

Green synthesis of silver nanoparticles using *Eucalyptus comadulensis* leaves extract and its immobilization on magnetic nanocomposite (GO-Fe₃O₄/PAA/Ag) as a recoverable catalyst for degradation of organic dyes in water

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The magnetically recyclable graphene oxide-Fe₃O₄/polyallylamine (PAA)/Ag nanocatalyst was prepared via a green route using *Eucalyptus comadulensis* leaves extract as both reducing and stabilizing agent. The catalytic activity of this nanocatalyst was investigated for the reduction reaction of methylene blue and methyl orange in the presence of NaBH₄ in aqueous medium at room temperature. The prepared nanocatalyst was characterized by different methods such as Fourier transformed infrared spectroscopy, X-ray diffraction, scanning electron microscopy–energy dispersive X-ray spectroscopy, thermogravimetric analysis, vibrating sample magnetometer, transmission electron microscopy, and UV–visible spectroscopy. The results show that graphene oxide/PAA/Ag nanocatalyst has good activity and recyclability, and can be reused several times without major loss of activity in the reduction process. The apparent rate constants of the methyl orange (MO) and methylene blue (MB) were calculated to be 0.077 s^{−1} (3 mg of catalyst) and 0.15 s^{−1} (2 mg of catalyst), respectively.

KEYWORDS

dye degradation, graphene oxide, green synthesis, magnetic nanocatalystsilver nanoparticles

1 | INTRODUCTION

Organic dyes are applied in various industries, including paper, textile, leather, and food processing,^[1] and are one of the most dangerous water contaminants identified worldwide.^[2,3] The presence of dye pollutants is extremely dangerous and they have undesirable effects on human life.^[4,5] Therefore, the use of an efficient method for removing these pollutants is necessary. Recently, noble metal nanoparticles have been utilized for the removal of poisonous dyes in wastewater. Among these nanoparticles, silver nanoparticles

(Ag NPs) have received great consideration due to their high conductivity, stability, good catalytic activity, and antibacterial and antiviral properties.^[6–11] The agglomeration of nanoparticles during the catalytic reaction is one of their disadvantages.^[12–16] Separation of nanoparticles from the reaction medium using simple methods such as filtration and centrifugation is difficult.^[17–20] To overcome these difficulties, different solid supports such as TiO₂, graphene, mesoporous materials, and Fe₃O₄ were applied for the immobilization of metal nanoparticles and the construction of the heterogeneous nanocatalyst.^[21–32] Graphene oxide

(GO) has been identified as a new group of interesting catalysts because of their unique properties such as good stability, large surface area, high mechanical strength, and good adsorption capacity. GO has various applications including as electrochemical sensors and biosensors,^[33,34] in drug delivery,^[35] as dye adsorbents,^[36] as electrodes for supercapacitors,^[37] in batteries,^[38] and as catalysts.^[39] The green procedures for the preparation of metal NPs by plant extracts have various benefits over chemical or physical synthetic procedures, including simplicity, elimination of toxic and dangerous materials, use of nontoxic solvents, for example water, and moderate reaction conditions.^[40–42] In this research, a green synthesis of Ag NPs using eucalyptus leaf extract was reported. In addition, polyallylamine (PAA), a water-soluble polymer with a large number of positively charged amine groups, was connected to the GO-Fe₃O₄ nanocomposite for the reduction of organic dyes. Therefore, we synthesized GO-Fe₃O₄/PAA/Ag nanocatalyst and applied it in the reduction of MB and MO using NaBH₄ as the reducing agent. In addition, the synthesized nanocatalyst can be easily collected from the reaction solution using an external magnet for several cycles without considerable loss of its catalytic activity. The magnetic nanocatalyst of GO-Fe₃O₄/PAA/Ag nanocomposite can be used to reduce the MB and MO pollutants from the water.

2 | EXPERIMENTAL

2.1 | Synthesis of GO

Nanosheets of GO were synthesized with the modified Hummers' procedure.^[43] In a common procedure, a flask containing 250 ml of H₂SO₄ was placed in an ice bath and 2.5 g of NaNO₃ and 5.0 g of graphite were added to it. The reaction mixture was stirred for 15 min at the same temperature, then 35 g of KMnO₄ was gently added to it. Following 30 min of magnetic stirring, the temperature was raised to 35°C and held for 60 min. Then, 200 ml of distilled water was added dropwise to the reaction mixture with vigorously stirring and the temperature was maintained at 98°C. After 30 min stirring, this mixture was diluted with 1,000 ml of distilled water. A given amount of 30% H₂O₂ was dropped into the mixture. After finishing the reaction, the product was eluted using HCl and distilled water to remove the remained KMnO₄ and up to the pH of supernatant became neutral. Finally, the resulting brown solid was dried at 60°C for 24 hr.

