

■ Catalysis

Au Nanoparticles Immobilized in Fe₃O₄/SBA-16 Functionalized Melamine- α -Chloroacetic Acid as a Recoverable Nanocatalyst for Reduction of Dye Pollutants in Water

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Fe₃O₄/SBA-16-functionalized melamine- α -chloroacetic acid group-supported Au nanoparticles are found to serve as heterogeneous and recyclable materials for the reduction of organic dyes such as Methylene Blue (MB) and Methyl Orange (MO) in aqueous solutions. A mesoporous structure with an average pore size of 3.39 nm and a surface area of 376 m² /g was characterized with the transmission electron microscopy (TEM), X-ray diffraction (XRD) patterns, scanning electron micro-

scopy (SEM), small angle x-ray scattering (SAXS), and FT-IR. Moreover, the specific surface area of the structure was determined using the Braunauer-Emmett-Teller (BET) method. The dye reduction activity of Fe₃O₄/SBA-16-MH-Au was investigated by reducing the MB and MO. The results demonstrated that the nonporous silica (NPS) was an effective system with high catalytic activities and had a prospective application for catalytic MB and MO.

Introduction

Structural characteristics such as unchanging and tunable nanometer cavity dimension, easy surface multi-modification, high stability and surface area, and large pore volume of mesostructures silica have an interesting architecture because they are diverse in special applications for example in catalysis, drug delivery system (DDS), adsorption, sensors and separation.^[1–4] There is much works appropriated to the synthesis of mesoporous silica, which present fewer problems of obstruction pores.^[5–8] Among them, SBA-16 (Santa Barbara Amorphous-16) is individually interesting because its Im3 m symmetry with the face-centered cage-type cubic structure is expected to be much superior to other hexagonally structural mesoporous silica.^[9–11] For the past decades, preparing nanocatalysts using eco-friendly methods with good catalytic activity, excellent yield and easy separation process has made them suitable for industrial and environmental purposes. The easy separation action of catalysts using a magnet from reaction mixtures has attracted much attention compared to filtration as it causes the catalyst very little loss.^[12–17] The composition of magnetite nanoparticles (Fe₃O₄-NPs) with mesoporous silica as a nanocatalyst has been extensively studied. The principle for the design of nanocomposites lies in the combination of magnetic properties with the textural and morphological features of mesoporous silica.^[18] Accordingly,

mesoporous silica can be modified and functionalized in many methods, upon request, for the purpose of various applications.^[19] Organic modification of this nanostructure has been broadly studied in biomedicine, separation, adsorption and catalysis.^[20–25] Contaminate water in the over the last few years has been an attention for researchers. This is because the major source of contaminated water is colored wastewater resulting from dye,^[26] pigment,^[27] textile,^[28] food processing,^[29] paper,^[30] and leather tanning^[31] industries. The discharge of color pollutants from these industries has endangered human, photosynthesis cycle in plant, aquatic and animal's life. Due to their complex structure, they cannot be easily decomposed and not biodegradable. In addition, the produced intermediates are also toxic.^[32–36] Methyl orange (MO) is in the acid/anion category, and has been increasingly used in the textile, pharmaceutical and paper industries. Moreover, methylene blue is a common dye in wood, silk and cotton. The MB and MO are known as toxic compounds and therefore should be reduced to harmless materials.^[37] SBA-16 is more porous than SBA-15, and the organic functional groups of the catalyst easily fall within its pores, giving it an excellent catalytic performance. Following our previous works in preparation of nanocatalysts^[38–40] and nanocomposites to degrade and eliminate organic dyes,^[41,42] the current study focused on the synthesis of Fe₃O₄/SBA-16-MH-Au magnetic mesoporous (MMS) nanocatalysts by grafting of melamine bearing α -chloroacetic acid groups on SBA-16 and subsequent deposition of Au nanoparticles. The nanocatalyst was shown an excellent magnetic property that can be easily separated from the reaction mixture by using of an external magnet. Thus, the impact of the catalyst performance was investigated on the degradation of organic dyes such as the MB and MO. And also the leaching test was done that shown this catalyst has good

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stability. So the prepared catalyst has a potential application in industry.

Results and Discussion

The synthesis of the prepared nanocatalyst was confirmed using the TEM, XRD, SEM, SAXS, and FT-IR as well as the BET's test.

