



## Original Research Article

# NiO Nanoparticles: Synthesis, Characterization, and Methyl Green Removal Study

Aliakbar Dehno Khalaji\*, Negin Mohammadi, Moslem Emami

Department of Chemistry, Faculty of Science, Golestan University, Gorgan, Iran

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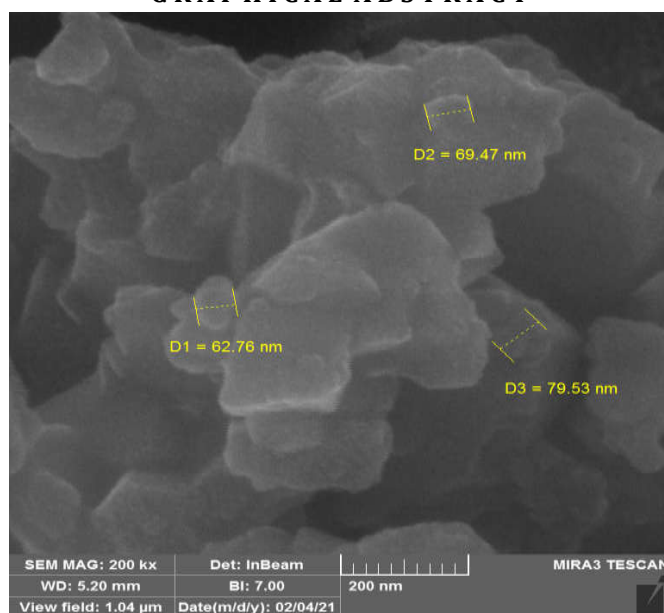
Methyl green

Removal

## ABSTRACT

Nickel oxide nanoparticles were prepared using solid-state thermal decomposition of  $\text{NiCl}_2$  at the presence of maleic acid at  $600^\circ\text{C}$ , characterized by FT-IR spectroscopy, XRD, FE-SEM and applied as new adsorbent for the removal of methyl green (MG) from aqueous solution. FT-IR and XRD results confirmed the preparation of cubic NiO nanoparticles. SEM image showed that NiO nanoparticles have a wide size distribution from 50 to 200 nm. The pH of the solution, adsorbent dosage and contact time effect on MG removal efficiency were considered. The uptake of MG was determined to be  $>96\%$  in 60 min and adsorbent dosage of 0.02 g

## GRAPHICAL ABSTRACT



\* Corresponding author: Aliakbar Dehno Khalaji

E-mail: [alidkhalaji@yahoo.com](mailto:alidkhalaji@yahoo.com)

Tel number: +98 - 1732245882

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## Introduction

Recently, the preparation of nickel oxide nanoparticles has drawn high interest on the part of researchers because of their various application and properties such as organic dyes removal [1-3], supercapacitors [4,5], Li-ion batteries [6,7], and choline sensing [8]. At the moment, water pollution caused by various organic dyes [9-13], is one the most environmental problems, because the dyes are very stable and poorly biodegradable [9-13]. So far, various techniques for water treatment have been examined [14-16]. However, among them, adsorption is the most favorable because of its low-cost, high effectiveness and simplicity [1-3, 9, 17].

Chemical and thermal stability of NiO is high [1], then NiO is used in various applications [1-8]. One of the best and important applications of NiO nanoparticles is removing organic dyes as an efficient adsorbent [1-3]. For example, Zhu *et al.* [1] prepared NiO nanospheres and considered the removal of congo red in aqueous solution. Ai and Zeng [2] synthesized the porous NiO nanoparticles and reported the excellent performance of 223.8 mg/g for congo red remove from aqueous solution. Al-Aoh synthesized NiO nanoparticles using microwave assisted method [3] and reported the high capacity of bromophenol blue adsorption of about 59, 79 and 93 mg/g at 303, 318 and 333 K, respectively. Up to now, various techniques have been used to prepare NiO nanostructures such as solvothermal [1,2], microwave assisted [3], hydrothermal [5], thermal decomposition [7,18-20] and desolvation [8].

