

Antimicrobial magnetic *Glycyrrhiza glabra* nanocomposite for decolouration of water through adsorption and photodegradation

Ankita Manchanda¹, Ahmed Hussain Jawhari², Ziaul Hasan³, Nazim Hasan^{2,4}, Sneha Shukla¹, Adiba Khan¹, Tabrez Alam Khan¹, Saif Ali Chaudhry^{1*}

¹Department of Chemistry, Jamia Millia Islamia, New Delhi-110025, India

²Department of Physical Sciences, Chemistry Division, College of Science, Jazan University, P.O. Box. 114, Jazan-45142, Kingdom of Saudi Arabia

³Department of Biosciences, Jamia Millia Islamia, New Delhi-110025, India

⁴Nanotechnology Research Unit, Jazan University, P.O. Box 114, Jazan 45142, Saudi Arabia
Corresponding E-mail: schaudhry@jmi.ac.in (Saif A Chaudhry)

Supplementary Information

Debye-Scherrer equation

The estimation of average crystallite grain size (D, nm) of GG/γ-Fe₂O₃ NPs from the full width at half maximum (FWHM) using Debye-Scherrer equation was performed using the given equation:

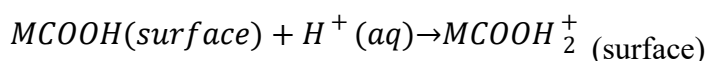
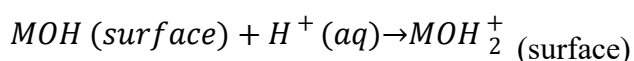
$$D = \frac{K \lambda}{\beta \cos \theta} \quad (S1)$$

where, λ is wavelength of incident X-ray radiation (λ = 0.154 nm), K is the Scherrer constant (=0.9 for cubic shape), θ is Bragg's angle of diffraction (radians), while β corresponds to FWHM (radians).

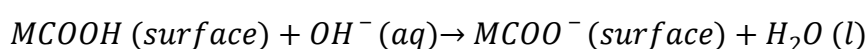
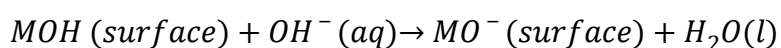
Zero-point charge of GG/γ-Fe₂O₃

The amount and nature of charge acquired by the solid surface directly correlates to the presence of H⁺ and OH⁻ in the aqueous dispersion phase, which either protonate or deprotonate the surface functional groups (MOH/MCOOH). This amphoteric behaviour of functional groups paves way for the development of either positive or negative charge on the solid surface above and below the neutral zero-point charge, ZPC condition as indicated below.

Protonation at low pH:



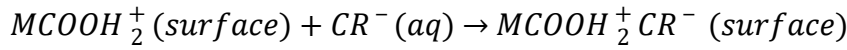
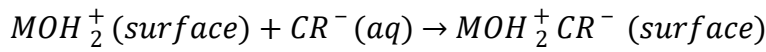
Deprotonation at high pH:



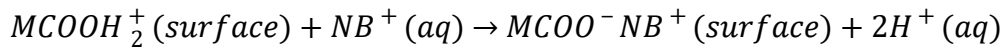
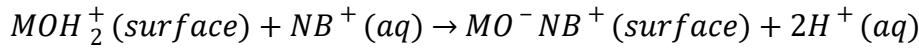
At ZPC, there is an equal concentration of both MOH₂⁺ / MCOOH₂⁺ and MO⁻ / MCOO⁻ species on the GG/γ-Fe₂O₃ surface, hence the total net electric charge density equilibrates to zero. During adsorption, the ZPC controls the ionic interaction between the surface functional active

sites and cationic (NB) and anionic (CR) dyes that adhere onto these sites, regulating the effect of change in pH of aqueous solution on the latter, as shown below.

For anionic dye Congo red (CR):



For cationic dye Nile blue (NB):



For GG/ γ -Fe₂O₃ surface, ZPC was found to be 6.9 (Fig. S5), and above and below pH 6.9, the surface acquired charge.

Thermodynamics of adsorption process

The adsorption data obtained at 30, 40, and 50 °C was substituted in the below stated

thermodynamic equations to compute ΔG [Eq. S2], ΔH (slope of $\log\left(\frac{Q_e}{C_e}\right)$ vs. $1/T$ plot), and

ΔS (intercept of $\log\left(\frac{Q_e}{C_e}\right)$ vs. $1/T$ plot) [Eq. S3].

$$\Delta G = -RT \ln K_C \quad (S2)$$

$$\log\left(\frac{Q_e}{C_e}\right) = \frac{\Delta S}{2.303R} - \frac{\Delta H}{2.303RT} \quad (S3)$$

where T is absolute temperature, $K_c (= Q_e/C_e)$, is the equilibrium constant, and R (= 0.008314 kJmol⁻¹K⁻¹) is the universal gas constant.

Webber-Morris Intraparticle diffusion (IPD)

The solute uptake is in proportion with time and expressed as:

$$Q_t = k_{IPD}t^{0.5} + C \quad (S4)$$

The equation produces a linear plot between Q_t and $t^{0.5}$, the slope of which determines the IPD rate constant, k_{IPD} (mgg⁻¹min^{-0.5}) (Table S6). If the experimental data fits well in the above equation producing a straight-line plot passing through origin, then IPD would be the rate limiting step, otherwise the kinetics is governed by the film diffusion model. However, intercept, C, incorporates partial film diffusion driven rate kinetics since both processes might occur simultaneously. Moreover, the intercept value provides a fair idea of the thickness of the boundary layer.

Boyd's-Liquid film diffusion (LFD)

$$- \ln (1 - F) = k_{LFD} t \quad (S5)$$

where, k_{LFD} (min^{-1}) is the film diffusion rate constant, while F ($= Q_t/Q_e$) depicts the fraction of pollutant ions adsorbed time t . If on plotting $-\ln(1-F)$ against t , a straight line is obtained with slope k_{LFD} that passes through origin, then the adsorption kinetics obey the LFD model.

Supplementary Figures:

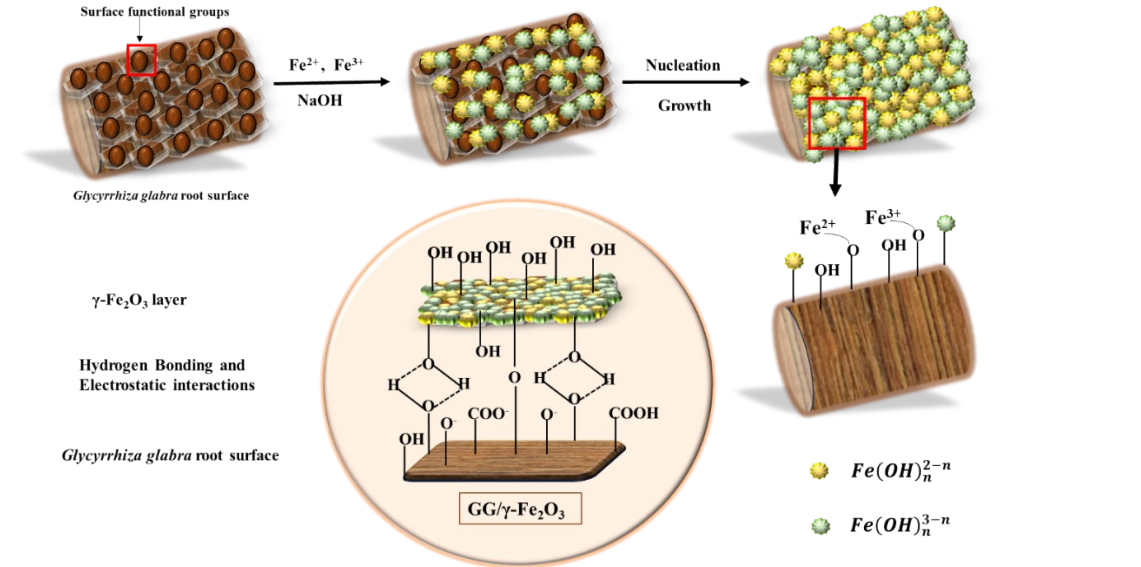


Fig. S1: Pictorial representation of interactions leading to the formation of GG/ $\gamma\text{-Fe}_2\text{O}_3$