The structure of the $\text{Fe}_3\text{O}_4/\text{SBA-16-MH-Au}$ magnetic mesoporous was obtained using the XRD analysis, as shown in Figure 1. Moreover, the analysis was performed with the X'Pert

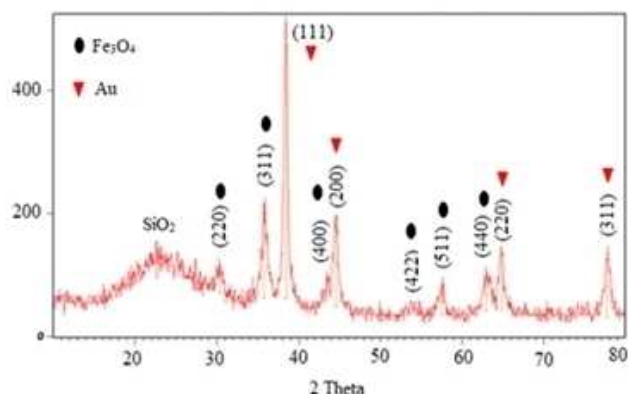


Figure 1. The XRD pattern of $\text{Fe}_3\text{O}_4/\text{SBA-16-MH-Au}$.

high score software. In this figure, the peaks at 2θ angles of 30.51° , 35.76° , 43.11° , 53.32° , 57.2° , and 61.90° were respectively related to the (220), (311), (400), (422), (511) and (440) reflection planes of Fe_3O_4 crystals with the cubic spinel structure. The peaks in $2\theta = 38.5^\circ$ (111), 44.1° (200), 64.83° (311) and 77.85° (220) were related to the Au NPs, confirming the formation of their crystalline structure. The peaks observed in the 2θ angle of 23° confirmed the amorphous structure of SiO_2 .^[38,40]

TEM images of $\text{Fe}_3\text{O}_4/\text{SBA-16-MH-Au}$ are demonstrated in Figures 2a and b. These images introduced a cubic structure of the SBA-16 mesoporous with I3ad space group. The black grain in the image showed the presence of Au and Fe_3O_4 NPs in the final structure. Moreover, the images demonstrated that the Fe_3O_4 and Au particles were uniformly stabilized on the surface and inside the cavities of the SBA-16 nanostructure.

The morphology of the synthesized nanocatalyst was shown by using SEM. Figure 3a shows the SEM image of the SBA-16 nanostructure. According to these images, spherical and homogeneous particles were obtained with a minimum particle size of 27.51 nm and maximum particle size of 38.60 nm after calcination and template removal. The SEM images of $\text{Fe}_3\text{O}_4/\text{SBA-16-MH-Au}$ in Figure 3b confirmed the spherical, homogeneous and uniform distribution of the particles with a minimum particle size of 21.98 nm and maximum particle size of 31.31 nm.