Methyl green (MG), as a mono/divalent cationic dye, is used in medicine and biology [21, 22]. In recent years, the application of various compounds to remove methyl green from aqueous solution has received considerable attention [23-33]. Maghni *et al.* [25] studied the removal of MG using activated bentonite and obtained the 353.33 mg/g adsorption capacity.

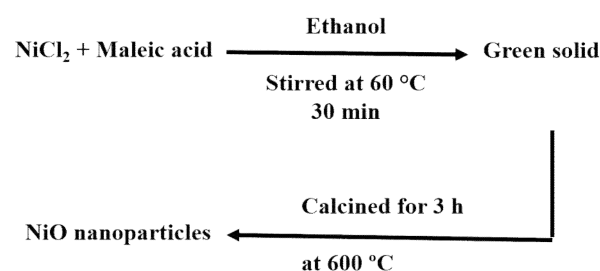
Rida *et al.* [24] reported the adsorption capacity of 67.93 mg/g of MG using activated carbon. Maximum adsorption capacity of MG using  $\text{CoFe}_2\text{O}_4/\text{rGO}$  was reported by Yin *et al.* [27]. Vargas-Rodriguez *et al.* [31] used halloy site nanotube as new adsorbent and reported the maximum adsorption capacity of MG found by 185 mg/g.

In line with the previous work on the preparation of various transition metal nanoparticles [34-39], we prepared new NiO nanoparticles by solid state thermal decomposition of  $\text{NiCl}_2$  at the presence of maleic acid at 600°C (Scheme 1) and characterized by FT-IR, XRD and FE-SEM techniques. In addition, methyl green removal was studied in aqueous solution.

## Experimental

### Materials and methods

$\text{NiCl}_2$ , maleic acid and ethanol was purchased from Merck Co. FT-IR spectrum of NiO was recorded by spectrophotometer instrument (Perkin-Elmer) (KBr disks, 4000–400  $\text{cm}^{-1}$ ). X-ray diffractometer ( $2\theta = 10\text{--}80^\circ$ , Bruker AXS-D8) was used to determine XRD pattern. FE-SEM images were recorded on the TESCAN Vega Model scanning electron microscope. UV-Vis spectra were carried out with a UV-Visible spectrophotometer (Perkin-Elmer).



**Scheme 1.** Schematic of the synthetic route for NiO nanoparticles

### Synthesis of NiO nanoparticles

To a solution of 2.0 g of NiCl<sub>2</sub> in 5 mL of ethanol, 2.0 g maleic acid was added and stirred for about 30 min at 60°C, up to a green solid precipitate. The solid was filtered off, washed with cold ethanol, and dried. Finally, the solid (1 g) was put into a tube furnace and calcined for 3 h at 600 °C under air atmosphere. The final black product was characterized by FT-IR, XRD, and FE-SEM.

### Adsorption experiments

All adsorption experiments were performed according to the previous work [23]. To a 25 mL of MG solution (40 ppm), we added NiO nanoparticles (0.01 and 0.02 g), and the solutions were shaken from 0 to 60 min. At various contact times, the adsorbent (NiO) was separated by centrifuging, and the concentration of MG was determined using a UV-Vis spectrophotometer. The removal percentage rate R (%) of MG onto the NiO was calculated according to the following equation [19, 20]:

$$R = \{(C_i - C_t) \times 100\} / C_i$$

Where C<sub>i</sub> (mg/L) is the initial and C<sub>t</sub> (mg/L) is the final concentration of MG.