2.2 | Synthesis of GO-COCl

The carboxylic acid groups of GO were reacted with SOCl₂ to form the acyl chloride. First, 0.3 g of GO was stirred in 25 ml of SOCl₂ for 24 hr at 70°C. The solution was then centrifuged and washed with dried tetrahydrofuran (THF) and dried in a freeze drier for 24 hr, to form acylated GO (GO-COCl).

2.3 | Synthesis of GO/PAA

GO-COCl (0.08 g) was dispersed in 1 ml of PAA and 100 ml of dry toluene for 15 min. The mixture was stirred for 3 hr at 50°C and the product was separated with an external magnet and washed with acetone then dried at ambient temperature.

2.4 | Synthesis of GO/PAA/Fe₃O₄

The desired amount of .03 g of GO/PAA powder was dispersed in 120 ml of water for 20 min. Then, 1.37 g (FeCl₃·6H₂O) and 0.5 g (FeCl₂·4H₂O) were added to the reaction mixture and it was stirred under an N₂ atmosphere at 60°C then 11 ml of NH₄OH was added to the mixture. The products were separated using an external magnet and eluted with distilled water.

2.5 | Preparation of leaf extract

E. comadulensis leaves were dried and powdered. Then 9.0 g of the powder was extracted using a Soxhlet (a device for extraction) for 24 h. After completing the extraction, the ethanolic extract was kept at 3–5°C until further use.

2.6 | Green preparation of Ag NPs

To synthesise the Ag NPs, 1 ml of the prepared leaf extract was added dropwise to 6 ml of 0.003 M solution of AgNO₃ at 60°C. The formation of Ag NPs from the reduction of silver ions occurred over about 1.5 hr. The reaction was controlled with UV-visible (UV-Vis) spectroscopy. The reaction mixture color changed after 1.5 hr, indicating the formation of the Ag NPs.

2.7 | Green synthesis of GO-Fe₃O₄/PAA/Ag nanocomposite

GO-Fe₃O₄/PAA (0.06 g) was added to 15 ml of ethanol and stirred for 15 min. Then, a 50 ml AgNO₃ (1.5 M) solution was added to the mixture and it was stirred at 60°C. Next 6 ml of aqueous extract was added to this mixture and it was stirred for 24 hr. The prepared product was separated with an external magnet, washed with distilled water and dried for 24 hr at ambient temperature.

2.8 | Catalytic degradation of organic dyes using GO-Fe₃O₄/PAA/Ag nanocatalyst

Catalytic reduction of MB and MO dyes was selected as the model reaction. The kinetics of the catalytic reduction reaction of these dyes were practically performed as below. To 2 ml of MB or MO solution (2.4×10^{-5} M) was added different amounts of GO-Fe₃O₄/PAA/Ag nanocatalyst (1, 2 or 3 mg), next, 2 mL of NaBH₄ aqueous solution (0.2 M) was added to it. The mixture was stirred at ambient temperature until it became colorless. The development of the conversion reaction was controlled using UV-Vis spectrophotometry in the range of 300–700 nm. After the reaction had finished, the nanocatalyst was isolated with an external magnet then eluted with ethanol and water. Since the prepared nanocomposite has magnetic properties, it can be easily separated from the reaction medium by an external magnet. This makes the catalyst recyclable.

3 | RESULTS AND DISCUSSION

3.1 | Structural and morphological characterization

3.1.1 | FT-IR spectroscopy

The nanostructure functional groups were characterized by Fourier transform infrared (FT-IR) spectroscopy, as illustrated in Figure 1a–e for GO, GO-COCl, GO/PAA, GO-Fe₃O₄/PAA, and GO-Fe₃O₄/PAA/Ag nanocomposite. The absorption peak at 3446 cm⁻¹ is related to the presence of stretching vibrations of OH groups in the GO structure. The characteristic peaks at 1408 and 1730 cm⁻¹ were attributed to the C–O and C=O stretching vibrations in the carboxylic acid groups at GO edges, respectively. Moreover, the peak at 1231 cm⁻¹ related to the epoxy groups confirmed the symmetric stretching, while the C–O deformation appeared at 1035 cm⁻¹ (Figure 1a). After modification of GO with SOCl₂, the peaks at

