c) MF0.035, which correspond to the first

replicate. A progressive reduction in intensity spectra was noted at extended irradiation time.

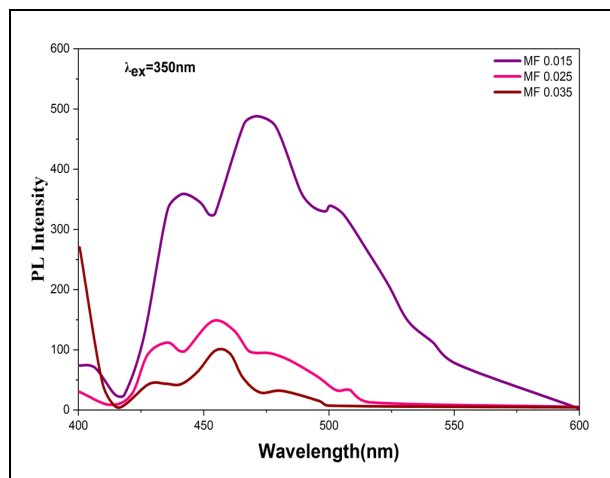


Fig. 14: PL spectra of MnFe₂O₄ NP samples

Fig. 15(d) presents the FTIR spectra of MB in aqueous solution after 130 min of solar irradiation in the presence of different samples, showing the degradation of MB in all cases. MB exhibited multiple characteristic vibrational bands within the 400–4000 cm⁻¹ region. An extensive band among 937.7 and 999.7 cm⁻¹ was ascribed to O–H and N–H, corresponding to aliphatic primary hydroxyl, water, and amine groups. Bands at 1144 and 1273 cm⁻¹ correspond to the C–H stretching of methyl groups. The absorption bands at 1356 and 1743 cm⁻¹ were attributed to C–O and C=O stretching vibrations of oxygen-containing functional groups generated during the photocatalytic degradation of MB molecules. The weak absorption band observed at 1795 cm⁻¹ corresponds to carbonyl (C=O) stretching vibrations. The prominent band at 2167 cm⁻¹ was assigned to C≡C stretching vibrations associated with intermediate degradation products. Band at 2203 cm⁻¹ corresponds to C–N vibrations, while the bands at 2203 and 2466 cm⁻¹ were attributed to C–N stretching vibrations of aliphatic and tertiary amines.

The degraded samples exhibited a significant decrease in the intensity of characteristic bands compared to the crystal violet reference in the spectra of the samples. The large peaks at 937, 999, 1144, 1273, and 1356 cm⁻¹ became slowly smaller in MF0.015 and MF 0.025 and strongly suppressed in MF0.035. On the same note, the bands at 1743 and 1795 cm⁻¹, as well as those at 2167 and 2203 cm⁻¹, showed significant weakening upon irradiation. The high absorption band around 2466 cm⁻¹ was also significantly reduced, especially with MF0.035. The overall reduction and attenuation of these characteristic vibrational peaks confirm the successful degradation of the dye molecules, with MF0.035 exhibiting the highest photocatalytic activity.