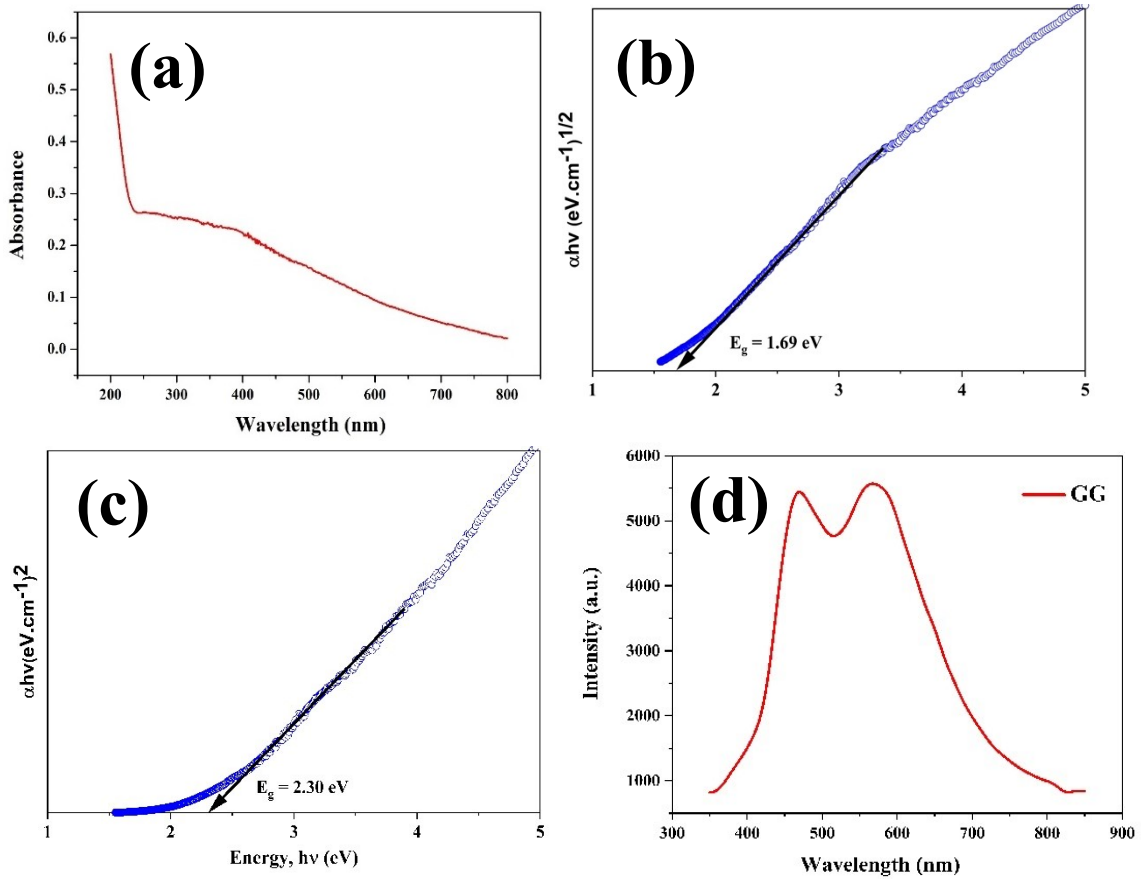


Fig. S2: (a) UV-Visible; (b) Indirect, and (c) Direct Tauc plot of GG/ γ -Fe₂O₃; (d) PL spectra of GG

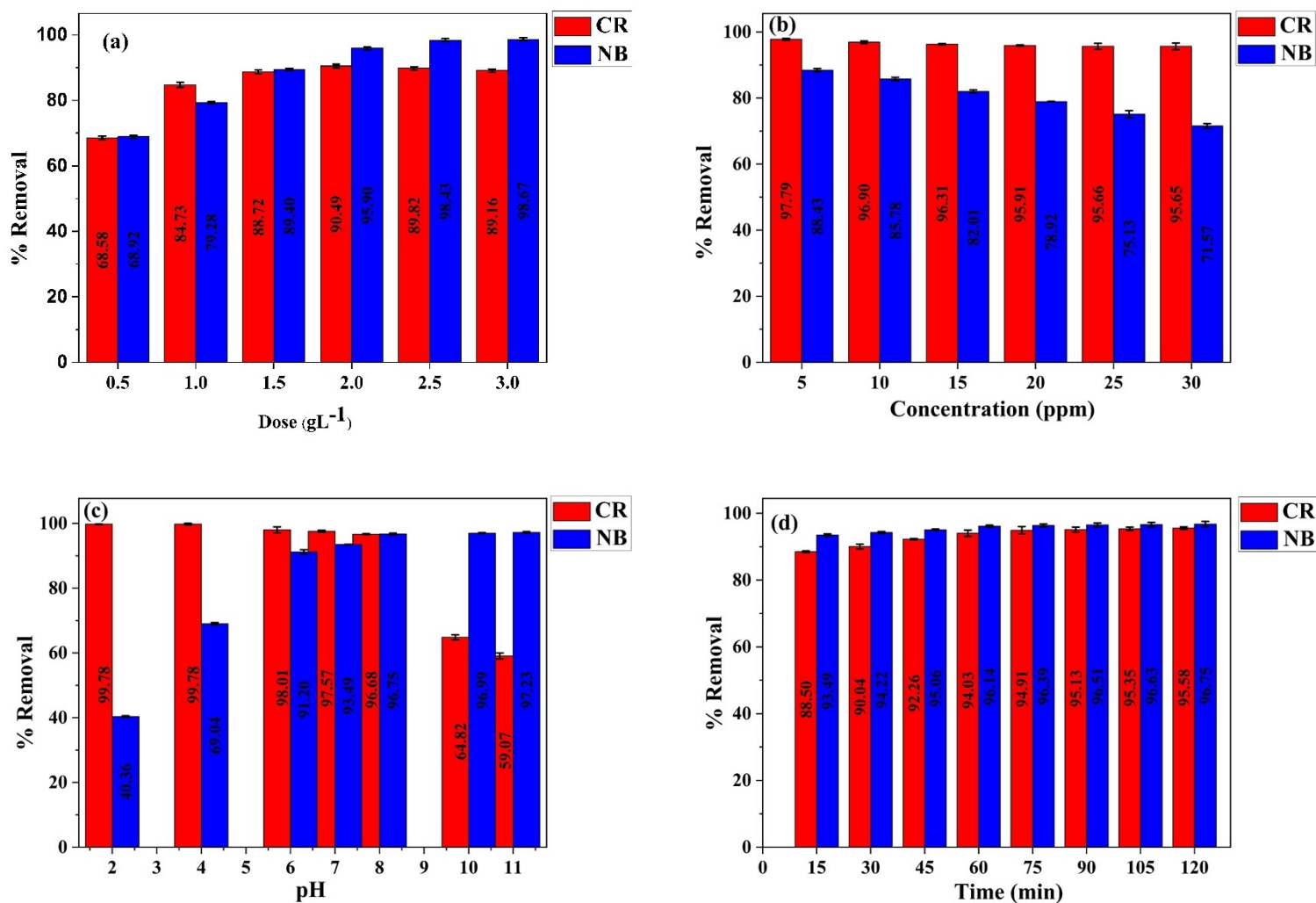


Fig. S3: Effect of operative variables on removal of CR and NB dyes, (a) Dosage of GG/ γ -Fe₂O₃, (b) Concentration of dyes, (c) pH and (d) Time of equilibration