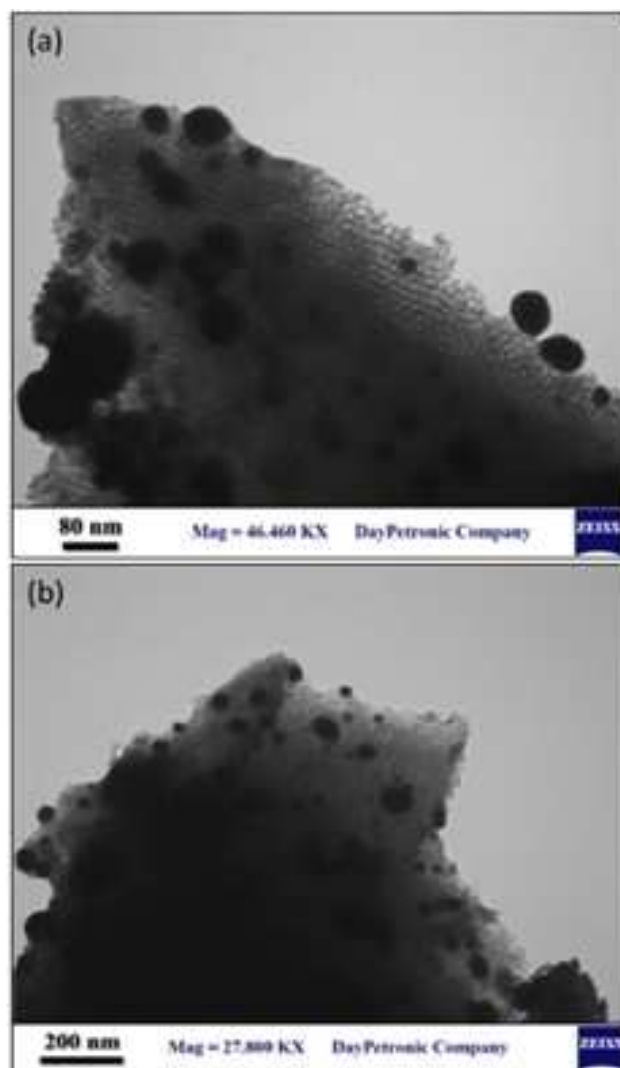


Figure 2. (a) and (b) TEM images of $\text{Fe}_3\text{O}_4/\text{SBA-16-MH-Au}$.

The SAXS analysis was used to distribute the particle size and pore in the SBA-16 and $\text{Fe}_3\text{O}_4/\text{SBA-16-MH-Au}$ samples. Figure 4a displays the SBA-16 mesoporous SAXS data after removing the template. In this pattern, one broad peak at around $2\theta = 0.99^\circ$ was observed, corresponding to d_{100} reflection. Moreover, the SAXS explanatory analysis confirmed the mesoporous nature of the products. Figure 4b shows the SAXS pattern of $\text{Fe}_3\text{O}_4/\text{SBA-16-MH-Au}$. It can be observed that the presented pattern is similar to the SAXS pattern of SBA-16 and peak of about $2\theta = 0.99^\circ$, which is due to d_{100} reflection. However, due to the connection of the functional groups on the mesoporous surface, the promotion of the SAXS pattern has slightly decreased. With the functionalized and treated SBA-16 in the presence of α -chloroacetic acid, Fe_3O_4 and Au nanoparticles, considerable decrease is observed in the intensities of the peaks resulting as the changes in the wall thickness or diminishing of scattering contrast between the channel walls of the mesosilicate. It might be also due to the

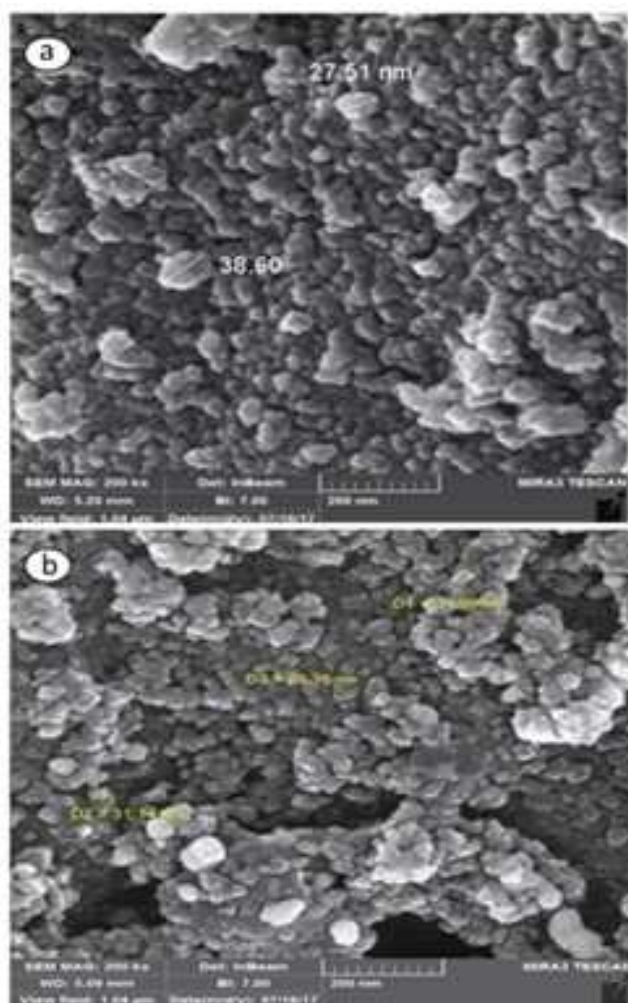


Figure 3. (a) SEM images of SBA-16 and (b) $\text{Fe}_3\text{O}_4/\text{SBA-16-MH-Au}$.

presence of some adsorbed Fe_3O_4 and Au particles within the channel structure.