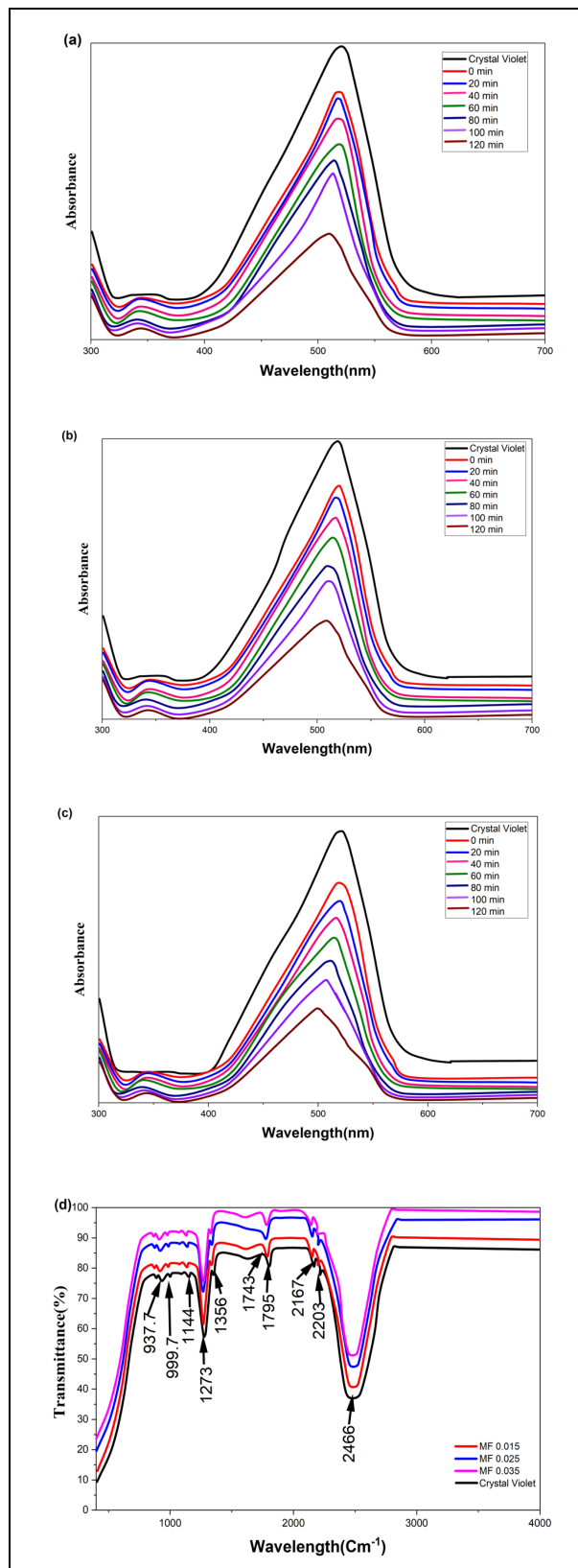


Fig. 15: (a–c) UV–Vis spectra of MB in the presence of MF0.015, MF0.025, and MF0.035, respectively, and (d) FTIR spectra of MB degradation

Fig. 16 illustrates the photocatalytic degradation (C_t/C_0) curves of MB in relation to solar irradiation duration, utilizing MnFe_2O_4 NPs as photocatalysts. The effectiveness of the degradation percentage of MnFe_2O_4 NPs was assessed using:

$$\text{Degradation efficiency (\%)} = \frac{C_0 - C_t}{C_0} \times 100 \quad \dots (12)$$

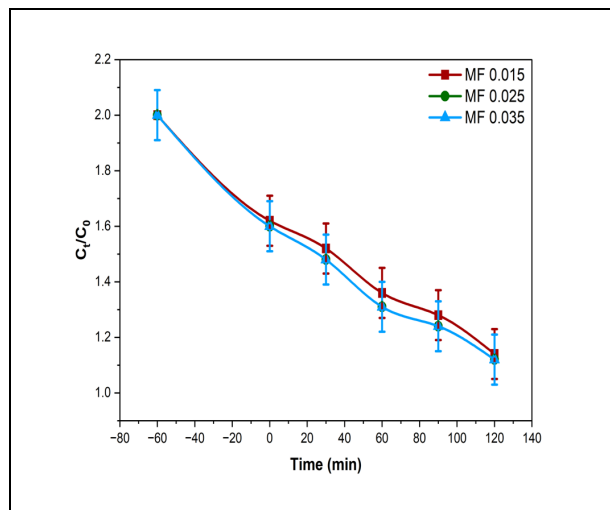


Fig. 16: Photocatalytic degradation profile of MB using MnFe_2O_4 NP samples

C_0 and C_t denote initial and ultimate concentrations of MB before and after light irradiation. After 120 min of solar irradiation, the MB degrading efficiencies were $43.0 \pm 2.0 \%$, $44.0 \pm 1.8 \%$, and $44.5 \pm 2.1 \%$ for the MF0.015, MF0.025, and MF0.035 samples, respectively. To better contextualize the photocatalytic performance, the obtained degradation efficiencies were compared with previous studies. Although the present system achieved a moderate degradation efficiency ($\sim 44.5\%$) under natural sunlight, several studies have reported higher efficiencies under optimized or modified conditions. For instance, $\text{MnFe}_2\text{O}_4/\text{CFA}$ composites demonstrated $\sim 99.9\%$ degradation under controlled conditions (Mushtaq *et al.* 2020). In a previous study, green biosynthesized Al-doped MnO nanoparticles using *Amomum longiligulare* fruit extract exhibited UV-light-driven degradation of MB (66%) and malachite green (38.1%) within 60 min, following first-order kinetics (Liu *et al.* 2020).