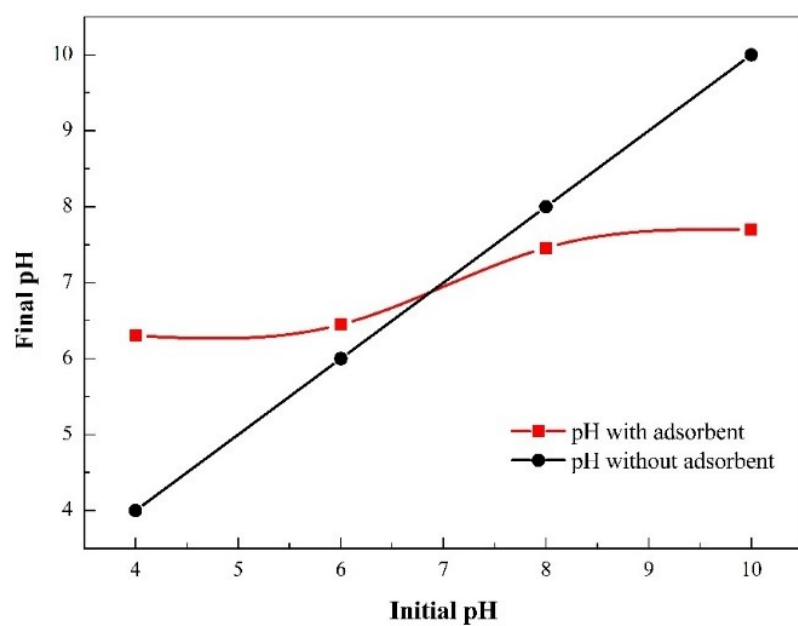


Fig. S4: Zero-point charge of GG/γ-Fe₂O₃

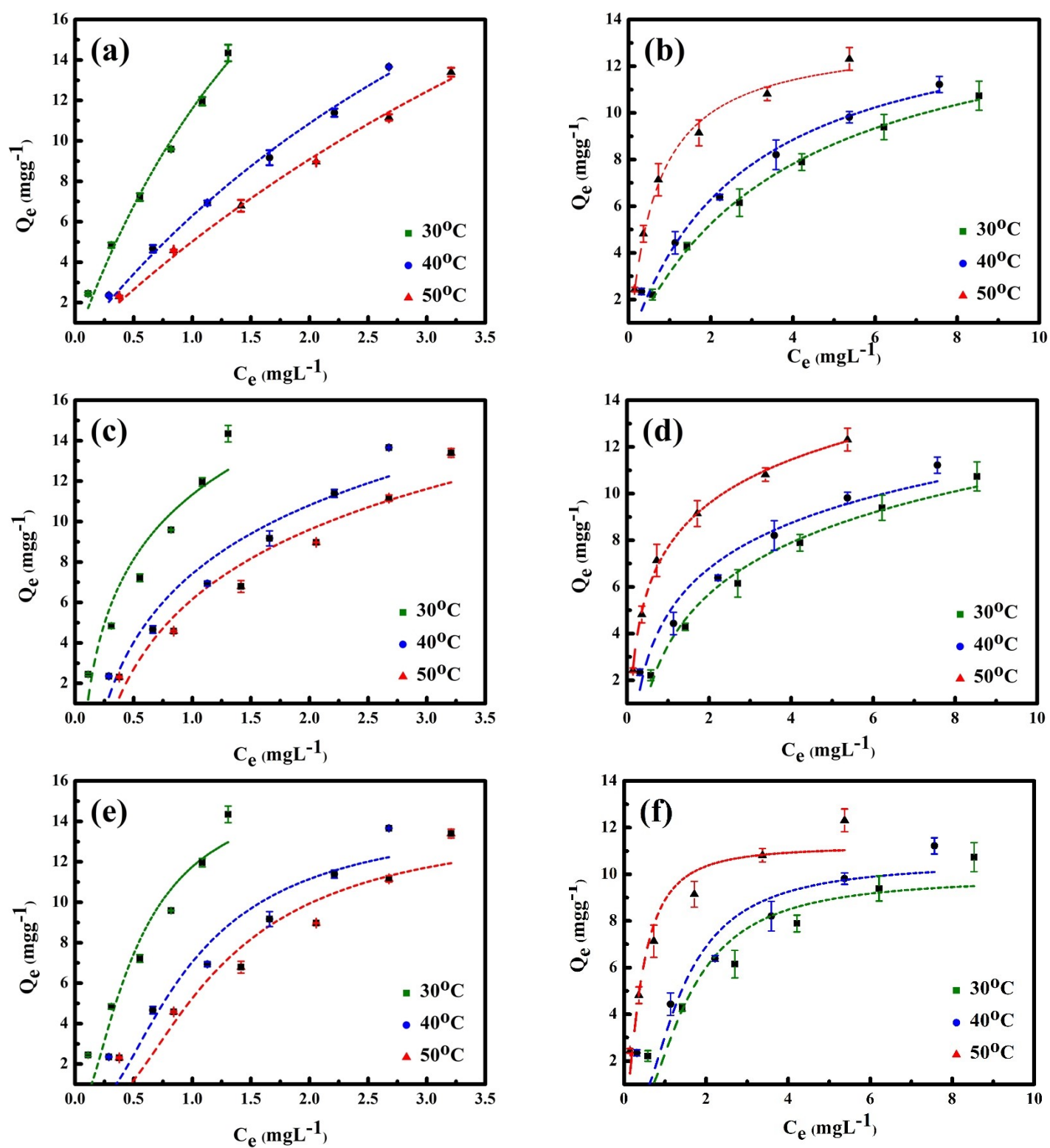


Fig. S5: Non-linear (a) Langmuir, (c) Temkin, and (e) Dubinin-Radushkevich isotherms for CR adsorption; (b) Langmuir, (d) Temkin, and (f) Dubinin-Radushkevich isotherms for NB adsorption