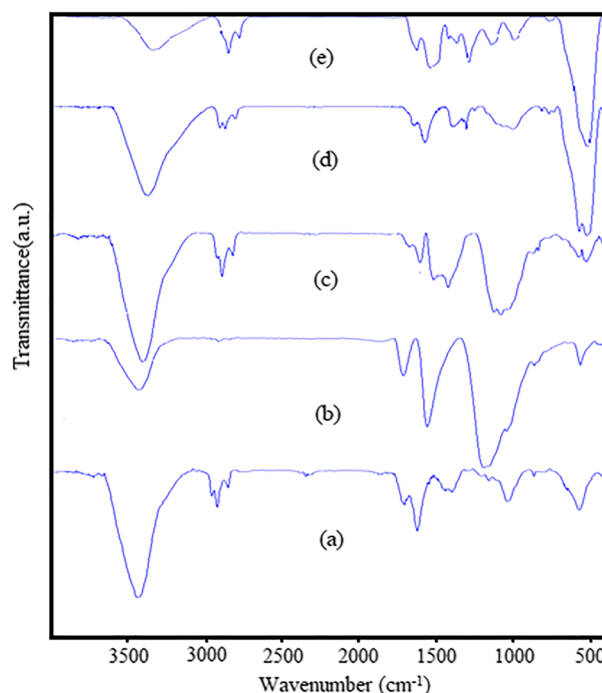


FIGURE 1 FT-IR spectra of (a) GO, (b) GO-COCl, (c) GO/PAA, (d) GO-Fe₃O₄/PAA, and (e) GO-Fe₃O₄/PAA/Ag

801 and 1729 cm⁻¹ confirmed C–Cl and C=O bonds (Figure 1b). On the GO-PAA spectrum, the bands at 2929 and 2852 cm⁻¹ were assigned to C–H stretching of the PAA alkyl group. In addition, the peaks at 1455, 1538, and 1632 cm⁻¹ indicated C–N stretching of the amide group, N–H of primary amine, and the coupling bending. The peak shifted to 1726 cm⁻¹ as a shoulder peak owing to the formation of the amide was related to the C=O band of the carboxylic group. This demonstrates that PAA polymer is grafted onto GO via an amide reaction between the PAA amine group and the GO carboxylic group, although some primary amine groups of PAA remain after the reaction (Figure 1c). Figure 1e shows the FT-IR spectrum of GO-Fe₃O₄/PAA/Ag nanocomposite. Since Ag NPs have no absorption peak in the IR zone, the FT-IR spectrum of the GO-Fe₃O₄/PAA nanocomposite is approximately the same as that of the GO-Fe₃O₄/PAA/Ag nanocomposite.

3.1.2 | X-ray diffraction

X-ray diffraction (XRD) is an important method used to study the crystal structures of materials. The XRD patterns of GO-Fe₃O₄/PAA and GO-Fe₃O₄/PAA/Ag nanocomposites are shown in Figure 2. The peak at $2\theta = 11.1^\circ$ can be assigned to the (001) plane of GO and the peaks at $2\theta = 30.2^\circ$ (220), 35.2° (311), 43.3° (400), 51.6° (422), 57.5° (511), and 62.5° (440) correspond to

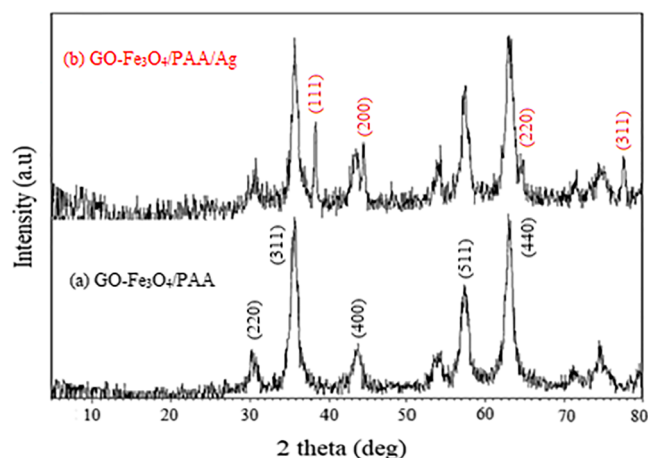


FIGURE 2 XRD patterns for (a) GO-Fe₃O₄/PAA and (b) GO-Fe₃O₄/PAA/Ag

Fe₃O₄ and are indexed in the Fd3m-Oh7 space group (no. 227) corresponding to a spinel cubic structure. Figure 2b also shows peaks at $2\theta = 38.1^\circ$, 44.3° , and 64.4° , which are related to the reflection of the (111), (200), and (220) Ag crystalline planes, with face-centered cubic structure. The average crystallite size was measured using the Debye–Scherrer formula^[44] which is calculated 26 nm for Ag NPs from the width of the plane (111).