The degradation rate of MB was assessed based on pseudo-first-order kinetics, and the rate constant values (k) were calculated by analyzing the slope of the $\ln(C_0/C_t)$ against time irradiation plot using (Mehenaoui *et al.* 2024):

$$\ln \frac{C_0}{C_t} = kt \quad \dots (13)$$

Fig. 17 illustrates graphs of $\ln(C_0/C_t)$ against irradiation time for MnFe_2O_4 NPs during the degradation

of MB. Kinetic rates (k) for MF0.015, MF0.025, and MF0.035 were $1.15 \times 10^{-2} \pm 0.001$, $1.22 \times 10^{-2} \pm 0.002$, and $1.45 \times 10^{-2} \pm 0.002 \text{ min}^{-1}$, respectively.

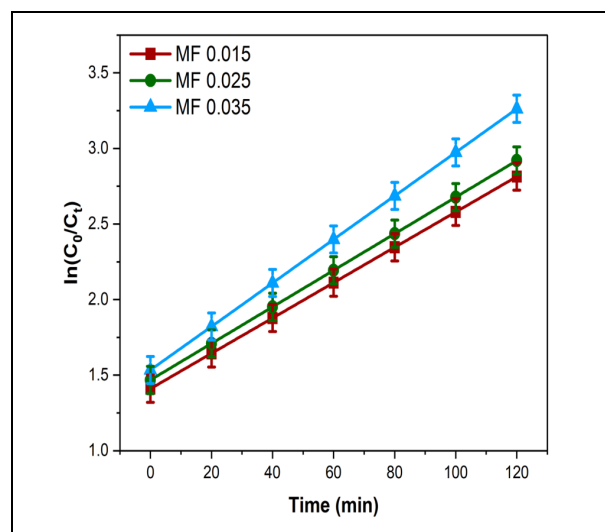


Fig. 17: $\ln(C_0/C_t)$ vs. irradiation time

3.7 Evaluation of Stability and Reusability

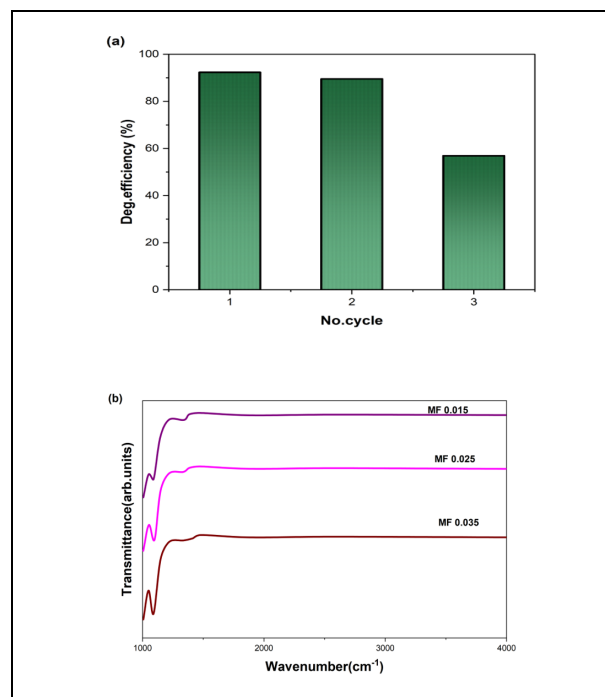


Fig. 18: (a) Photocatalytic cycling performance of MF0.035 for MB degradation and (b) FTIR spectra of MF0.015, MF0.025, and MF0.035 samples

Fig. 18 illustrates the photocatalytic cycling performance of MF0.035 for MB degradation. Following the third cycle, degradation performance diminished by merely 35%, signifying a negligible decline in photocatalytic efficacy and excellent recyclability. The spectra distinctly exhibit bands linked to Fe–O and Mn–

O vibrations, typical of MnFe_2O_4 , indicating no alterations in the material's area. Signals over 800 cm^{-1} were significantly associated with materials derived from dye residues.

