

original article

Decolorization of synthetic wastewaters by nickel oxide nanoparticle

Roya Nateghi, Gholam Reza Bonyadinejad¹, Mohammad Mehdi Amin¹, Hamed Mohammadi²

Department of Environmental Health Engineering, Zanjan University of Medical Sciences, Zanjan, Iran, ¹Environment Research Center, Isfahan University of Medical Sciences, Isfahan, Iran, ²Department of Environmental Health Engineering, Shahid Beheshti University of Medical Sciences, Tehran, Iran

Address for correspondence:
Eng. Gholam Reza Bonyadinejad,
Environment Research Center,
Isfahan University of Medical Sciences,
Hezar Jerib Avenue, Isfahan, Iran.
E-mail: r_bonyadinejad@yahoo.com

ABSTRACT

Aims: In this study, the adsorption process using nickel oxide nanoparticles was studied in a laboratory scale for wastewater treatment containing mono azo Orange II dye.

Materials and Methods: The effect of various parameters such as initial dye concentration, pH, contact time and different concentrations of nickel oxide was investigated. The adsorption experiments were done with different concentrations of dye in a solution using a specific amount of nickel oxide nanoparticle with different pH values and a agitator speed of 100 revolutions per minute (rpm) for about 2 hours. Samples were centrifuged and the concentration of each dye was determined by ultraviolet (UV)-Vis spectrophotometer (DR 5000).

Results: Based on obtained results, the optimum pH range to dye removal is acidic pH, and under lab conditions, 0.6 g/L of nickel oxide can completely remove the 50 mg/L of dye. Although increasing the dye concentration resulted in decreased process efficiency, up to 100 mg/L of dye concentration, considerable efficiency was obtained.

Conclusions: The data showed that the nickel oxide nanoparticles could be used as an efficient adsorbent for decolorization of azo dyes.

Key words: Adsorption, azo dye, decolorization, nickel oxide nanoparticles

INTRODUCTION

About one million tons of different chromogens are synthesized annually worldwide, of which 10-15% remain in textile wastewaters after application. Azo dyes represent the largest class of dyes listed in general category of dyes (60-70% of all dyes).^[1,2] Azo dyes contain at least one chromogenic

factor of nitrogen-nitrogen double bond (-N = N-) called azo group with one or more aromatic groups^[3-5] and their discharge into natural receiver waters such as lakes and rivers decreases light transmission, soluble O₂ levels and increases chemical oxygen demand (COD), thus disturbing marine life. Researchers have found that some dyes can produce carcinogenic and mutagenic aromatic amines during the process of degradation.^[6] Without adequate and appropriate treatment, these compounds can remain in the environment for a very long time and permanently.^[7] There are different methods for decolorization of textile wastewater including different physical, chemical and biological processes. Exploitation and maintenance cost of most of these processes is high, so using them for treatment of wastewaters containing dye is not suitable and economical for many countries.

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Physical adsorption is an effective and practical method for a rapid decrease in the concentration of dissolved dyes in wastewater. Significant researches have been done regarding adsorption of dyes by adsorbents such as activated carbon, silica, clay, natural and artificial polymers, etc. Nickel oxide (NiO) is considered as a good adsorbent due to its chemical and magnetic properties.^[8] Nickel oxide powders with same sizes and good dispersion are used in different fields such as producing films, magnetic materials, ceramic, heterogeneous catalytic materials, alkaline batteries, electrochrom, etc.^[9] Nickel oxide as an effective catalyst has been used for oxidation of a wide range of organic compounds.^[10,11] In a study conducted by Sa'adatjoo *et al.* (2008) on red basic dye 46 at initial dye concentration of 15 parts per million (ppm), using adsorbent of hardened pieces of Portland white cement, under the condition of the pH value of 10 and 10 g/L adsorbent concentration, more than 80% decolorization was achieved after 120 minutes contact time.^[12] Asgari and Ghanizade (2009) could completely remove 200 mg/L methylene blue dye after 2 hours contact time using 1 g/L bone ash.^[13] In a research conducted by Arami *et al.* (2009), about 76% of basic blue dye 41 with the concentration of 50 mg/L were removed using 0.8 g/L Tamarind shell at the pH value of 8.^[14] Song *et al.* (2009), using nickel oxide nano-sheets, could completely remove 100 mg/L reactive dye at acidic pH after 6 hours.^[8] This study aims to investigate the efficiency of Nickel oxide nanoparticles as one form of adsorption processes for decolorization of Orange II azo dye.