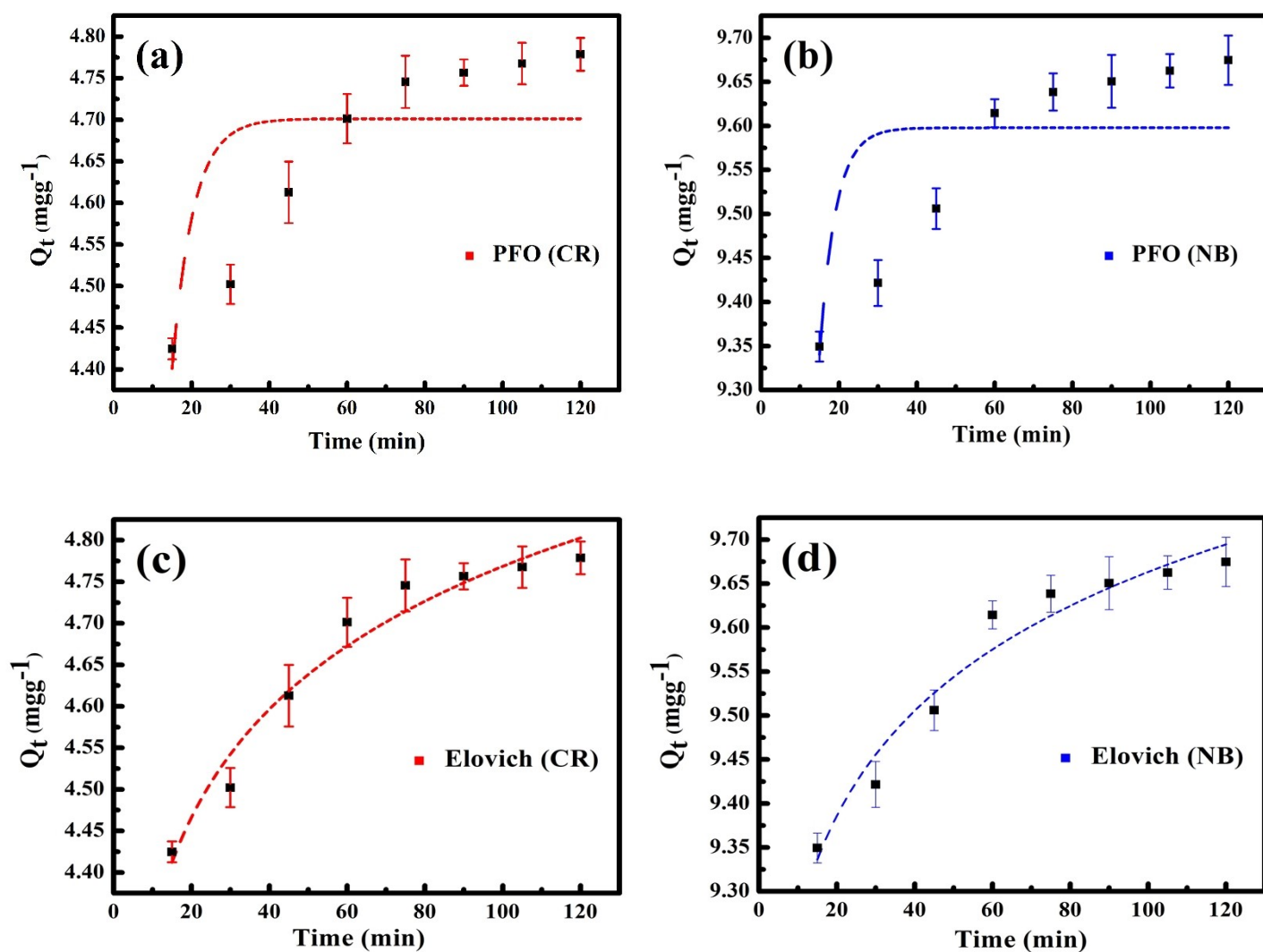


Fig. S6: Non-linear (a) Pseudo-first order (PFO) and (c) Elovich plots for CR adsorption; (b) Pseudo-first order (PFO) and (d) Elovich plots for NB adsorption

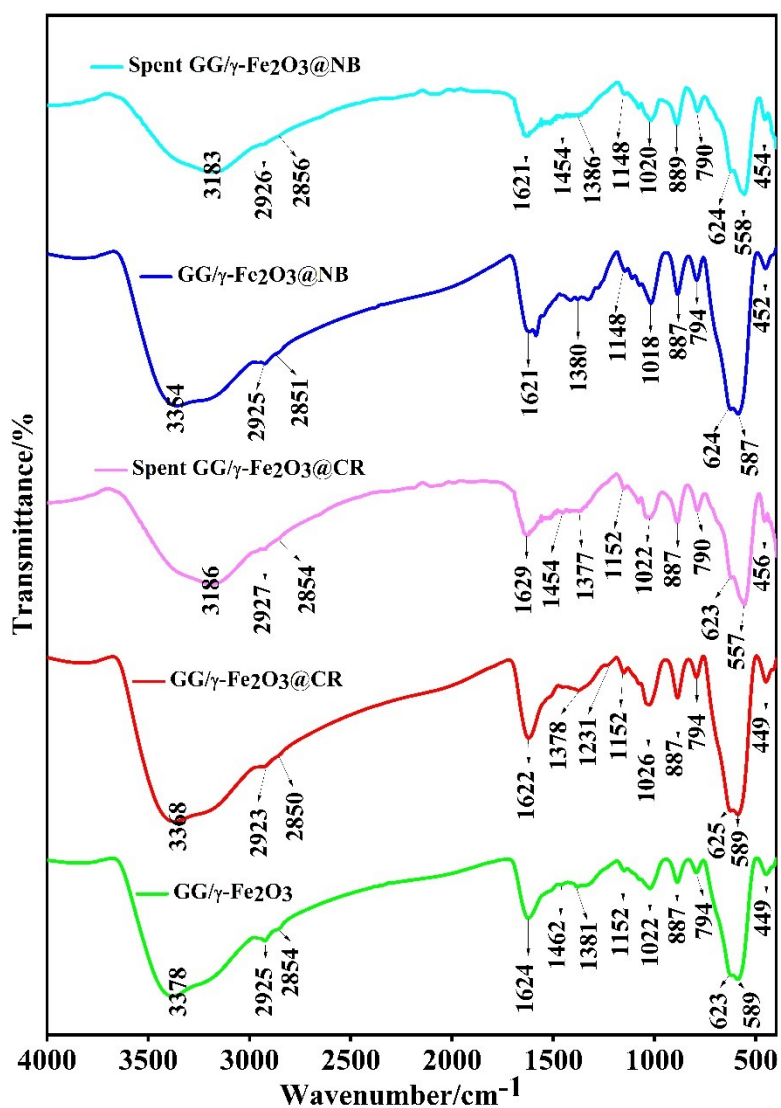


Fig. S7. FT-IR spectra of GG/γ-Fe₂O₃ before and after CR/NB adsorption, and of spent GG/γ-Fe₂O₃ after desorbing CR/NB dyes

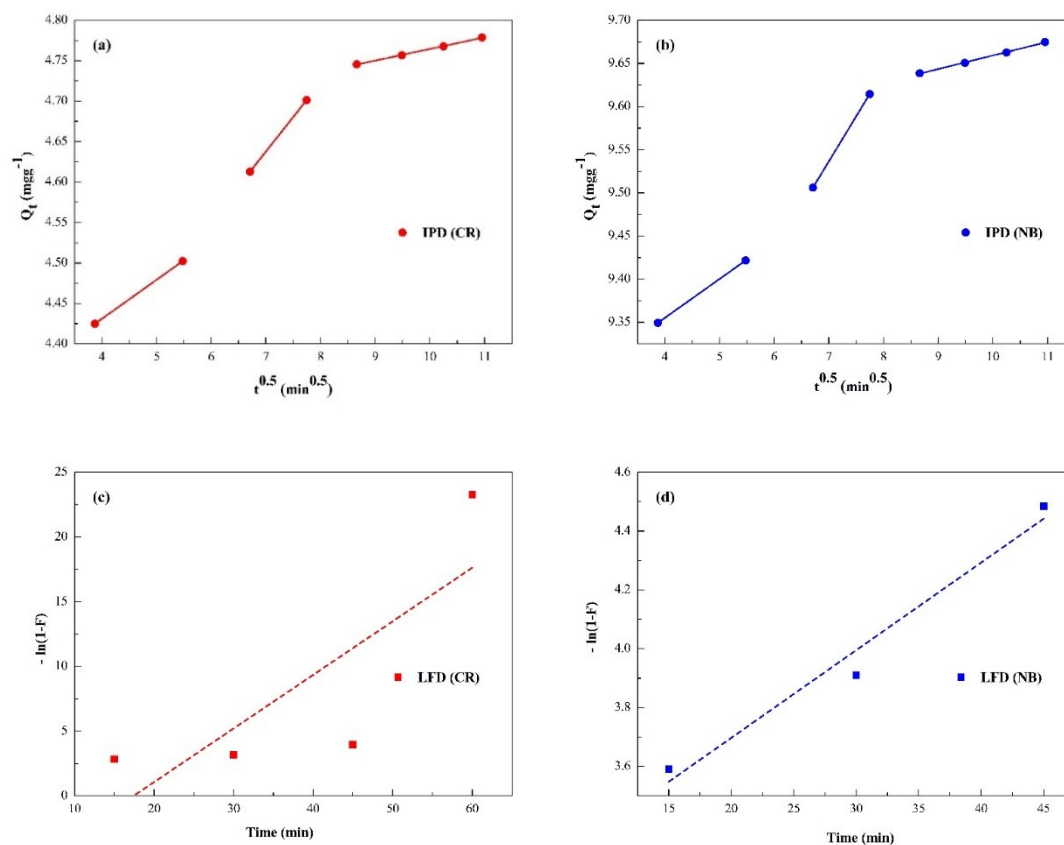


Fig. S8: (a) and (b) depicts Webber-Morris/ Intra-particle diffusion (IPD) model, and (c) and (d) depicts Liquid film diffusion model (LFD) for CR and NB sorption on GG/ γ -Fe₂O₃, respectively

