



Highly efficient degradation of hazardous dyes in aqueous phase by supported palladium nanocatalyst—A green approach



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ABSTRACT

In this report, supported palladium nanocatalyst on silica coated Fe₃O₄ magnetic nanoparticles was employed as a novel catalyst towards the degradation of methyl red, methyl orange and methylene blue. The reaction between palladium nanoparticles and dissolved oxygen in acidic media produced palladium hydroperoxide, which has strong ability to oxidize the proposed dyes. The results showed that, the concentration of dyes changes exponentially with time and Langmuir–Hinshelwood model was used to describe the catalytic decolorization of these dyes in the presence of palladium nanoparticles. The efficiency of the catalyst and the effects of different variables such as initial pH, catalyst loading and temperature have been discussed in detail. Degradation sequence of dyes was further investigated by liquid chromatography–mass spectroscopy technique, which confirmed the degradation of dyes to non-toxic end products.

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Introduction

The discharge of several hazardous dyes from many textiles industries in waste water is a main reason for serious environmental problems that concern with human health and the aquatic medium due to the toxicity and the carcinogenic effect of these materials [1–4]. Therefore, the complete removal of the pollutant dyes from wastewater is necessary and subject of widespread research. For this purpose, several processes have been studied to reach partial or complete removal of pollutant compounds such as adsorption, coagulation, biodegradation, chemical or photochemical degradation and advanced oxidation processes [2–12].

As an alternative strategy, some magnetic functionalized materials have been used as photocatalysts or adsorbents in the field of water purification, owing to their high specific surface area, unique magnetic properties, chemical stability, low toxicity, ease in synthesis and especially excellent recycling ability. For example, adsorbents based on chitosan, glutaraldehyde, aminoguanidine, carbon nanotube and microporous carbon have been reported to remove dyes efficiently [13–17]. Moreover, a number of magnetic photocatalysts were fabricated using the magnetic particle and photoactive material (for instance, BiOBr@SiO₂@Fe₃O₄, SrFe₁₂O₁₉,

AgBr–TiO₂/SiO₂@Fe₃O₄ and Fe₃O₄@TiO₂) and they were successfully used for photocatalytic elimination of different organic dyes molecules [18–22]. Recently, Safavi and Momeni introduced palladium/hydroxyapatite/Fe₃O₄ nanoparticles and evaluated their catalytic activity towards the degradation of some organic dyes [23]. Their results showed high catalytic activity of palladium nanoparticles for the degradation of organic azo dyes.

Among different metal nanocatalysts, palladium nanoparticles (Pd NPs) have gained much attention due to their advantages as heterogeneous and green chemistry catalyst [24–27]. These nanoparticles show superior activity, attributed to their larger surface-to-volume ratio which enhances the contact between reactants and catalyst. Although the higher surface area provides nanoparticles with more active sites, it facilitates their agglomeration and impedes their consistent distributions. Therefore, it is essential to design an efficient catalyst having facile reusability and well-dispersion. In this regard, iron oxide magnetic nanoparticles (Fe₃O₄ MNPs) as the Pd supports have got more attentions as they can be easily recovered from the reaction mixture, simply by using an external magnet [28–32].

Recently we have demonstrated a facile method for deposition of Pd NPs on amino-vinyl functionalized silica coated Fe₃O₄ MNPs (Fe₃O₄@(A–V)-silica–Pd MNPs) [33]. The results showed that the supported Pd nanocatalyst could be simply recovered by using an external magnet and reused for several times without significant loss of its catalytic activity. In continuation of our previous work,

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herein, the catalytic performance of $\text{Fe}_3\text{O}_4@(\text{A-V})\text{-silica-Pd}$ MNPs was investigated in the degradation of some organic pollutant dyes and the effects of different variables such as initial pH, catalyst loading and temperature were studied.

Experimental

Apparatus and reagents

Iron(II) chloride tetrahydrate, iron(III) chloride hexahydrate, palladium(II) chloride, triethoxyvinylsilane (VTEOS), *n*-(2-aminoethyl)-3-aminopropyltrimethoxysilane (AEAPS), tetraethyl orthosilicate (TEOS), methyl orange, methyl red and methylene blue were purchased from Sigma–Aldrich and Merck and were used without further treatment. Ultrapure water (Millipore) was used throughout the whole experiment.

A Shimadzu 1601 PC UV–vis spectrophotometer (Kyoto, Japan) was used for recording the spectrophotometric data. The liquid chromatography–mass spectroscopy system (LC–MS) (LCMS-2010 EV Shimadzu) was equipped with Shimpack XR-ODS C18 column (5 mm × 20 mm i.d., 5 μm). The mobile phase consisted of methanol and 10 mM ammonium acetate (30/70 v/v). The flow rate of the mobile phase was 0.3 mL min⁻¹. 10 μL of sample was injected by an autosampling device. The eluent from the chromatographic column enters UV–vis detector, electrospray ionization (ESI) interface and then the mass analyzer. The ESI conditions were as follows: capillary voltage 1.5 kV, gas flow 1.5 L min⁻¹ and desolvation temperature 200 °C. The mass spectrophotometer was scanned from $m/z=100$ to 400 for recording the mass spectra.