Figure 5 revealed the FT-IR spectra of the Fe_3O_4 (a), SBA-16 (b), SBA-16-Cl (c), SBA-16-Melamine (d) and $\text{Fe}_3\text{O}_4/\text{SBA-16-MH-Au}$ (e). In Figure 5a, three bands at 585 cm^{-1} , 635 cm^{-1} and 1635 cm^{-1} can be observed from spectrum of Fe_3O_4 nanospheres. Two bands at 585 cm^{-1} and 635 cm^{-1} are corresponding to the vibration of the Fe–O bonds. The O–H bending group were appeared at 1635 cm^{-1} .^[43,44] The adsorption bands at 960 and 1100 cm^{-1} are indicative of the siloxane (–Si–O–Si–), symmetric and asymmetric vibration.^[45] The peak at 3494 cm^{-1} attributed to the Si–OH stretching band of the silanol groups, and a characteristic peak at 1650 cm^{-1} is indicated to the deformation vibrations of adsorbed water molecules in the FT-IR spectrum of SBA-16, as illustrated in Figure 5b.^[42] The FT-IR spectrum of SBA-16-Cl shows the stretching band of C–Cl observed at 540 and 785 cm^{-1} . Moreover, three absorption bands at 960 cm^{-1} , 1100 cm^{-1} , and 3485 cm^{-1} are respectively related to the –Si–O–Si– symmetric and asymmetric vibration as well as the Si–OH stretching bond, as indicated in Figure 5c. In Fig. 5d, the functional groups of SBA-16-Melamine have been

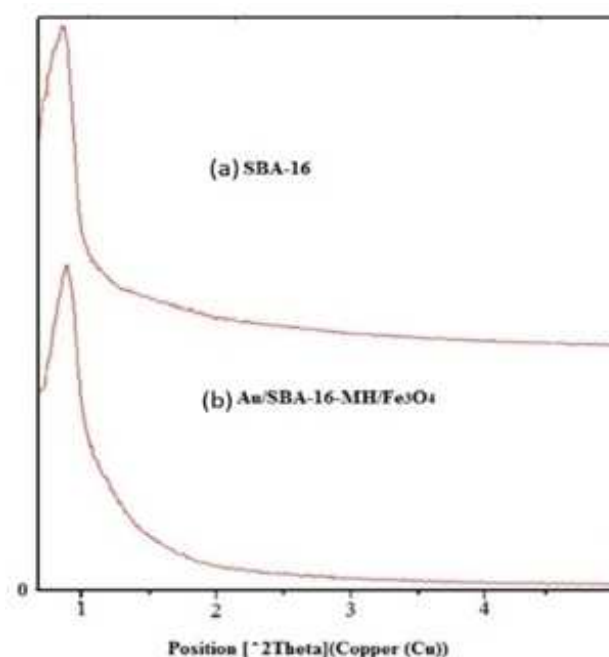


Figure 4. (a) SAXS pattern of SBA-16 and (b) $\text{Fe}_3\text{O}_4/\text{SBA-16-MH-Au}$.

investigated and utilized by the FT-IR spectroscopy. It is well documented that the band near 3500 cm^{-1} is dedicated to overlapping of the stretch N–H and O–H groups. The small absorption band at 3100 cm^{-1} represents the aromatic stretch =C–H group. The FTIR spectrum of this compound revealed a bond in the aromatic C=C stretch region of 1600 cm^{-1} and 1502 cm^{-1} . A band is indicated at 1640 cm^{-1} in the FT-IR spectrum of $\text{Fe}_3\text{O}_4/\text{SBA-16-MH-Au}$ in Figure 5e. The peak at 1640 cm^{-1} is assigned to the C=O. The overlapping of the hydroxyl group was shown at 3500 cm^{-1} due to water absorption in the Si–OH and N–H stretch groups. The typical peaks at 960 cm^{-1} and 1100 cm^{-1} belong to the stretching vibration of the Si–O–Si bonds in the silica mesh. Moreover, the absorption peak at 3100 cm^{-1} belongs to the aromatic stretch group =C–H.^[45,46]