3.8 Radical Scavenger Evaluation

Fig. 19 illustrates the degrading performance of MB using the MF0.035 sample with different scavengers. The maximum efficiency of 92% was attained without the presence of any scavengers (Tran *et al.* 2025). The addition of AgNO_3 resulted in a modest fall in efficiency to 87%, indicating that electrons play a marginal role at degrading process. The introduction of EDTA-2Na (disodium ethylenediaminetetraacetate) resulted in a notable decrease in efficiency, declining to 76%. Isopropyl alcohol (IPA) subsequently reduced the efficiency to 73%. The most significant decrease was observed in the presence of benzoquinone (BQ), with the degradation efficiency declining to 69%. The data suggest h^+ functions as a secondary oxidizing agent, demonstrated by the mild influence of EDTA-2Na.

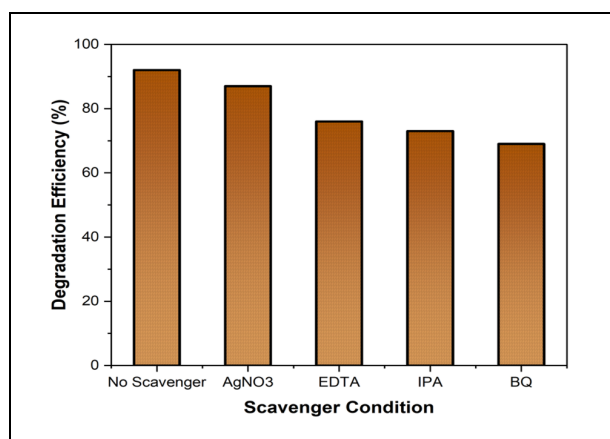


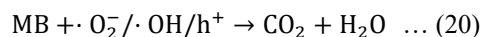
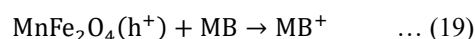
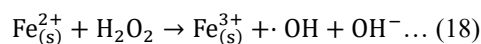
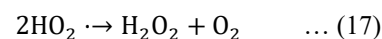
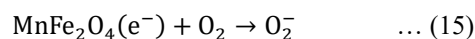
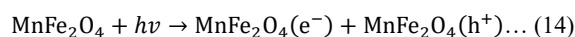
Fig. 19: Photocatalytic degradation for MB using MF0.035 with different scavengers

3.9 Proposed Photocatalytic Reaction Pathway Mechanism

The photocatalytic degradation pathway of MB using manganese ferrite nanoparticles involves the excitation of electrons from the valence band to the conduction band under solar-light irradiation, thereby generating electron-hole pairs that participate in the degradation process. Specifically, electrons in the conduction band convert molecular oxygen adsorbed on the surface of nanoparticles to superoxide radicals. This transpires due to conduction band potentials of MnFe_2O_4 NPs (-0.22 , -0.11 , and -0.22 eV for the MF0.015, MF0.025, and MF0.035, respectively) being negative redox. Consequently, O_2 emerges as the principal reactive species in the photocatalytic degradation of MB dye.

In situ-generated H_2O_2 was activated at the surface of MnFe_2O_4 Fe^{2+} by heterogeneous reaction, resulting in the production of reactive $\cdot\text{OH}$ radicals. The $\cdot\text{OH}$ radicals acted as secondary reactive species participating in the photocatalytic breakdown of MB dye. Direct generation of $\cdot\text{OH}$ from H_2O was not feasible in this system because the valence band (VB) potentials of the MnFe_2O_4 NPs were lower than the oxidation potentials required for $\text{OH}/\cdot\text{OH}$ (1.99 eV) and $\text{H}_2\text{O}/\cdot\text{OH}$ (2.38 eV).

Concurrently, h^+ at VB were oxidized by MB, augmenting the overall oxidative capability of the mechanism. The synergistic effects of $\cdot\text{OH}$ radicals, h^+ , and $\text{O}_2^{\cdot-}$ facilitate the efficient photocatalytic degradation of MB into non-toxic byproducts, primarily H_2O and CO_2 .



Under sunlight irradiation, MnFe_2O_4 NPs generate charge carriers, where excited electrons interact with dissolved oxygen to produce superoxide radicals ($\text{O}_2^{\cdot-}$), while holes contribute to oxidation reactions. These reactive species, along with hydroxyl radicals ($\cdot\text{OH}$) formed during the process, collectively drive the degradation of MB into harmless end products. Thus, the photocatalytic activity is primarily governed by effective charge separation and reactive oxygen species generation.