MATERIALS AND METHODS

The present study was designed as an experimental-interventional laboratory scale study in a batch mode. Nickel oxide nanoparticles (NiO) and Orange II dye were purchased from Sigma – Aldrich company. Dye was used without purification. Dye chemical structure is presented in Figure 1. Other chemicals were purchased from Germany Merck company. pH meters (WTW), centrifuge (SIGMA) and DR5000 spectrophotometer devices were used. At first, the mother solution of Orange II dye with concentration of 1000 mg/L was prepared. Then using distilled water, standard solutions with concentrations of 5, 10, 15, 20, 25 and 30 mg/L were extracted from this solution. After scanning the absorption range of each dye, the wavelength of maximum absorption (λ_{max}) for Orange II dye was achieved at 500 nm.

Next, standard colors' absorption, with specified concentrations was read by adsorption spectrophotometer (DR 5000) and standard curve of each color was drawn and the concentration of each dye at different stages was determined by equation elicited from the standard curve. Figure 2 shows the standard calibration curve of Orange II dye. As nickel oxide nanoparticles are completely insoluble in water, we can separate them completely from wastewater, and they also have a high physical resistance compared with activated carbon, and hence were chosen as adsorbent ion

our study. Radiographs/X-rays (XRD) and scanning electron microscopy (SEM) techniques were used to determine the structural characterization of nickel oxide nanoparticles. This study aimed to investigate the effect of various parameters such as initial dye concentration, pH, contact time and different concentrations of nickel oxide. Hence, adsorption experiments were done with different concentrations of dye in solution, and using a specific amount of nickel oxide nanoparticles, and with different pH values and agitator speed of 100 rpm for about 2 hours. Samples were centrifuged and the concentration of each dye was determined by UV-Vis spectrophotometer (DR 5000).

RESULTS

XRD and SEM techniques were used to determine the structural characterization of nickel oxide Nanoparticles. Figures 3 and 4 show images of XRD and SEM of nickel oxide nanoparticles respectively. The value of accessible surface is determined about 50 m²/g according to calculations done by Sigma company, and according to the experiments, the average size of these powders is estimated less than 10 nm. For this research, at first, the effect of used dosage of nickel oxide nanoparticles on decolorization at fixed concentration of 50 mg/L dye, neutral pH and different concentrations of adsorbent was investigated. The results are shown in Figure 5. The results indicate that about 81% of the Orange II dye has been removed by 0.6 g/L adsorbent after 30 minutes contact time. Therefore this value was used as the optimum amount for examination of dye decolorization process.

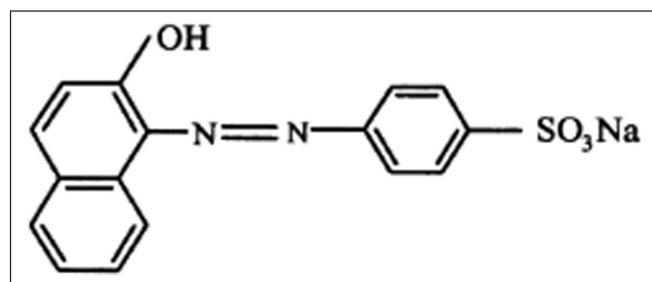


Figure 1: Chemical structure of orange dye 2

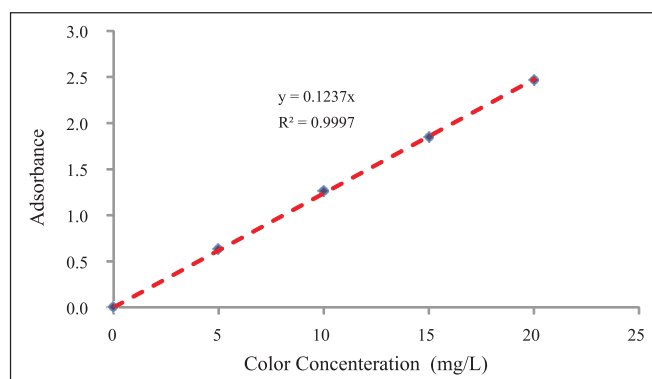


Figure 2: Standard absorption curve for measuring Orange II dye in unknown samples