The BET analyses of the SBA-16 are demonstrated in Figure 6 a. It is clear that both the samples show the typical H_2 hysteresis loop with IV type isotherm, suggesting their bottle-like pore structures. Figures 6b and c present the mesoporous hybrid material. The BET of the SBA-16-melamine and $\text{Fe}_3\text{O}_4/\text{SBA-16-MH-Au}$ nanostructures were similar, but had a lower surface than the SBA-16 isotherm. The volume of nitrogen gas adsorption was diminished due to the organic compound linkage to the SBA-16 surface. The organic group melamine interaction with SBA-16 causes to increase the mean size of the particle, thereby creating a pore. Table 1 summarizes the structural data such as surface area, total pore volume, and pore size. Diminishing these parameters ($\text{SBET} (\text{m}^2\text{ g}^{-1})$, $V (\text{cm}^3\text{ g}^{-1})$ and $\text{DBJH} (\text{nm})$) suggests that organic functional groups are located within the pores

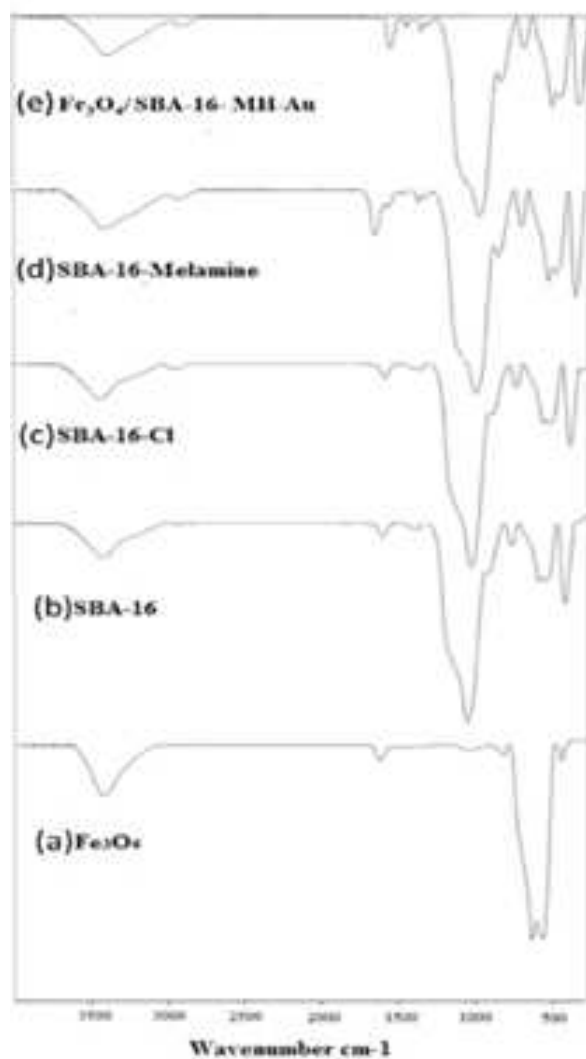


Figure 5. (a) FT-IR spectroscopy of Fe_3O_4 , (b) FT-IR spectroscopy of SBA-16, (c) FT-IR spectroscopy of SBA-16-Cl, (d) FT-IR spectroscopy of SBA-16-Melamine and (e) FT-IR spectroscopy of SBA-16-MH.

Catalytic activity

The reduction of dye contamination is carried out to determine for efficiency and catalytic activity of metallic nanoparticles.^[35] Thus, the reason for the reduction of the MB and MO dyes in water in the presence of sodium borohydride (NaBH_4) as reducing agent with $\text{Fe}_3\text{O}_4/\text{SBA16-MH-Au}$ as catalyst was explored. Effects of several different dosages of the nanocatalyst and the time dependence of the efficiency were considered using the UV-Vis spectroscopy. The diagram of absorbance versus reaction time for reducing the MB and MO with NaBH_4 are shown in Figure 7. The rate of the catalytic activity of the catalyst was calculated by the kinetic studies, which results in the show with an increase in the amounts of the nanocatalyst, the catalytic activity was increased. This can be due to the existence of the catalytic active sites according to the increase in the amounts of the nanocatalyst. Therefore, it