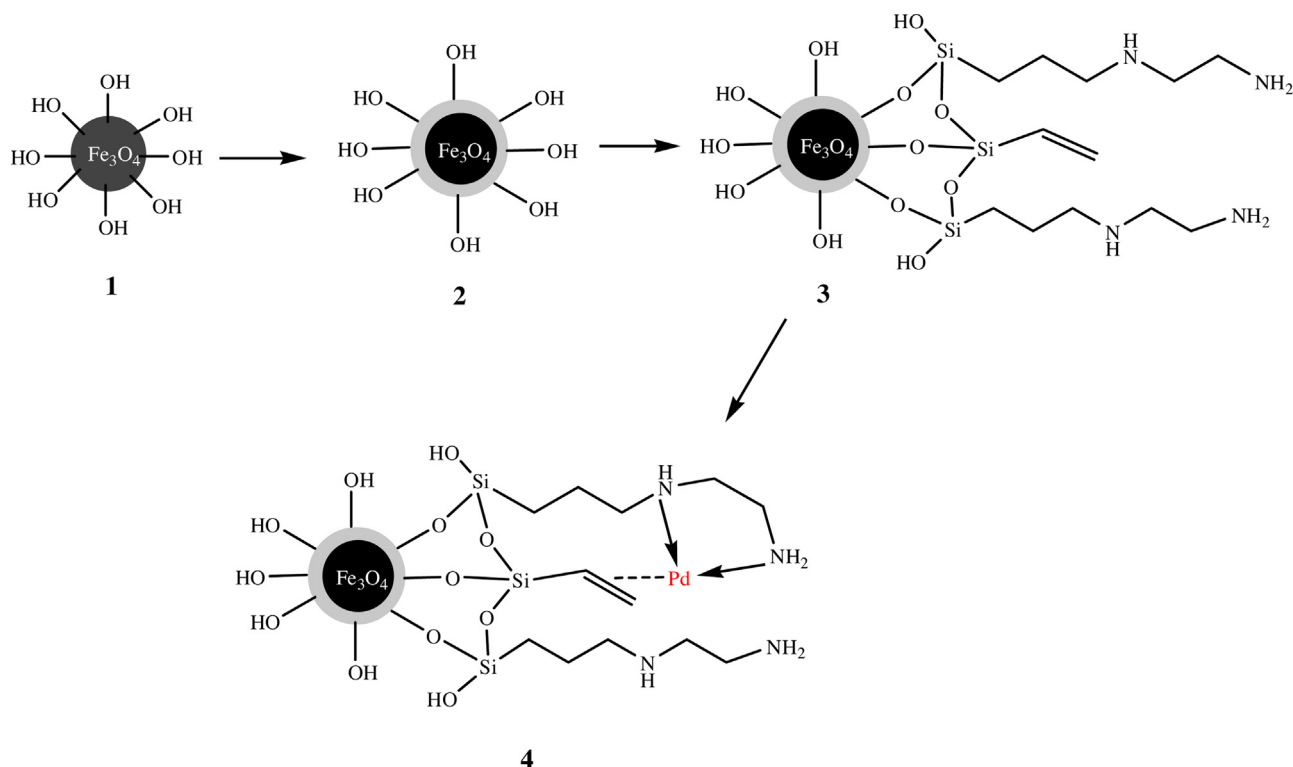
Preparation of $\text{Fe}_3\text{O}_4@(\text{A-V})\text{-silica-Pd}$ nanoparticles

$\text{Fe}_3\text{O}_4@(\text{A-V})\text{-silica}$ MNPs were synthesized via the following method [33]. 1 g of Fe_3O_4 MNPs which were prepared via the

co-precipitation method [34,35], were suspended in 1-propanol (100 mL) by sonication for 30 min at room temperature. Next, NH_4OH (25% w/w, 14 mL), H_2O (10 mL) and TEOS (3.5 mL) were added and the mixture was stirred at 50 °C for 1 h. The black precipitate was washed three times with 1-propanol and then added to a mixture of AEAPS (1.5 mL) and VTEOS (1.5 mL) in 100 mL of H_2O , followed by the addition of 0.01 M HCl (50 mL). The mixture was shaken at room temperature for 24 h. The resulted dark brown precipitate ($\text{Fe}_3\text{O}_4@(\text{A-V})\text{-silica}$ MNPs) was washed with H_2O and was kept in an oven at drying for 48 h until complete drying. In order to deposit the Pd NPs, 0.1 g of the $\text{Fe}_3\text{O}_4@(\text{A-V})\text{-silica}$ MNPs was dispersed in PdCl_2 solution (10 mL of 2 mM PdCl_2 in 1 mM H_2SO_4) and the mixture was stirred for 10 min at 80 °C to adsorb and reduce Pd ions completely. The obtained nanocomposite was carefully washed with H_2O and Ethanol and dried in an oven at 80 °C. The ICP results showed that the loading amount of Pd on $\text{Fe}_3\text{O}_4@(\text{A-V})\text{-silica-Pd}$ MNPs was 2.0×10^4 mmol mg⁻¹.

Catalytic degradation study

The catalytic activity of the $\text{Fe}_3\text{O}_4@(\text{A-V})\text{-silica-Pd}$ MNPs was investigated for the degradation of methyl orange, methyl red and methylene blue. For this purpose, 10 mL of dye solution (5 μg mL⁻¹) was transferred in to the test tube and catalytic testing was done by the addition of the catalyst to the dye solution under ambient conditions. The ultrasonic bath (Elma S180H) was used for dispersion of supported Pd NPs in the dye solution and they could be easily separated from the solution mixture by putting an external magnet into the vicinity of the tube. The decomposition of dyes was monitored spectroscopically with a UV–vis absorption spectrophotometer. The measurements were carried out in fast mode (350–700 nm in 30 s) in every 3–5 min interval. After complete degradation, the separated $\text{Fe}_3\text{O}_4@(\text{A-V})\text{-silica-Pd}$ MNPs was washed with distilled water and applied in recycling purposes.



Scheme 1. Synthesis of $\text{Fe}_3\text{O}_4@(\text{A-V})\text{-silica-Pd}$ MNPs.

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