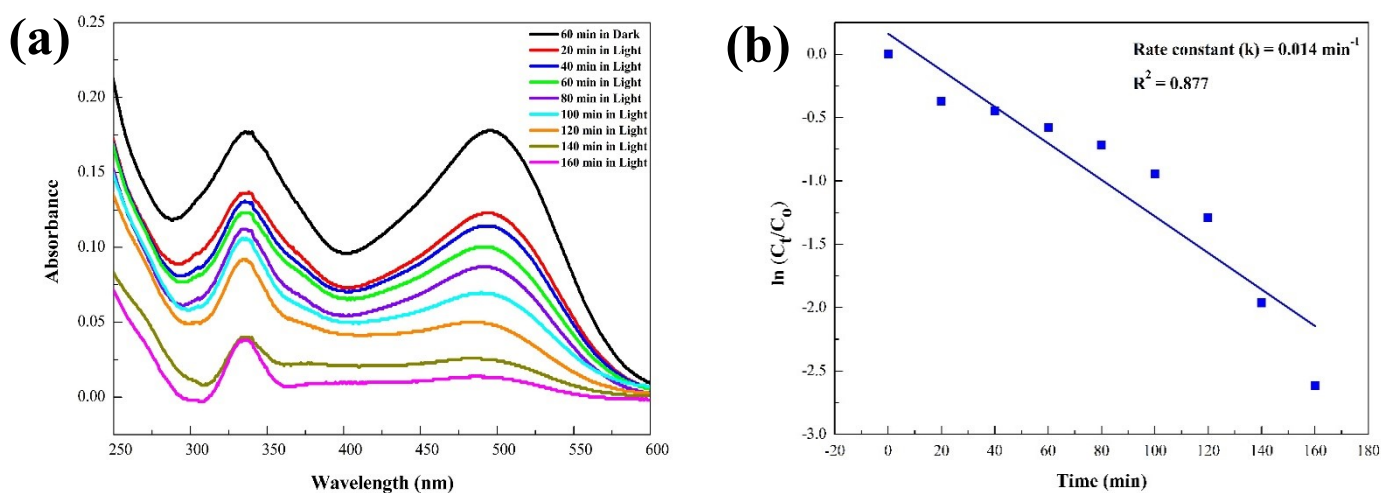


Fig. S9: (a) Time dependent UV-Visible absorption spectra, and (b) Pseudo-first order (PFO) kinetic plot for photodegradation of CR dye using GG/ γ -Fe₂O₃

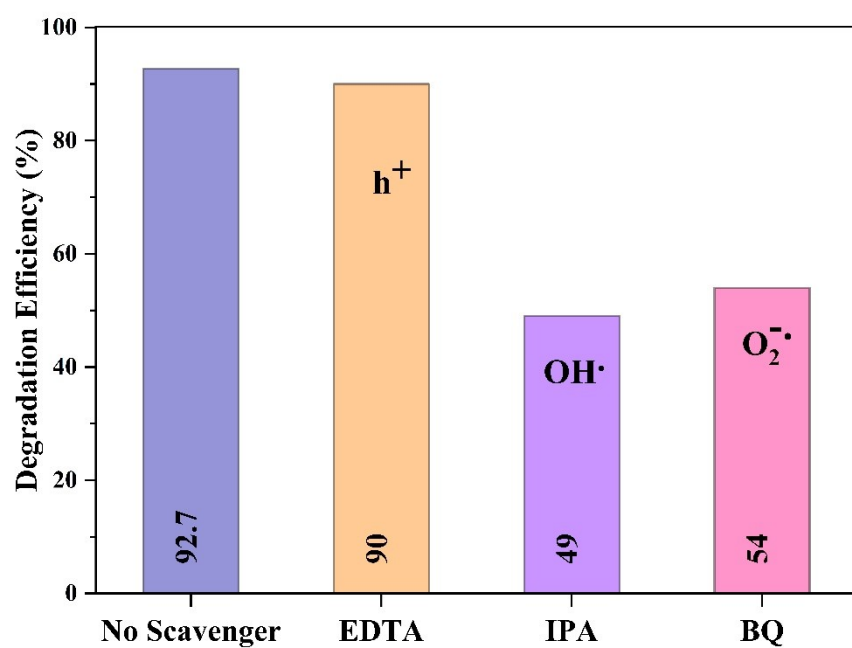


Fig. S10. Effect of radical scavengers on the photocatalytic degradation performance of GG/γ-Fe₂O₃

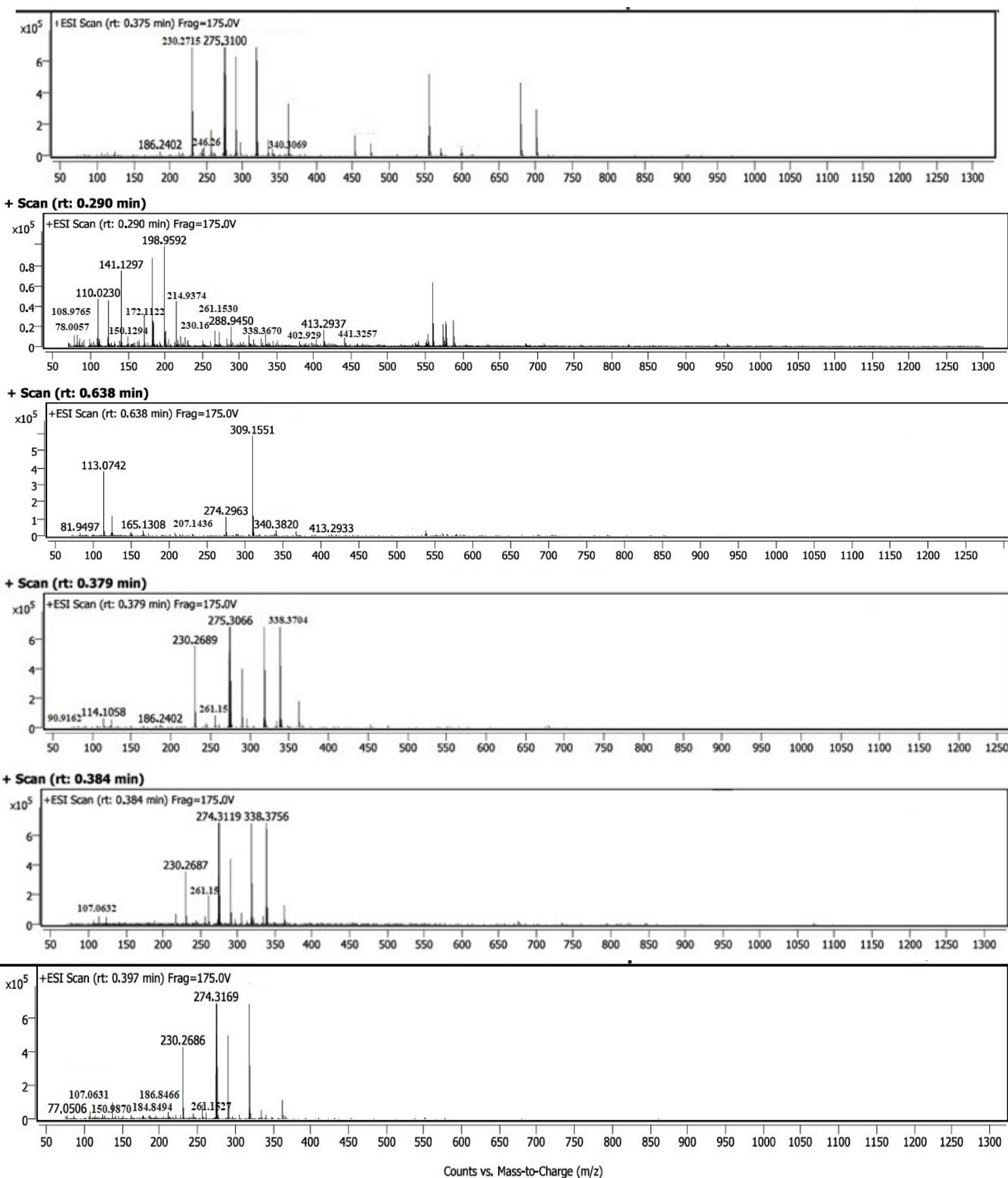


Fig. S11. Time resolved high resolution LC-HRMS mass spectra for CR degradation by