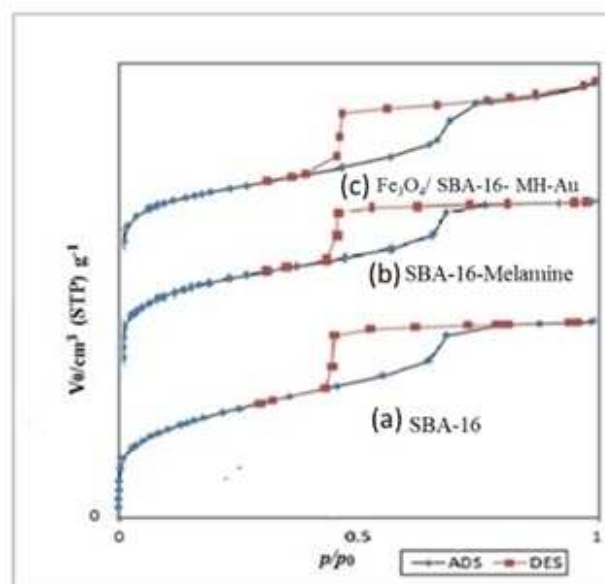


Figure 6. The adsorption and desorption isotherm of (a) SBA-16, (b) SBA-16-melamine and (c) $\text{Fe}_3\text{O}_4/\text{SBA-16-MH-Au}$.

Table 1. Textural parameters for SBA-16, SBA-16-Melamine, $\text{Fe}_3\text{O}_4/\text{SBA-16-MH-Au}$

Material	S_{BET} ($\text{m}^2 \text{g}^{-1}$)	V ($\text{cm}^3 \text{g}^{-1}$)	D_{BJH} (nm)
SBA-16	918.23	0.7784	3.39
SBA-16- Melamine	660.92	0.5607	3.39
$\text{Fe}_3\text{O}_4/\text{SBA-16-MH-Au}$	376	0.4474	4.76

is observed increases gradually the apparent rate constant (k_{app}) of the reduction reaction of dyes. The k_{app} for different amounts of nanocatalyst were obtained from the diagram slope of $\ln(A_t/A_0)$ versus reaction time. The k_{app} increases considerably as the concentration of the nanocatalyst increases from 0.001 g (as shown in Figures 7a and 7c for the MB and MO, respectively) to 0.002 g (as indicated in Figures. 7b and 7d for the MB and MO, respectively). After $\text{Fe}_3\text{O}_4/\text{SBA-16-MH-Au}$ was added, the absorbance intensity disappeared completely within 75 s (0.001 g catalyst), 40 s (0.002 g catalyst) for the MB, 100 s (0.001 g catalyst), and 60 s (0.002 g catalyst) for the MO (Figure 7). The solution of MO and MB became colorless after $\text{Fe}_3\text{O}_4/\text{SBA-16-MH-Au}$ was recovered by an external magnet. The reaction kinetics of reduction reactions can be easily monitored from the time-dependent absorption spectra. Since the concentration of NaBH_4 was in excess, the reduction reaction can be supposed to follow the pseudo-first order kinetics reaction. The plot of $\ln(A_t/A_0)$ versus time (s) is linear in the presence of $\text{Fe}_3\text{O}_4/\text{SBA-16-MH-Au}$, as shown in Figure 8. The k_{app} of reactions were obtained from the slope of the plots were 0.026 s^{-1} and 0.046 s^{-1} (for the MB as illustrated in Figures 8a and 8b) and 0.019 s^{-1} and 0.031 s^{-1} (for the MO as shown in Figures 8c and 8d) in the presence of 1 mg and 2 mg of the catalyst, respectively.