3.1.3 | TEM and FE-SEM analysis

The transmission electron microscopy (TEM) image of the GO-Fe₃O₄/PAA/Ag nanocomposite (Figure 3a)

illustrates the uniform dispersion of the Fe₃O₄ and Ag nanoparticles, which are well located on GO sheets with a size range of 12–18 nm. A crumpled and sheet-like structure is clearly seen for the GO sheets, with the Ag and Fe₃O₄ nanoparticles randomly dispersed on them. The light-gray regions are the GO plates, and the dark zones are Fe₃O₄ and Ag NPs.

The morphology of the nanocomposite was studied with scanning electron microscopy (SEM), as shown in Figure 3b. The SEM image shows that Fe₃O₄ and Ag nanoparticles are dispersed on the GO plates. It can be seen that an aggregate consisting of Fe₃O₄ and Ag crystallites is dispersed as small, almost spherical nanoparticles with approximate sizes in the range 10–30 nm stabilized on the GO sheets. Energy-dispersive X-ray spectroscopy (EDS) analysis (Figure 3c) confirms the presence of Fe, O, C, and Ag in the GO-Fe₃O₄/PAA/Ag nanocomposite that was carried out with a field emission SEM (FE-SEM).

Inductively coupled plasma (ICP) analysis revealed that the loading of Ag nanoparticles in the nanocomposite was about 3.25 wt%, while the weight percentage of Fe was around 35.2 wt%. These amounts were close to the values obtained from the EDS spectrum.

3.1.4 | Vibration sampling magnetometer and TGA measurements

Figure 4a shows the magnetization curves for the Fe₃O₄, GO-Fe₃O₄/PAA and GO-Fe₃O₄/PAA/Ag nanocomposites. It has been reported that the saturation magnetization of Fe₃O₄ nanoparticles (Figure 4a, curve a) is 69.91 emu g^{-1} ,

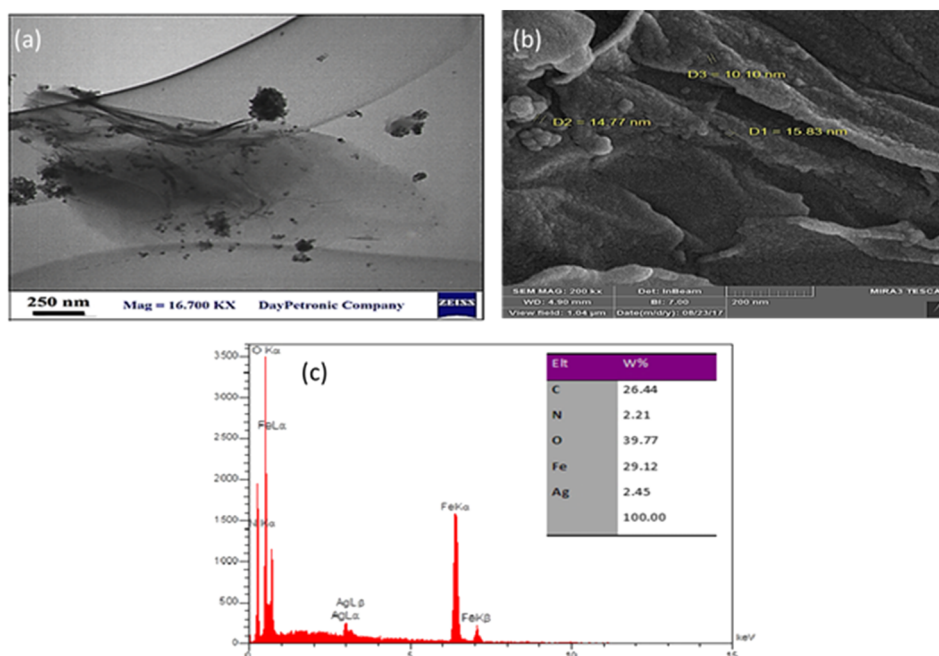


FIGURE 3 (a) TEM image, (b) SEM image, and (c) EDS spectrum of GO-Fe₃O₄/PAA/Ag nanocomposite