Studying the effect of pH value in the adsorption process as depicted in Figure 6 shows that the adsorption capacity of nickel oxide nanoparticles has decreased with the increase of pH. In other words, the highest decolorization efficiency (about 99.5%) is obtained at pH value of 3. To investigate the effect of initial dye concentration in the adsorption process, different dye concentrations of 100-75-50-25 mg/L respectively were studied. To investigate this part of the research, the best effect point of pH and the optimal concentration of NiO obtained from the previous steps, along with different dye concentrations were used. As Figure 7 shows, decolorization speed decreases with the increase of initial dye concentration from 25 mg/L to 100 mg/L.

DISCUSSION

The results of XRD and SEM experiments indicate that nickel oxide powders have non-crystallized structure and pores and suitable surfaces for adsorption. Regarding NiO surface having periodic polar single-layers of O₂ Ions and Nickel cations, a strong electrostatic field perpendicular to NiO surface is created which may cause the adsorption of the dye molecules in the solution, and help in the decolorization of the surface area.^[8] The results of investigation show the effect of adsorbent value on adsorption process efficiency. Figure 5 shows that efficiency increases with decreasing adsorbent and this may be due to increasing adsorbent surface and more accessibility to adsorption site. These results are confirmed with studies done by other researchers.^[12,14] According to Figure 5, about 81% of Orange II dye has been removed in 30 minutes and then decolorization efficiency has become fixed. Generally, adsorption capacity and decolorization efficiency by adsorbent are directly related to contact time, hence the surface adsorption increases with increasing contact time due to increased possibility of contacting dye molecules with adsorbent surface; however, reaches a fixed value at a specific time. In this moment, the amount of adsorbed dye is in a dynamical balance with the amount of not adsorbed dye, and after this time, the amount of dye adsorption by adsorbent remains approximately fixed.^[14]

The value of solution's pH has an important role in the whole process and adsorption capacity, and affects not only the adsorbent surface charge but also the ionization degree of materials in solution, and the also separation of agent groups in active places of adsorbent and the solution's chemistry. Many researchers believe that pH has an important role in electrostatic attraction between the adsorbent and the target.^[8,14] The results indicate that the adsorption capacity has decreased with increasing pH. In the best conditions, i.e. at pH value of 3, about 99.5% of the dye has been removed [Figure 6]. Higher uptakes of dye obtained at lower pH values may be due to the electrostatic attractions between the negatively charged dye anions and positively charged adsorbent surface.^[8] At lower pH, more protons will be available due to which electrostatic attraction between