GG/ γ -Fe₂O₃ at different time intervals

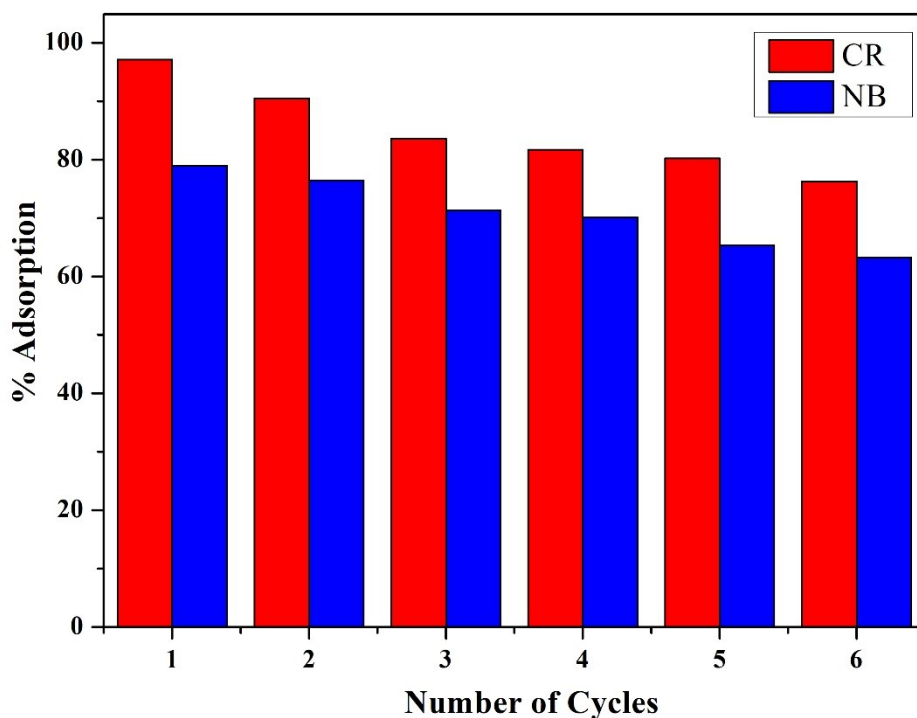


Fig. S12. Regeneration-reusability cycles of GG/ γ -Fe₂O₃

Supplementary Tables:

Table S1. Chemicals utilised for the synthesis of GG/ γ -Fe₂O₃

Chemicals/Reagents* (AR grade)/Materials	Purchased from
<i>Glycyrrhiza glabra</i> plant roots	Local market in West Delhi, India
Ferrous sulphate heptahydrate {FeSO ₄ .7H ₂ O, minimum assay > 99.5%}, Calcium chloride dihydrate {≥ 98%}, Direct Red-81 {DR-81}	Merck India Ltd., New Delhi
Ferric nitrate nanohydrate {Fe(NO ₃) ₃ .9H ₂ O, minimum assay 98%}	Thermo Fisher Scientific India Pvt. Ltd., New Delhi
Congo red {C ₃₂ H ₂₂ N ₆ Na ₂ O ₆ S ₂ , M.W.= 696.65gmol ⁻¹ , λ _{max} = 498 nm}, Isopropanol {IPA, min 99%}, Malachite green {MG}, Eosine yellow {EY}, New fuchsin {NF}, Thymol blue {TB}, Crystal violet {CV}, Phloxin B {Ph}, Chlorophenol red {Ch-R}	Loba Chemie Pvt., Ltd., India
Nile blue {C ₂₀ H ₂₀ ClN ₃ O, M.W.= 353.85 gmol ⁻¹ , λ _{max} = 627 nm}	HiMedia Laboratories Pvt. Ltd., India
Bismark brown-R {BB-R}	CDH (P) Ltd., India
Sodium hydroxide pellets {minimum assay > 97%}, Hydrochloric acid {minimum assay (alkalimetric) ≥ 32%}, and Potassium nitrate {minimum assay 99%, Sodium sulphate {minimum assay 99.5%, Sodium nitrate {minimum assay ≥	Merck India Ltd., New Delhi

99%}, Sodium chloride {minimum assay $\geq$ 99%}, Sodium carbonate {minimum assay $\geq$ 99.5%}	
Acetone {minimum assay $>$ 99.5%, Alizarin red S {AR-S}	Fisher Scientific Ltd., India
EDTA {99%}	SRL Pvt. Ltd., India
1,4-benzoquinone {BQ, 99%}	Ottokemi, India
Ethanol, Yeast extract, Peptone, Dextrose, Dimethyl sulphoxide (DMSO), and Agar	Sigma Chemical Co., St. Louis, MO, USA
Fungal strains of <i>Candida albicans</i> SC-5314 and ATCC-90028	National Institute for Microbial Resource (NCMR), Pune, India
Bacterial strains of <i>Escherichia coli</i> (ATCC-25922) and <i>Staphylococcus aureus</i> (MTCC-902)	Institute of Microbial Technology (IMTECH), Chandigarh, India

*All chemicals and reagents were used as obtained without any additional purification procedure. Doubly distilled water was used throughout the experiments.

Table S2. Characterization of GG/ γ -Fe₂O₃

Characterization Technique	Sample(s)	Instrument Used	Range and Specifics	Information
FT-IR	GG, GG/ γ -Fe ₂ O ₃ , GG/ γ -Fe ₂ O ₃ @CR, GG/ γ -Fe ₂ O ₃ @NB	Perkin-Elmer spectrometer	4000-400 cm ⁻¹ using pellets in KBr	Specific interactions between metal oxide, GG and dyes, functional group analysis
	Spent GG/ γ -Fe ₂ O ₃ @CR, Spent GG/ γ -Fe ₂ O ₃ @NB	Nicolet iS50		
P-XRD	GG, GG/ γ -Fe ₂ O ₃	Rigaku diffractometer	Continuous scanning angular range 05 to 90° (2 θ), Cu-K α radiation of λ =1.54 Å (40 kV, 30 mA) at RT	Crystal structure, phase and crystal size
FE-SEM	GG and GG/ γ -Fe ₂ O ₃	Thermal field emission type Zeiss Gemini SEM 500	10-15 kV accelerating voltage	Morphological and microstructural investigation
HR FE-SEM	GG/ γ -Fe ₂ O ₃ and GG/ γ -Fe ₂ O ₃ @CR	Nova Nano SEM-450, FEI, Netherlands	10 kV	
EDX	GG and GG/ γ -Fe ₂ O ₃	Thermal field emission type Zeiss Gemini SEM 500 microscope	15 kV	Identification and mapping of elemental composition
TEM and SAED	GG/ γ -Fe ₂ O ₃	F30 S-Twin, Tecnai microscope	-	Morphological and microstructural distribution of particles, and particle size
BET	GG/ γ -Fe ₂ O ₃	Micromeritics' Gemini VII 2380 Series surface analyzer	N ₂ adsorption-desorption isotherm	Surface area and porosity

VSM	GG/ γ -Fe ₂ O ₃	MicroSense, Model ADE-EV9	-2 to +2 T	Magnetic property measurements
XPS	GG/ γ -Fe ₂ O ₃	Thermo Scientific K-Alpha XPS System	0-1350 eV; Al K α source (h ν = 1486.6 eV)	Elemental analysis and bonding interactions
TGA	GG/ γ -Fe ₂ O ₃	TA TGA 5500	N ₂ atmosphere, 30-800° C, Heating Rate: 10°Cmin ⁻¹	Thermal stability
UV-Vis and CR degradation	GG/ γ -Fe ₂ O ₃	UV-1800, Shimadzu UV-spectrophotometer	-	Band gap estimation
PL	GG, γ -Fe ₂ O ₃ , GG/ γ -Fe ₂ O ₃	WiTech alpha 300R, Germany	Excitation λ = 325 nm (He-Cd laser)	Charge transfer efficiency, and rate of recombination
LC-HRMS	CR degradation intermediates	Agilent G6530AA (LC-HRMS-BIO-QTOF)	Source: ESI and APCI; HPLC pressure: 600 Bar, MS pressure: 10 ⁻⁴ to 10 ⁻⁵ Torr	CR degradation intermediates and byproducts

Table S3. Standard deviation (SD) values for chosen parameter space for dye adsorption by GG/ γ -Fe₂O₃