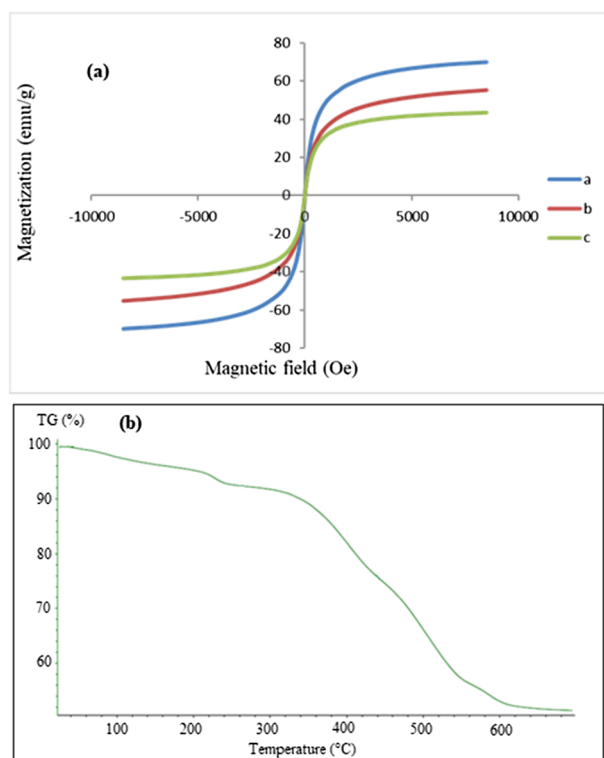


FIGURE 4 (a) Magnetization curves of Fe₃O₄ nanoparticles (curve a), GO-Fe₃O₄/PAA nanocomposites (curve b), and GO-Fe₃O₄/PAA/Ag (curve c). (b) TGA of GO-Fe₃O₄/PAA nanocomposites

the saturation magnetization of GO-Fe₃O₄/PAA nanocomposites (Figure 4a, curve b) is 55.25 emu g⁻¹, and the saturation magnetization of the GO-Fe₃O₄/PAA/Ag nanocatalyst (Figure 4a, curve c) is 43.43 emu g⁻¹. Due to the presence of the polymerized coating of PAA, the saturation magnetization value of Fe₃O₄ nanoparticles is greater than that of the GO-Fe₃O₄/PAA nanocomposites and higher than that of the GO-Fe₃O₄/PAA/Ag nanocatalyst. GO-Fe₃O₄/PAA/Ag nanocomposite has a smaller saturation magnetization value than GO-Fe₃O₄/PAA nanocomposite because of the presence of Non-magnetic silver NPs on the surface of the GO-Fe₃O₄/PAA nanocomposites. It should be noted that the GO-Fe₃O₄/PAA/Ag nanocatalyst still exhibits powerful magnetization, demonstrating its magnetic separation.

The thermal stability of the GO-Fe₃O₄/PAA/Ag nanocomposite was studied using thermogravimetric analysis (TGA) (Figure 4b). The TGA curve of GO-Fe₃O₄/PAA/Ag showed several steps of mass loss following the rise in temperature. The first mass loss (7.19%) at below 260°C is due to the evaporation of entrapped water in the GO layers. The second mass loss (16.70%) at 260–440 °C is due to the decomposition of PAA. The final mass loss (23.23%) occurs at a higher temperature that is due to the loss of oxygen-containing functional groups and the carbon skeleton in the GO.

3.1.5 | UV-Vis spectra of *E. comadulensis* leaf extract and green synthesised Ag NPs

Ag NPs were prepared using *E. comadulensis* leaf extract. The UV-Vis spectrum of the Ag NPs obtained in this way is shown in Figure 5b and the UV-Vis spectrum of *E. comadulensis* leaf extract is shown in Figure 5a. After the adding the AgNO₃ solution to the *E. comadulensis* leaf extract, the solution color changed to dark brown, indicating the synthesis of Ag⁰ from Ag⁺. The reaction development and the synthesis of the nanoparticles were monitored via UV-Vis spectrophotometry.