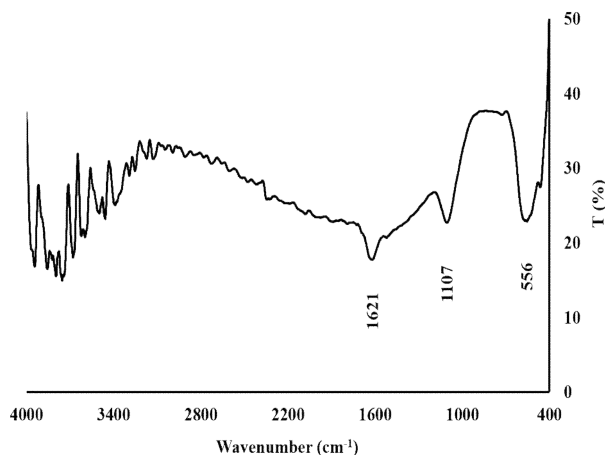
## Results and Discussion

### FT-IR spectrum

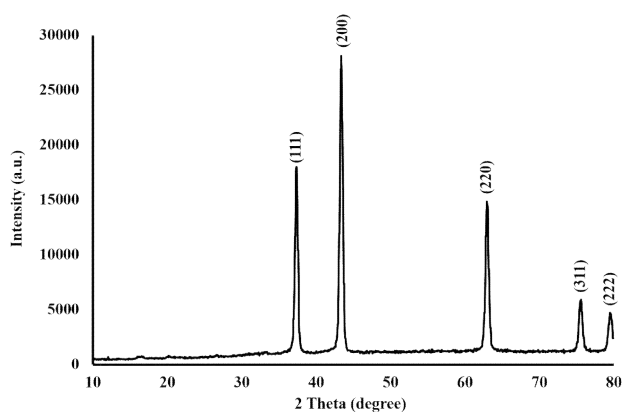
Figure 1 represents the FT-IR spectrum of the as-prepared NiO nanoparticles. The weak peak at 1621 cm<sup>-1</sup> is assigned to the water molecules adsorbed on the surface of NiO nanoparticles [18-20]. The peak at 556 cm<sup>-1</sup> is assigned to the Ni-O stretching vibration [18-20].

### XRD pattern

Fig 2 represents the XRD pattern of the as prepared cubic phase of NiO nanoparticles and exhibits characteristic diffraction peaks at 2θ = 37.4, 43.4, 62.95, 75.6 and 79.5° corresponded to reflections from (111), (200), (220), (311) and (222) miller planes, which were similar to the data of JCPDS card number of 71-1179, confirming the pure cubic structure of NiO nanoparticles [1,2].



**Fig 1.** FT-IR spectrum of the as-prepared NiO



**Figure 2.** XRD pattern of the as-prepared NiO

### FE-SEM images

Figure 3 shows the SEM image of the as-prepared NiO nanoparticles. It shows that the particles lack uniform morphology. The diameter sizes of the particles are in the range of 50-200 nm with average crystal sizes of 135.57 nm, calculated by Scherrer equation.

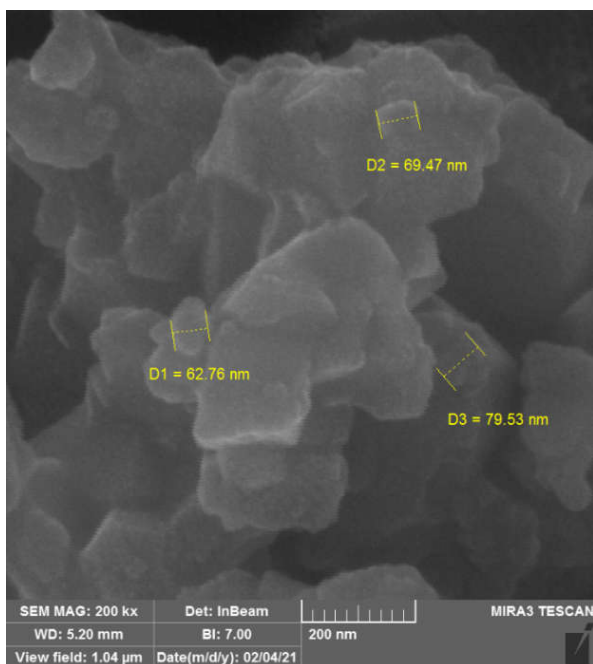
$$D = 0.9 \lambda / \beta \cos \theta$$

Where D is crystallite size, λ is wavelength of X-ray, β is full width at half maximum in radian (FWHM) and θ is the Bragg angle.