3.10 Phytotoxicity Evaluation

The percentage of seed germination was determined using (Ssekatawa *et al.* 2022):

$$\text{Seed germination (\%)} = \frac{\text{Seed germinated}}{\text{Total seed germinated}} \times 100 \dots (21)$$

Fig. 20 illustrates the percentage of seed germination across three treatments: control, treated water, and untreated water. The control treatment exhibited huge germination of $97.8 \pm 5.2\%$, followed by treated water ($89.6 \pm 5.4\%$). The lowest value ($79.8 \pm 5.3\%$) was associated with untreated water.

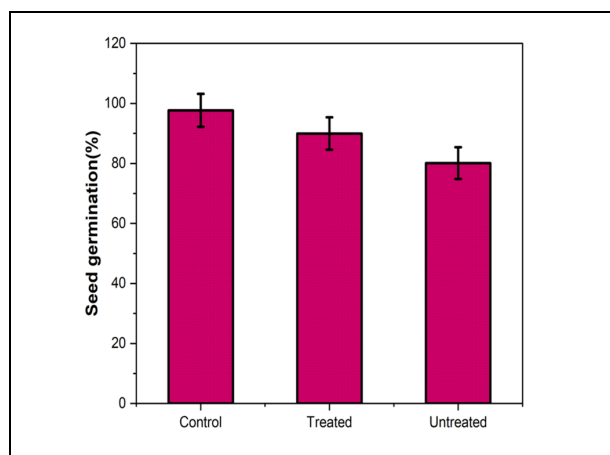


Fig. 20: Percentage seed growth response

Fig. 21 illustrates mean root lengths for identical treatments. The control treatment exhibited the largest length of root, measuring 3.02 ± 0.15 cm. Untreated water produced an average length of 2.69 ± 0.12 cm, while treated water yielded a length of 2.39 ± 0.11 cm. Ultimately, these findings clearly corroborate the results, indicating that the most extensive root development occurred in seeds from the control treatment.

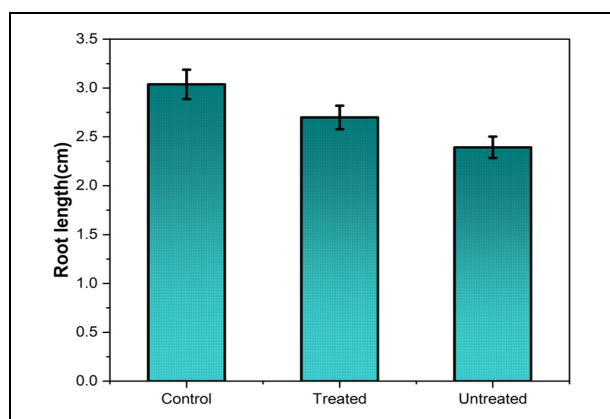


Fig. 21: Mean root length variation

The phytotoxicity results demonstrate that the treated water exhibits significantly lower toxicity compared to the untreated dye solution, as evidenced by improved seed germination and root growth of *Vigna radiata*. This indicates that the photocatalytic process effectively reduces harmful contaminants and enhances water quality. From an environmental perspective, these findings suggest that the treated water can be safely reused for agricultural irrigation and green space applications, thereby reducing freshwater demand and supporting sustainable water management practices.

4. CONCLUSION

MnFe₂O₄ nanoparticles were successfully synthesized through a green co-precipitation method using *Camellia sinensis* extract as a reducing and

stabilizing agent. Structural analysis confirmed the formation of single-phase cubic spinel nanoparticles with crystallite sizes ranging from 9.41 to 9.68 nm, indicating effective control over particle growth. Optical studies revealed a significant reduction in bandgap from 2.33 eV to 1.11 eV with increasing extract concentration, enhancing visible-light absorption. The synthesized nanoparticles demonstrated stable photocatalytic performance under natural sunlight, achieving a maximum degradation efficiency of approximately 44.5% for MB. Kinetic analysis confirmed pseudo-first-order behavior, while scavenger studies identified superoxide (O₂^{•-}) and hydroxyl (•OH) radicals as the dominant reactive species governing the degradation process. Furthermore, phytotoxicity evaluation using *Vigna radiata* confirmed reduced toxicity in treated water compared to untreated dye solution, indicating its suitability for reuse. Overall, this study highlights the potential of green-synthesized MnFe₂O₄ nanoparticles as cost-effective and environmentally sustainable photocatalysts for wastewater remediation and the safe reuse of water for agricultural applications.

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CONFLICTS OF INTEREST

The authors declare that there is no conflict of interest.

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