3.1.6 | Raman spectroscopy of GO

Since Raman spectroscopy is beneficial for investigating defects and disorder in the crystal structure, it is frequently used to characterize graphite and its derivatives. Raman spectra facilitate the study of conjugated and carbon-carbon double bonds, which cause high-intensity peaks in the Raman spectrum. The Raman spectrum of GO exhibited the presence of D peaks at 1352 cm⁻¹ and G peaks at 1603 cm⁻¹, confirming the formation of lattice distortions. The peak intensity of D to G band (ID/IG) was about 0.92 for GO, confirming the decrease in the mean size of sp² domains and edge defects due to the decrease of some C=C bonds in GO.

3.2 | Application of GO-Fe₃O₄/PAA/ag nanoparticles in the catalytic reduction of MB and MO

The UV-Vis spectra of MB and MO had maximum absorbance wavenumbers at 465 and 663 nm, respectively. When the GO-Fe₃O₄/PAA/Ag nanocomposite was added, the reaction proceeded quickly and the MB and MO color of dye solutions gradually diminished and eventually became colorless, indicating that the catalytic reduction had occurred at the nanocatalyst surface. This discoloration was monitored by UV-Vis spectrophotometry at regular intervals. The reduction spectra of MO dye for 1, 2 or 3 mg of nanocatalyst are shown in Figure 7. The characteristic absorption peak for MO is at 465 nm, and this disappears after 120 s (1 mg), 60 s (2 mg), and 40 s (3 mg) as a result of MO reduction by GO-Fe₃O₄/PAA/Ag nanocomposite in the presence of NaBH₄. Because of the large excess of NaBH₄, the reduction rate appears to be independent of the amount of NaBH₄. Hence, a pseudo-first-order kinetic equation can be used. The apparent rate constants for the MO reduction are 23 × 10⁻³ s⁻¹ (1 mg), 26 × 10⁻³ s⁻¹ (2 mg), and 77 × 10⁻³ s⁻¹ (3 mg),

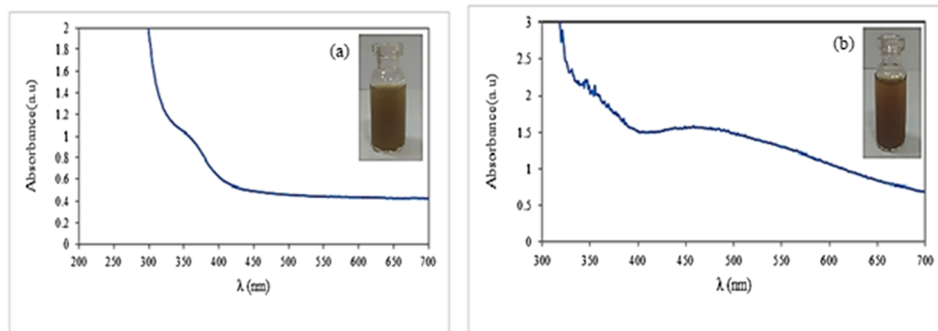


FIGURE 5 UV-Vis spectra of (a) *E. comadulensis* leaf extract and (b) green synthesized Ag NPs using *E. comadulensis* leaf extract

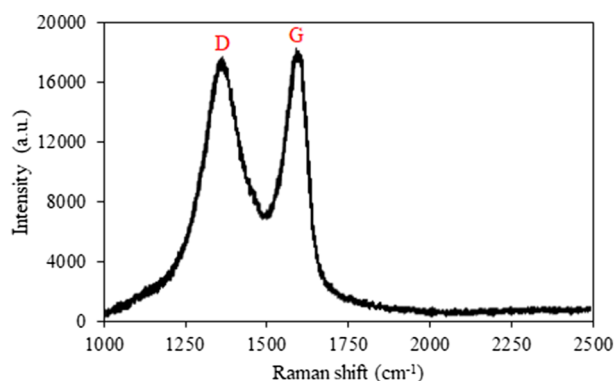


FIGURE 6 Raman spectrum of GO

calculated from the slope of the $\ln(A_t/A_0)$ versus time plot in Figure 7.^[45]

The reduction of MB with NaBH_4 was also used as another model reaction to examine the catalytic activity of the GO- $\text{Fe}_3\text{O}_4/\text{PAA}/\text{Ag}$ nanocomposite. The progress of the catalytic reduction of MB can be observed by following the decrease in absorbance at λ_{max} of MB with time. The reaction rate constants were calculated to be $38 \times 10^{-3} \text{ s}^{-1}$ (1 mg), $150 \times 10^{-3} \text{ s}^{-1}$ (2 mg), and an extremely fast reaction for 3 mg catalyst (reaction time < 10 s), for which we could not calculate the apparent rate constant (Figure 8).