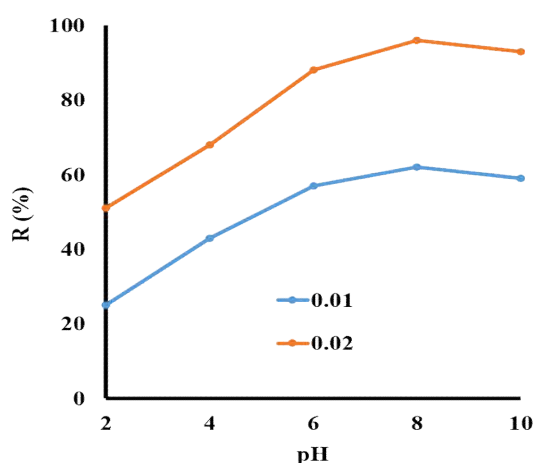
### Effect of pH on MG removal

One of the important factors on dyes removal is pH of the solution [3, 17, 24, 25, 27, 30, 31]. In this paper, MG adsorption was studied in the pH range of 2 – 10 using initial concentration of 40

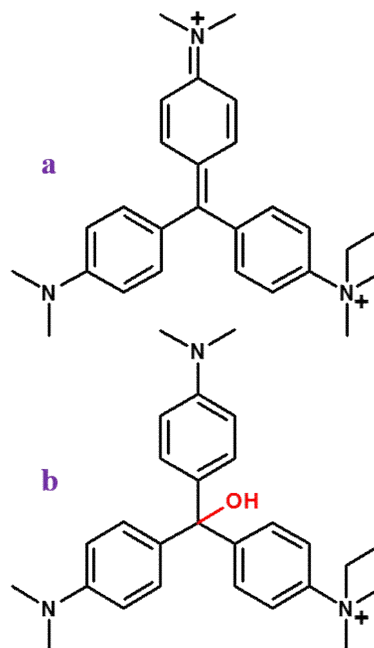
ppm methyl green (Fig. 4). As seen in Figure 4, the MG removal increased with increases of pH from 2 to 8, due to greater negative charge of adsorbent, increasing the electrostatic attraction of cationic MG (Scheme 2) and the adsorbent [31]. At basic solution (pH from 8 to 10), the removal of MG remained almost constant, due to the carbinol form MG (Scheme 2) [31, 40].



**Fig. 3.** FE-SEM image of the as-prepared NiO



**Fig. 4.** The effect of pH (2 – 4) solution on MG removal percentage (%)



**Scheme 2.** a) Cationic and b) carbinol form of MG.

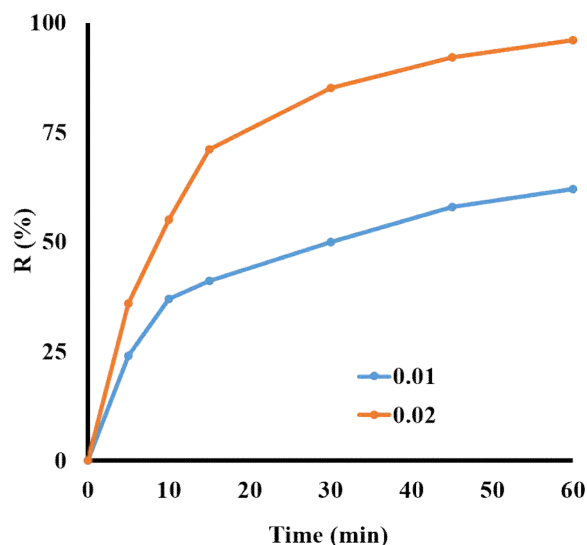
#### *Effect of contact time and adsorbent dosage on MG removal*

The effect of NiO dosage (0.01 and 0.02 g), and contact time (0-60 min) on the percentage removal of MG was studied as represented in Figure 5. As seen in Figure 5, with increasing the contact time, increase in MG removal happened. Adsorption equilibrium is achieved at 60 min, followed by the saturation of the active sites on the surface of NiO nanoparticles. Also, increasing NiO dosage causes the increase of MG removal, from 62% using 0.01 g to 96% using 0.02 g of NiO nanoparticles [17, 23, 24, 31].