Dose (g L ⁻¹)		0.5	1	1.5	2	2.5	3		
	CR	0.57518	0.72514	0.60069	0.57518	0.52452	0.45894		
	NB	0.43035	0.32741	0.4	0.4	0.44576	0.50863		
Concentration (mg L ⁻¹)		5	10	15	20	25	30		
	CR	0.25007	0.7	0.19757	0.92738	1.10036	0.75		
	NB	0.47085	0.50319	0.46184	0.0755	1.05057	0.71042		
pH		2	4	6	7	8	10	11	
	CR	0.07211	0.25239	0.91504	0.32512	0.12583	0.78307	0.91198	
	NB	0.39345	0.66091	1.11355	0.09	0.45	0.2623	0.40415	
Contact time (min)		15	30	45	60	75	90	105	120
	CR	0.45	0.54	0.41	0.43	0.61	0.20421	0.5	0.38
	NB	0.34828	0.28844	0.22539	0.29462	0.4293	0.55973	0.66461	0.80467

Table S4. Statistical treatment using one-way ANOVA analysis of operational conditions for CR and NB adsorption by GG/ γ -Fe₂O₃

Parameter	Degrees of Freedom (DF)		Sum of Squares (SS)		Mean Square (MS)		F value		Prob>F (P-value)	
	CR	NB	CR	NB	CR	NB	CR	NB	CR	NB
Dosage	5	5	1060.93574	2063.3058	212.18715	412.66116	625.89684	2314.42042	4.53281E-14	1.80951E-17
Concentration	5	5	10.56629	611.692	2.11326	122.3384	3.93177	318.82483	0.02416	2.52611E-12
pH	6	6	5760.22846	8326.02903	960.03808	1387.6715	2717.8581	4271.01005	1.22826E-20	5.20416E-22
Contact time	7	7	151.737	32.26886	21.67671	4.60984	104.76271	19.11903	3.65731E-12	1.17045E-6

Table S5. d-spacing calculated from TEM-SAED pattern of GG/ γ -Fe₂O₃

S.No.	1/2r (nm ⁻¹)	1/r (nm ⁻¹)	r (nm)	d-spacing	Miller indices (hkl)
1.	6.797	3.398	0.294	2.942	(220)
2.	7.986	3.993	0.250	2.504	(311)

3.	9.667	4.833	0.207	2.069	(400)
4.	11.715	5.857	0.171	1.707	(422)
5.	12.590	6.295	0.159	1.589	(511)
6.	13.771	6.885	0.145	1.452	(440)

Table S6. MIC ($\mu\text{g mL}^{-1}$) and MBC ($\mu\text{g mL}^{-1}$) of GG and GG/ $\gamma\text{-Fe}_2\text{O}_3$ against bacterial stains

MIC & MBC ($\mu\text{g mL}^{-1}$) against tested bacterial strains		<i>E. coli</i>	<i>S. aureus</i>
GG	MIC	550.25 ± 1.85	425.5 ± 1.25
	MBC	1100.5 ± 2.85	850.2 ± 1.85
GG/ $\gamma\text{-Fe}_2\text{O}_3$	MIC	250 ± 3.15	200 ± 2.62
	MBC	500 ± 1.98	400 ± 3.67

Table S7. MIC ($\mu\text{g mL}^{-1}$) and MFC ($\mu\text{g mL}^{-1}$) of GG and GG/ $\gamma\text{-Fe}_2\text{O}_3$ against tested fungal strain

MIC & MFC ($\mu\text{g mL}^{-1}$) against tested fungal strains		<i>C. albicans (SC5314)</i>	<i>C. albicans (ATCC90028)</i>
GG	MIC	300.85 ± 8.58	320.74 ± 5.65
	MFC	600.56 ± 7.56	645.32 ± 3.64
GG/ $\gamma\text{-Fe}_2\text{O}_3$	MIC	131.25 ± 2.56	125 ± 1.15
	MFC	465.5 ± 3.12	450 ± 2.30

Table S8. Parameters governing the mechanism of adsorption of CR and NB dyes on GG/ $\gamma\text{-Fe}_2\text{O}_3$

Pollutant	Intra-particle diffusion (Webber-Morris model)			Liquid film diffusion (Boyd model)	
	$k_{\text{IPD}} (\text{mg g}^{-1} \text{min}^{-0.5})$	C	R^2	$k_{\text{LFD}} (\text{min}^{-1})$	R^2
CR	0.053	4.240	0.923	0.414	0.468
NB	0.049	9.178	0.919	0.030	0.946

Table S9. Removal of CR and NB dyes by GG/ $\gamma\text{-Fe}_2\text{O}_3$ in real wastewater setup, and amidst competing ions

1. Real water analysis			
Type of water sample	% CR adsorption	% NB adsorption	% CR degradation
Sewage water	33.03	36.6	26.19
Tap water	57.84	44.18	42.85
RO water	91.81	89.55	54.79
Distilled water	97.51	91.38	92.7
2. Effect of co-existing ions			
NO_3^-	95.6	90.45	74.29
Cl^-	97.02	87.66	89.25
SO_4^{2-}	94.34	82.72	64.63
CO_3^{2-}	0.81	84.46	6.85
Na^+	96.9	80.66	89.25
Ca^{2+}	90.17	82.15	85.71

Table S10. Effect of dye mixtures on CR and NB adsorption by GG/ γ -Fe₂O₃

10 mg ⁻¹ Dye 1 (A)	10 mg ⁻¹ Dye 2 (A)	20 mg ⁻¹ Dye 3 (C)	20 mg ⁻¹ Dye 4 (C)	% CR adsorption	% NB adsorption
CR	-	-	-	97.51	-
-	-	NB	-	-	91.38
CR	-	NB	-	85.44	86.99
CR	EY	NB	NF	67.88	87.37
CR	DR-81	NB	BB-R	82.87	82.76
CR	AR-S	NB	CV	90.14	89.07
CR	TB	NB	Ph	84	73.26
CR	ChR	NB	MG	46.77	78.02

Table S11. Cost estimation for laboratory-scale synthesis of GG/ γ -Fe₂O₃

Breakup of cost	Amount per batch	Unit Cost	Cost (USD, \$)	Remarks
<i>Glycyrrhiza glabra</i> root	10 g	Negligible	\$0/g	Locally sourced
Fe(NO ₃) ₃ ·9H ₂ O	2.02 g	\$35.09/250 g	\$0.28/g	ThermoFisher
FeSO ₄ ·7H ₂ O	0.695 g	\$43.91/250 g	\$0.12/g	Merck
NaOH, Ethanol	Small	Estimate	\$0.15	Neutralisation/purification
Distilled water (DW)	~1 L	Estimate	\$0.06	Lab-prepared
Energy	-	Estimate	\$0.18-0.27	For heating, drying, stirring
Total (per batch)	-	-	\$0.79-0.88	Lab-scale estimation

Table S12. Cost of water treatment by GG/ γ -Fe₂O₃ under optimized conditions

Factor	Approx. Value	Cost Implication (USD, \$)
GG/ γ -Fe ₂ O ₃ dose	0.2 g L ⁻¹	\$0.158–\$0.176/L (\$158–\$176/1000 L)
Optimum adsorption time	60 min	Feasible for batch adsorption setups
Sunlight-driven photodegradation	160 min	None
pH Adjustment	pH=2-4 (CR); pH=11 (NB)	\$0.012–\$0.024 per 1000 L (Estimate)
Regeneration cycles	4 cycles (practically feasible)	Recurring cost reduced by 70–80%

Table S13. Comparative study of other adsorbents used for CR and NB removal

Pollutant	Adsorbent	Dose (gL ⁻¹)	Conc. (mgL ⁻¹)	Time (min)	pH	Temp (°C)	Adsorption capacity (mgg ⁻¹)	Reference
CR	Fly ash	1	30	60	-	50	22.12	10
	Zeolite	-	-	-	-	-	8.1	11
	<i>Padina gymnospora</i> (PG)	-	-	-	-	-	11.59	
	Zeolite/algae composite (ZPG)	1	20	480	7	25	11.26	
	SiO ₂ -PDA-PTAC	0.2	60	180	3	65	125	12

	Cu/TiO ₂	1	20	10	6	-	46.49	13
	GO-MnFe ₂ O ₄	1	40	60	-	-	8.597	14
	G-3PS	1	22	30	5	40	14.82	15
	G-5PS		22	20	5	40	16.04	
	GG/ γ -Fe ₂ O ₃	0.2	10	60	2-4	50	47.504	Present Study
NB	Ag-rGO	0.5	20	60	6	30	6.685	16
	RAM	-	-	-	-	-	21.2	17
	P(AAm-co-AcA)	1.5	-	-	-	-	16.86	18
	Natural clay	4	500	480	5	-	25	19
	GG/ γ -Fe ₂ O ₃	0.2	20	60	11	30	15.361	Present Study

Table S14. Comparative analysis of CR degradation efficiency with previously reported catalysts.