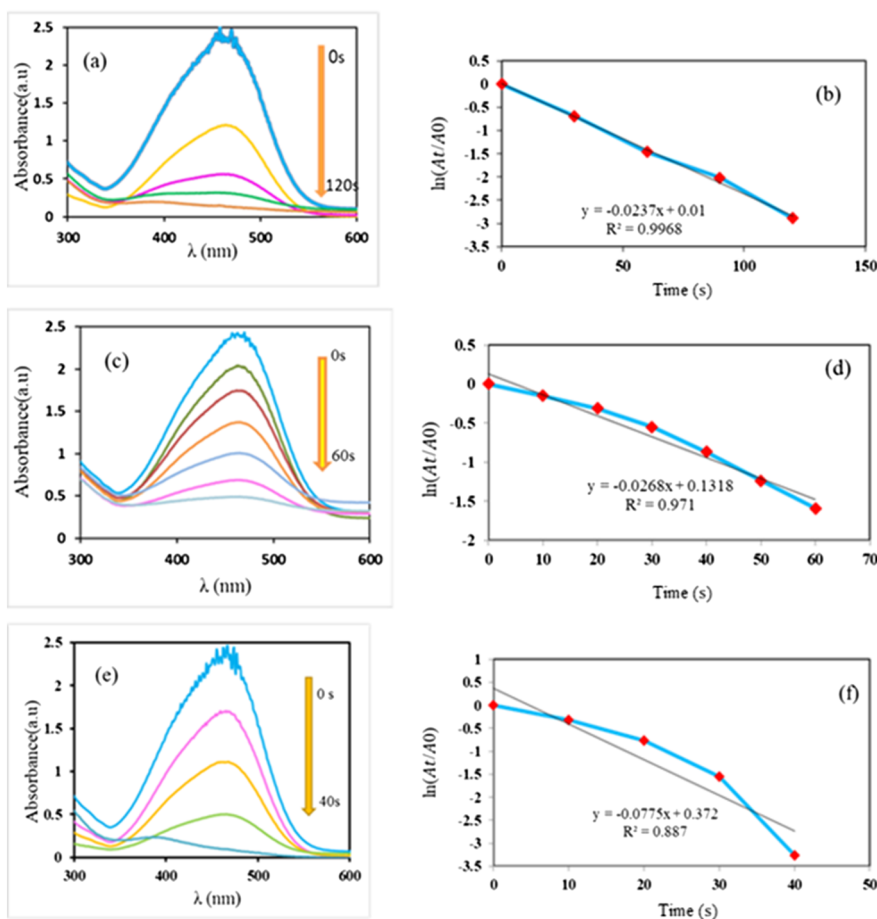
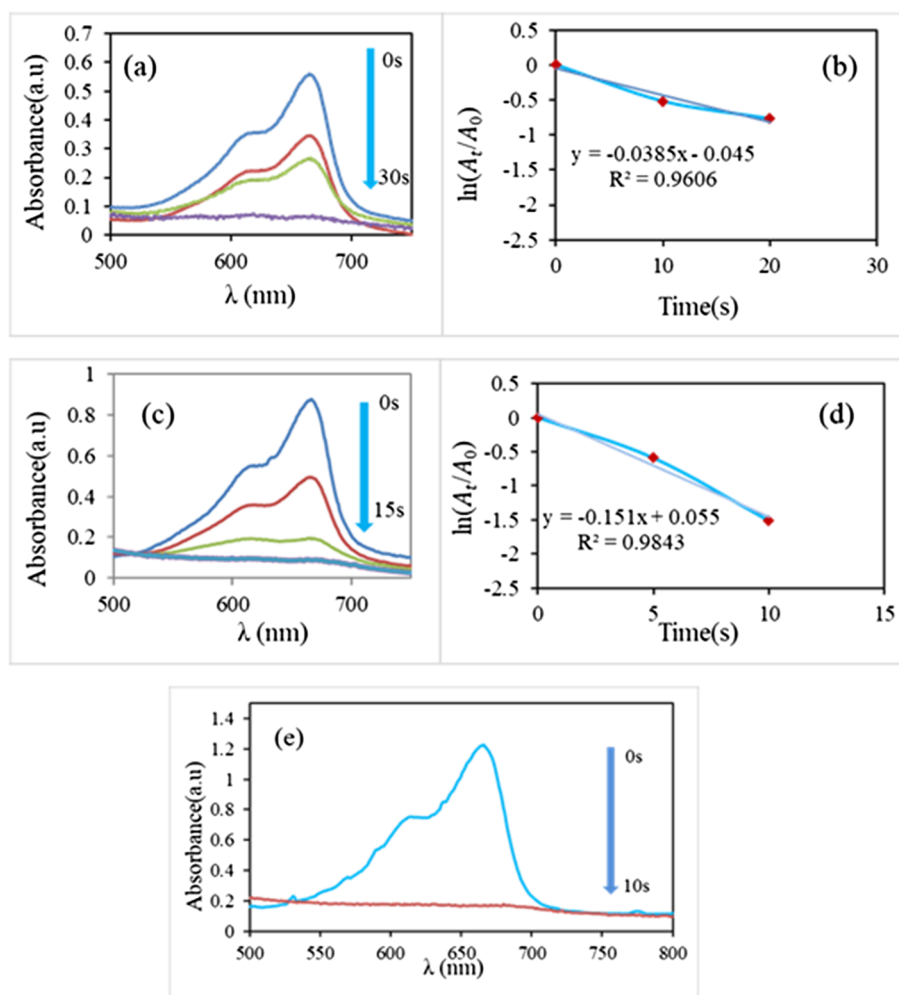