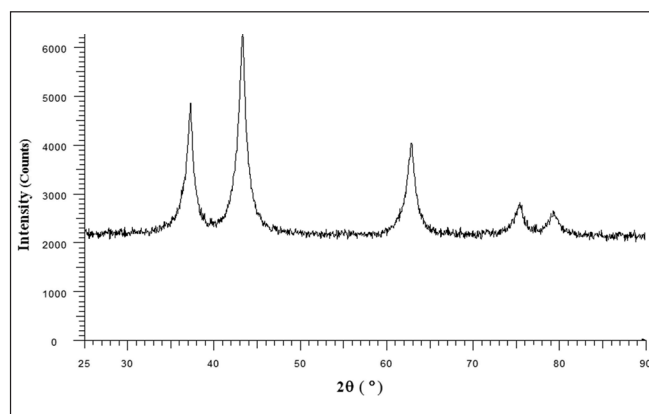


Figure 3: X-ray techniques of Nickel Oxide Nanoparticles

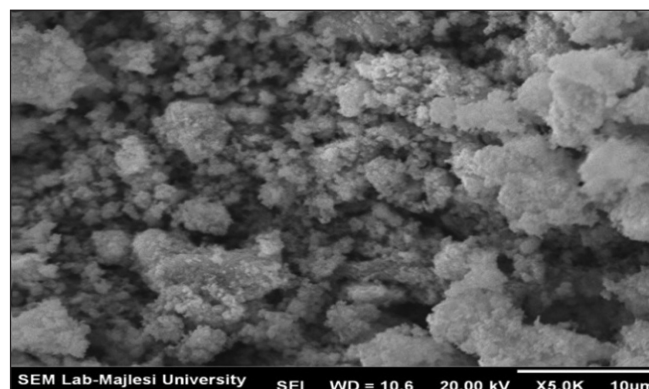


Figure 4: Scanning electron microscopy of nickel oxide Nanoparticles

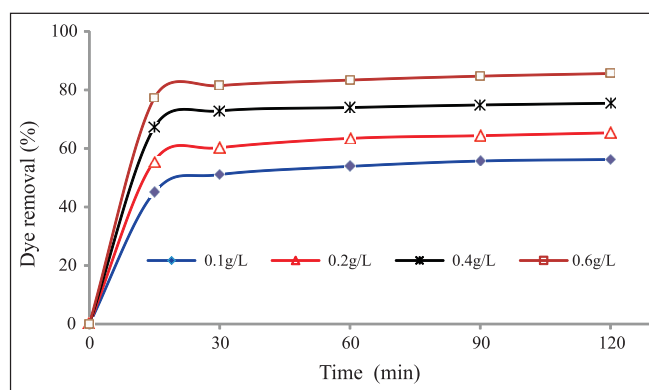


Figure 5: The effect of the nickel oxide adsorbent value and contact time on decolorization of Orange II dye

colorful ions with negative load and adsorption places with positive load increases, so that dye adsorption increases. Lower absorption at higher pH may be due to abundance of OH⁻ ions and the repulsive force between negatively charged surface and anionic dye molecules. Similar results have been obtained by Song *et al.*^[8]

Different dye concentrations of 25 – 50 – 75 – 100 mg/L respectively, were studied. As Figure 7 shows, decolorization speed decreases with increasing initial dye concentration

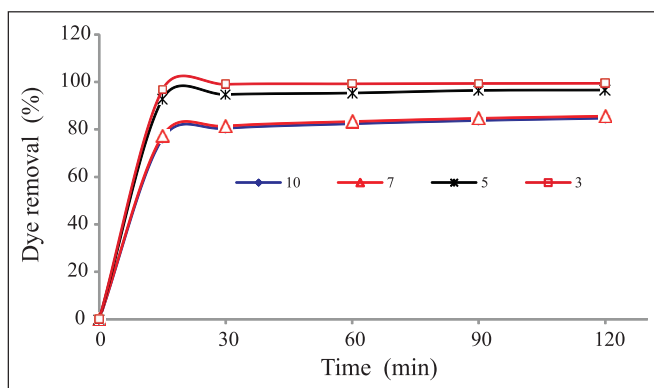


Figure 6: The effect of initial dye pH in the adsorption process

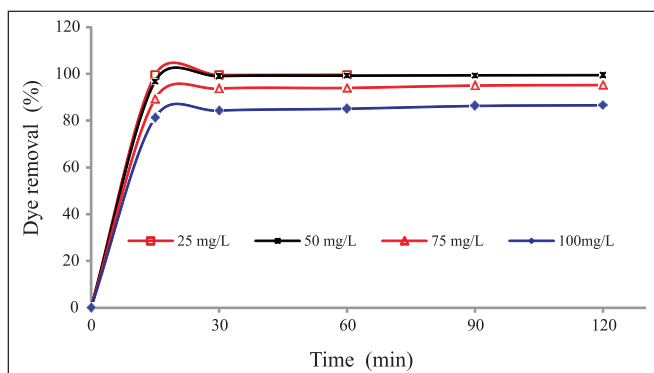


Figure 7: The effect of dye concentration on decolorization process efficiency using nickel oxide adsorbent

from 25 mg/L to 100 mg/L. At lower initial concentration, dye adsorption on adsorbent is very high and equilibrates rapidly. In other words, ratio of the initial number of dye molecules to available adsorption places would be lower, and therefore, some part of adsorption would be independent of initial concentration.^[15] At higher concentrations, the number of available adsorption places is lower, and therefore decolorization depends on the initial concentration.^[14] The results of this study showed that pH is the strongest influencing factor in the process efficiency of the Orange II dye adsorption by nickel oxide nanoparticles. Generally, nickel oxide nanoparticle is an effective adsorbent for decolorizing Azo dye from contaminated wastewaters. 0.6 g/L nickel oxide was considered as the optimum amount of adsorbent for complete decolorization under the condition of 50 mg/L initial dye concentration, pH value 3 and agitator speed 100 rpm about 30 minutes.

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