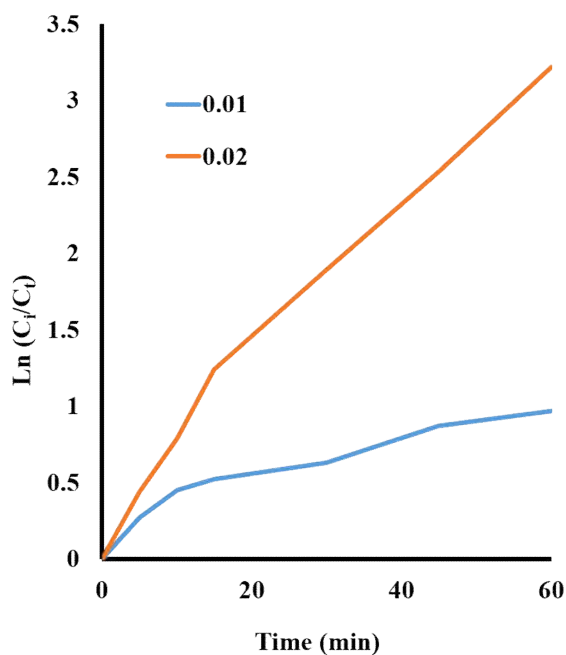
The Langmuir isotherm is one of the best models to study the adsorption of cationic dyes on the surface of adsorbent as a homogeneous monolayer without any interaction [32, 42, 43], according to the following equation:

$$\ln (C_i/C_t) = kt$$

rate constant  $k$  was calculated by the slope of fitting curve of  $\ln (C_i/C_t)$  versus  $t$  (Fig. 6). The rate constant ( $k$ ) was calculated 0.016  $\text{min}^{-1}$  and 0.054  $\text{min}^{-1}$  for using 0.01 g and 0.02 g of NiO, respectively.



**Fig. 5.** MG removal percentage (%) in different contact times (0-60 min) and adsorbent dosages (0.01 and 0.02 g)



**Fig. 6.** Kinetic curve of MG adsorption using NiO nanoparticles (0.01 and 0.02 g)

Table 1 comparing the maximum adsorption capacity ( $q_t$ ) of various adsorbents used for removal of methyl green in aqueous solution. The  $q_t$  (mg/g) was calculated using the following equation [31].

$$q_t = \{(C_i - C_t) \times V\} / M$$

Where  $V$  (L) and  $M$  (g) are the volume of the solution and the mass of NiO, respectively.

The maximum amount of MG adsorbed on the surface of Ni was calculated 48 mg/g for the conditions of pH 8, 60 min and 0.02 g of NiO.

**Table 1.** Comparison of the adsorption of MG onto various adsorbents

Adsorbent	$q_t$ (mg/g)	Ref
Activated carbon	67.93	24
Activated bentonite	353.33	25
CNTs	181.2	26
Chitosan Schiff base	1536.6	23
NiFe <sub>2</sub> O <sub>4</sub> -CNTs	110.3	26
CoFe <sub>2</sub> O <sub>4</sub> -rGO	88.3	27
ZSM-5	39.48	30
Silica gel/ZnO	24.91	32
MCM-41	20.97	29
NiO	48	This work

### Conclusion

In this study, NiO nanoparticles were prepared using thermal decomposition technique and characterized by FT-IR, XRD and SEM. The as-prepared NiO nanoparticles had a distribution size from 50 to 200 nm with the average size of 135.57 nm; they were calculated using XRD. The as-prepared NiO nanoparticles were used as an adsorbent to remove MG from aqueous solution. Amount of 96% (48 mg/g) was adsorbed within 60 min from 40 mgL<sup>-1</sup> methyl green solution using 0.02 g of NiO nanoparticles. Therefore, the as-prepared NiO is a good adsorbent to remove dye from aqueous solution.

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