FIGURE 7 The UV-Vis spectra and apparent rate constants of MO dye in the presence of 1 mg (a and b), 2 mg (c and d), and 3 mg (e and f) of nanocatalyst

FIGURE 8 The UV–Vis spectra and apparent rate constants of MB dye in presence of 1 mg (a and b) and 2 mg (c and d) of nanocatalyst, and (e) the UV–Vis spectrum for 3 mg of nanocatalyst



To recognize the performance of the synthesized nanocatalyst, it is essential to find the turnover frequency (TOF) and the turnover number (TON) of the nanocatalyst. In heterogeneous catalysis, TON is equal to the number of reactant molecules converted into products, so the amount of nanocatalyst used is 1 g. The TOF is TON/time. Since the concentration of both reactants

(MB and MO) was 2.4×10^{-5} M and the catalyst amount was kept at 2 g/L, the calculated TOFs of the catalyst for the MB and MO are 1.4×10^{18} and 5.6×10^{19} molecules·mg⁻¹·s⁻¹, respectively, and the calculated TON of the catalyst for the MB and MO is 8.4×10^{20} molecules/mg.

To demonstrate benefit of this work, our catalyst was compared with that used in other published methods.^[24–29] Some results for the reduction of MO and MB are summarized in Table 1. These results show that GO-Fe₃O₄/PAA/Ag is a more effective catalyst with respect to the rate constant than previously reported catalysts. Moreover, the synthesized nanocatalyst was stable, recyclable, and exhibited good catalytic activity.

TABLE 1 Comparison of reduction reactions of MB and MO in the presence of GO-Fe₃O₄/PAA/Ag catalyst

Dye	Catalyst	Rate constant	Reference
MB	Ag/PSNM-3	0.134 min ⁻¹	[46]
	Au NPs	0.241 min ⁻¹	[47]
	PAA-modified Tm/Ag	0.077 min ⁻¹	[48]
	GO-Fe ₃ O ₄ /PAA/Ag	0.15 s ⁻¹	This work
MO	Pt NPs	4.145 min ⁻¹	[49]
	Au NPs	0.0017 s ⁻¹	[50]
	C@Fe MOF	0.92 min ⁻¹	[51]
	GO-Fe ₃ O ₄ /PAA/Ag	0.077	This work

4 | CONCLUSION

A novel magnetically recoverable GO-Fe₃O₄/PAA/Ag nanocatalyst was synthesized by the adsorption-reduction of Ag⁺ ions on GO-coated Fe₃O₄/PAA. The nanocatalyst

could be easily recovered from the reaction solution using an external magnet. We showed that the catalytic reduction of MO and MB with borohydride in the presence of GO-Fe₃O₄/PAA/Ag nanocomposite can be represented in terms of the Langmuir–Hinshelwood model, which assumes the adsorption of both reactants on the nanocatalyst surface. Furthermore, the GO-Fe₃O₄/PAA/Ag nanocatalyst was observed to be very stable with high catalytic activity even after eight cycles. This work may be useful in developing a process for the reduction of MB and MO in aqueous solution under mild conditions.

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