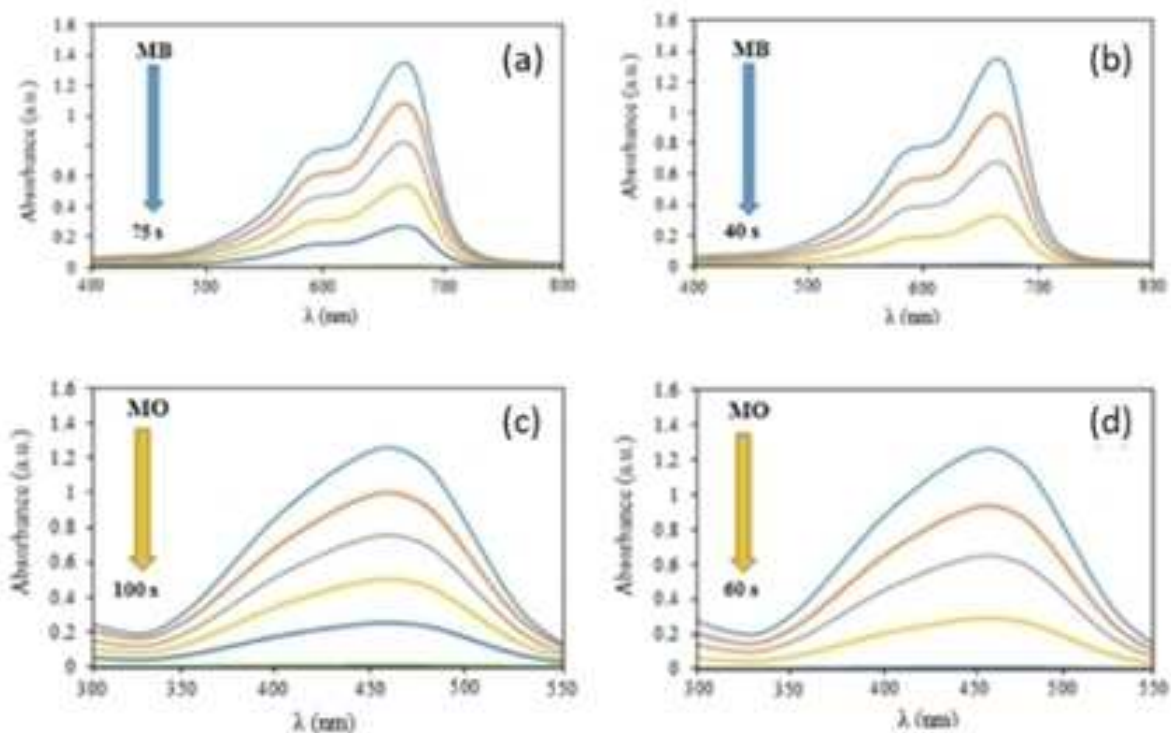


Figure 7. (a) UV-Vis spectrum for reduction of MB in 75 s, (b) UV-Vis spectrum for reduction of MB in the 40 s, (c) UV-Vis spectrum for reduction of MO in 100 s and (d) UV-Vis spectrum for reduction of MO in 60 s.