Photocatalyst	Dose (g/L ⁻¹)	Conc. (mg/L ⁻¹)	Time (min)	PFO rate Constant ($\times 10^{-3} \text{ min}^{-1}$)	Light Source	% CR degradation	Reference
Co ₃ O ₄ /TiO ₂ /GO	0.25	10	90	23.5	Xe 300 W	91	1
CS/n-CdS	1.5	20	180	11.08	Xe 300 W	86	2
P25 TiO ₂	1	40	210	-	W 300W	90	3
Cu foam	-	30	300	3.53	Sunlight	60	4
CuO				5.01		75	
ZnO@CuO				7.12		90	
Fe ₃ O ₄ @CuO				7.95		94	
PAC	2	50	1.5	1513.8	Microwave	87.79	5
Iron promoted TiO ₂ AC NCs	10	$5 \times 10^{-5} \text{ M}$	300	2.11-4.70	Visible	84.46	6
Cellulose/ γ -Fe ₂ O ₃ -ZrO ₂	50 mg	100	30	9.1	Visible	98.5	7
α -Fe ₂ O ₃ /rGO	0.2	50	30	31.8	Visible	87	8
BaDy _x Fe _{12-x} O ₁₉	0.3	10	90	19	Sunlight	89.29	9
GG/ γ -Fe ₂ O ₃	0.2	10	160	14.00	Sunlight	92.7	Present Study

References

- 1 W. K. Jo, S. Kumar, M. A. Isaacs, A. F. Lee and S. Karthikeyan, Cobalt promoted TiO₂/GO for the photocatalytic degradation of oxytetracycline and Congo Red, *Appl Catal B*, 2017, **201**, 159–168.
- 2 H. Zhu, R. Jiang, L. Xiao, Y. Chang, Y. Guan, X. Li and G. Zeng, Photocatalytic decolorization and degradation of Congo Red on innovative crosslinked chitosan/nano-CdS composite catalyst under visible light irradiation, *J Hazard Mater*, 2009, **169**, 933–940.
- 3 H. X. Guo, K. L. Lin, Z. S. Zheng, F. Bin Xiao and S. X. Li, Sulfanilic acid-modified P₂₅ TiO₂ nanoparticles with improved photocatalytic degradation on Congo red under visible light, *Dyes and Pigments*, 2012, **92**, 1278–1284.
- 4 D. Malwal and P. Gopinath, Enhanced photocatalytic activity of hierarchical three-dimensional metal oxide@CuO nanostructures towards the degradation of Congo red dye under solar radiation, *Catal Sci Technol*, 2016, **6**, 4458–4472.
- 5 Z. Zhang, Y. Shan, J. Wang, H. Ling, S. Zang, W. Gao, Z. Zhao and H. Zhang, Investigation on the rapid degradation of congo red catalyzed by activated carbon powder under microwave irradiation, *J Hazard Mater*, 2007, **147**, 325–333.
- 6 D. Negoescu, V. Bratan, M. Gherendi, I. Atkinson, D. C. Culita, A. Neacsu, A. Baran, S. Petrescu and V. Parvulescu, Iron Promoted TiO₂-Activated Carbon Nanocomposites for Photocatalytic Degradation of Congo Red in Water, *Catalysts*, 2024, **14**, 844.
- 7 H. Helmiyati, N. Fitriana, M. L. Chaerani and F. W. Dini, Green hybrid photocatalyst containing cellulose and γ -Fe₂O₃-ZrO₂ heterojunction for improved visible-light driven degradation of Congo red, *Opt Mater (Amst)*, 2022, **124**, 111982.
- 8 Y. Wang, S. Wang, Y. Wu, Z. Wang, H. Zhang, Z. Cao, J. He, W. Li, Z. Yang, L. Zheng, D. Feng, P. Pan, J. Bi, H. Li, J. Zhao and K. Zhang, A α -Fe₂O₃/rGO magnetic photocatalyst: Enhanced photocatalytic performance regulated by magnetic field, *J Alloys Compd*, 2021, **851**, 156733.
- 9 Himanshi, J. Makasana, S. Ganesan, B. Lal, K. S. Naidu, J. Ahmed, J. Prakash, A. Verma, N. Lakshmaiya, R. Jasrotia, X. Yong and C. K. Chan, Insights into the enhanced photocatalytic degradation of congo red using advanced BaD_yxFe_{12-x}O₁₉ catalytic hexamaterials, *Sci Rep*, 2025, **15**, 1–20.
- 10 M. Harja, G. Buema and D. Bucur, Recent advances in removal of Congo Red dye by adsorption using an industrial waste, *Sci Rep*, 2022, **12**, 1–18.
- 11 A. R. Dryaz, M. Shaban, H. AlMohamadi, K. A. A. Al-Ola, A. Hamd, N. K. Soliman and S. A. Ahmed, Design, characterization, and adsorption properties of Padina gymnospora/zeolite nanocomposite for Congo red dye removal from wastewater, *Sci Rep*, 2021, **11**, 21058.
- 12 J. Zhu, K. Wang, S. Fan, A. Ouyang, J. Gu, J. Tian, M. Liu, F. Deng, X. Zhang and Y. Wei, Preparation of SiO₂ composites for efficient removal of Congo red via the combination of mussel-inspired surface modification and anionic ring-opening polymerization, *Colloids and Surfaces C: Environmental Aspects*, 2025, **3**, 100078.
- 13 S. Zhang, Q. Luo, X. Sun, L. Chi, P. Sun and L. Zhang, Facile synthesis copper-modified titania (Cu/TiO₂) nanoparticles for high-efficiency Congo red adsorption, *Chin J Chem Eng*, 2025, **81**, 87–94.
- 14 A. Zourou, A. Ntziouni, N. D. Adamopoulos, T. Roman, F. Zhang, M. Terrones and K. V. Kordatos, Graphene oxide-MnFe₂O₄ nanohybrid material as an adsorbent of Congo red dye, *Journal of Physics and Chemistry of Solids*, 2023, **181**, 111490.
- 15 A. Ebelegi, N. Ayawei and D. Wankasi, Efficient Sequestration of Congo Red Dye From Aqueous Solutions Using Pamam Dendrimer – Silica Composite, *Water Air Soil Pollut*, 2024, **235**, 1–15.

- 16 N. Natasha, A. Khan, U. U. Rahman, N. Sadaf, M. Yaseen, R. A. Abumousa, R. Khattak, N. Rehman, M. Bououdina and M. Humayun, Effective Removal of Nile Blue Dye from Wastewater using Silver-Decorated Reduced Graphene Oxide, *ACS Omega*, 2024, **9**, 19461–19480.
- 17 G. Güçlü, Removal of basic dyes from aqueous solutions by dimethyl terephthalate distillation residue, *Desalination*, 2010, **259**, 53–58.
- 18 Y. Işıkver, Removal of some cationic dyes from aqueous solution by acrylamide- or 2-hydroxyethyl methacrylate-based copolymeric hydrogels, *Fibers and Polymers*, 2017, **18**, 2070–2078.
- 19 T. B. Iyim and G. Güçlü, Removal of basic dyes from aqueous solutions using natural clay, *Desalination*, 2009, **249**, 1377–1379.