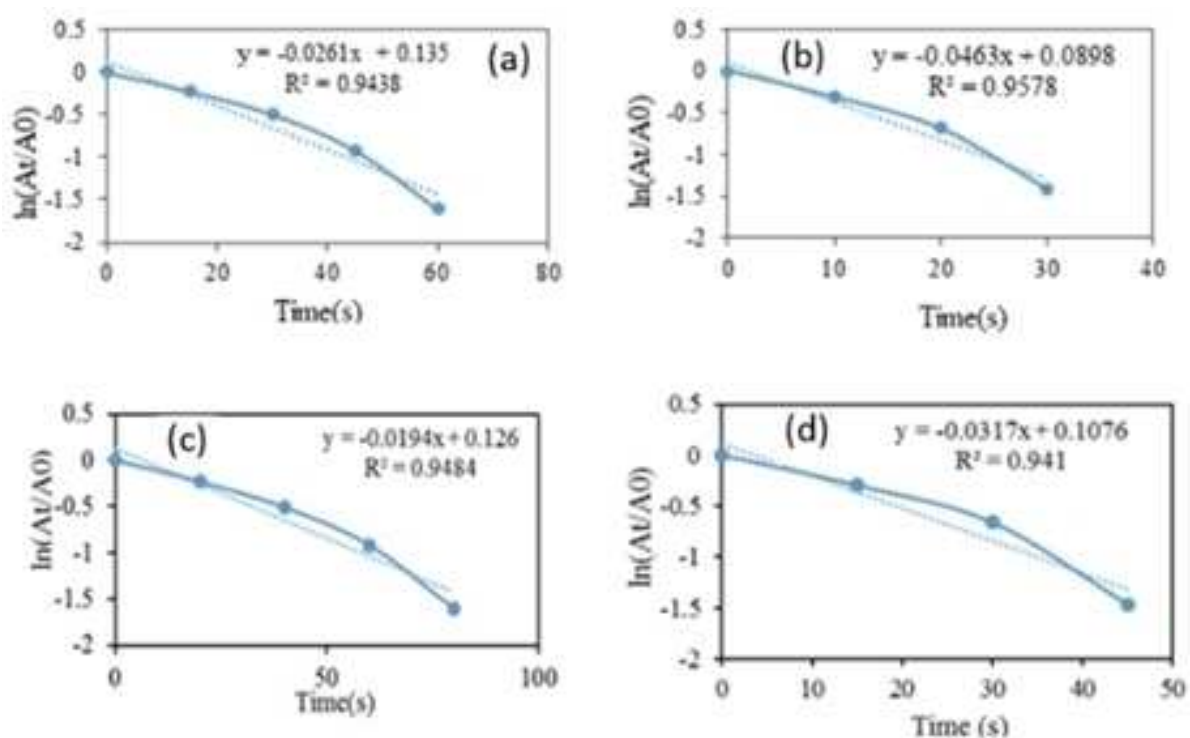


Figure 8. (a) UV-Vis spectrum for reduction of MB in 75 s, (b) UV-Vis spectrum for reduction of MB in the 40 s, (c) UV-Vis spectrum for reduction of MO in 100 s and (d) UV-Vis spectrum for reduction of MO in 60 s.

The leaching test was done, according to the results of ICP analysis, the amount of Au was determined 0.15 mmol/g that shown the prepared catalyst has good stability. The performance of the Fe₃O₄/SBA-16-MH-Au nanocatalyst with other catalysts that have been reported to catalyse reduction of the MB dye. As the k_{app} values demonstrated in Table 2, the Fe₃O₄/

Table 2. Comparison of the catalytic activity of the Fe₃O₄/SBA-16-MH-Au with various catalysts reported in the literature for the reduction or degradation of MB dye.

Catalyst	k_{app}	Ref.
Fe ₃ O ₄ @Nico@Cu	0.04 s ⁻¹	[47]
Fe ₃ O ₄ /SiO ₂ -Pr-S-Ag	0.138 s ⁻¹	[48]
Fe ₃ O ₄ @SiO ₂ -Ag	0.09 s ⁻¹	[38]
α-MnO ₂	0.116 min ⁻¹	[49]
Cu@SBA-15	0.51 min ⁻¹	[50]
MnFe ₂ O ₄ @SiO ₂ @Ag	0.254 min ⁻¹	[51]
Fe ₃ O ₄ @Histidine@Ag	0.15 min ⁻¹	[52]
Fe ₃ O ₄ /SBA-16-MH-Au	0.046 s ⁻¹	This work

SBA-16-MH-Au nanocatalyst is better than mostly other catalysts to reduction of MB dye.

Conclusions

In this paper, we described the mesoporous Fe₃O₄/SBA-16-MH-Au synthesis in a simple way, which reduced hazardous dye from the aqueous solution. The morphology of the synthesized Fe₃O₄/SBA-16-MH-Au was considered by the FT-IR, XRD, BET, TEM, SAXS, TEM and SEM analyses, and the mesoporous was prepared. The results showed that the synthetic material had a good surface area (376 m²g⁻¹), simple surface modification and high catalytic activity. These three capabilities have made the reported applications to potentially reduce the organic compounds of dyes, especially the MB and MO. The k_{app} of reactions were obtained from the slope of the plots were 0.026 s⁻¹ and 0.046 s⁻¹ (for the MB) and 0.019 s⁻¹ and 0.031 s⁻¹ (for the MO) in the presence of 1 mg and 2 mg of the catalyst, respectively. The leaching test was done, according to the results of ICP analysis, the amount of Au was determined 0.15 mmol/g that shown the prepared catalyst has good stability (0.2 mmol/g was leached.).

Supporting Information Summary

The following data is provided in supporting information: The experimental section and any associated references

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Conflict of Interest

The authors declare no conflict of interest.

Keywords: Magnetic nanocatalyst · Nanoporous silica · Reduction reaction · Dye pollutants

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