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Degradation studies of Organic Pollutants using green synthesized NiO NPs from Clove Extract and investigation of antimicrobial properties

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Abstract

In this study nanoscaled Nickel oxide nanoparticles were synthesized using clove extract under atmospheric condition through green synthesis. The synthesized nanoparticles were characterized using FT-IR, UV-Visible spectroscopy, EDAX, SEM and XRD techniques. Using Debye Scherer's formula, the crystallite size was calculated. The presence of Ni was confirmed using EDAX. Using the synthesized nanoparticle, degradation of Congo Red dye was carried out. The effect of various parameters on dye degradation like catalyst dosage, dye concentration, pH were investigated systematically. The results revealed that NiO nanocatalyst are an efficient catalyst to carry out the degradation of Organic dyes.

Keywords: Dye degradation, organic dyes, XRD, SEM , Congo Red , NiO nanoparticle.

Introduction

Among all the various fields of nanotechnology, photocatalysis has attracted a huge scientific interest with significant developments during the last decade. Modern day photocatalysis began with Fujishima and Honda reporting photocatalytic splitting of water in 1972[1]. In the past 30 years, numerous semiconductors and molecular assemblies have been investigated as photocatalyst candidates[2]. In recent years, transition metal oxides (TMOs) have attracted considerable interest due to their potential applications such as supercapacitors, sensors, solar cells, photocatalysis and electrochromic devices[3-8].

Water pollution, along with the increasing production of industrial wastewater, has become a concern all over the world, which reduces the availability of clean water for living beings. Dyes wastewater from textile, paper, and leather industries are the main problem of contamination in the aquatic environment. Reactive dyes are the most commonly used (60-70%) among various type of synthetic dyes due to low energy consumption in the dyeing process, water fastness, and color brightness.

The complex chemical structure makes this dye stable and difficult to biodegrade and the concentration in the environment tends to persist if this dye enters the water bodies[9,10]. Meanwhile, the negative effects of this dye on humans and aquatic animals have been reported due to its toxic and carcinogenic properties[11-13]. Thus, an effective method is needed to remove the concentration of this dye from wastewater before being discharged into the environment. This method has gained considerable attention because it is simple to handle and significantly produces lower residues compared to the conventional treatment process[14-16].

NiO NPs may have many applications such as in the manufacture of electrochromic, films, magnetic materials, p-type transparent conducting films, gas sensors, catalyst, alkaline batteries cathode, and solid oxide fuel cells anode[17–26].

The present work investigates on the degradation of organic dyes using green synthesized NiO nanoparticles. Antimicrobial activities of the nanoparticles were investigated in the present study. Antibacterial and antifungal activities were carried out and the results were shown.

Materials and Methods

This chapter deals with the synthesis and characterization of the photocatalyst. The instrumentation is also discussed here. Congo Red Dye was used for the study. Double distilled deionized water was used for the studies. Systronics Double beam Spectrophotometer - 2203 was used for the absorption measurements. JASCO-FP 8200 Spectrofluorometer was used to measure the emission spectra.

Preparation of Clove extract: About 5 grams of the cloves were boiled with 100 mL of double distilled water in a 250 mL beaker for about 15 minutes. It is then cooled to room temperature. The solution is filtered and the filtrate is stored for further use.

Preparation of NiO-NPs: Nickel oxide nanoparticles were synthesized using modified protocol from the previous studies. About 0.01M solution of Nickel sulphate solutions was added to the clove extract in 1:1 volume ratio. NiO NPs were formed respectively immediately by the reduction process. The mixture was stirred magnetically for 30 minutes and the allowed to stand for about an hour at room temperature to get a colloidal suspension of the nanoparticles. The mixture was then centrifuged and washed with water several times and dried in an air oven at 100°C for about 2 hours. The dried nanoparticle is calcined at 400°C in a muffle furnace and it is then stored in air tight container for further use.

Characterization of the Photocatalyst: The synthesized photocatalyst was characterized by UV-Visible spectroscopy, FT-IR, EDAX, SEM, XRD. The characterization studies were carried out to understand the structural, morphological, elemental composition, crystallite size, functional groups. The dye degradation studies were carried out and they were analyzed using UV- visible spectroscopic Techniques. The degradation studies were carried out in presence of induced light, sonication in presence of sunlight. Studies were carried out by varying the pH of the dye solution.

EDAX: EDAX Analysis stands for Energy Dispersive X-ray analysis. It is sometimes referred to also as EDS or EDAX analysis. It is a technique used for identifying the elemental composition of the specimen. It's an investigative method which is widely employed in elemental or chemical characterization of objects. The sample may be of any form such as solid thin films, solid powder, liquid sample or even a pellet etc. Elemental composition and purity of the photocatalyst is acquired using EDAX Studies. SEM is used to know about the morphology of the particle.

Figure-1(a): SEM image of NiO NPs.

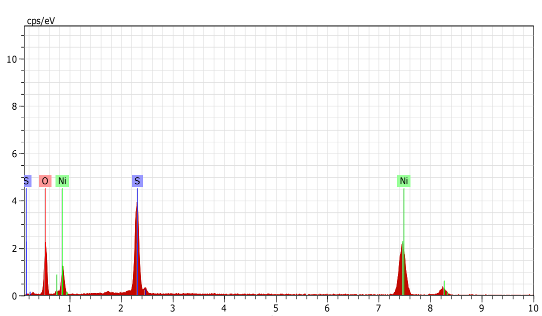


Figure-1(b): EDAX spectrum of NiO-NPs.

The SEM and EDAX of NiO nanoparticle are depicted in Figure-1. The presence of Ni and O are confirmed from the EDAX result.

Table-1: Chemical composition of NiO nanoparticle based on EDAX.

Element	At. No	Series name	wt. %	At. %
O	8	K-series	51.79	70.27
Ni	28	K-series	30.98	14.21
S	16	K-series	13.24	8.96
Total			100	100

XRD pattern of NiO –NPs: The crystallite size was calculated using Debye Scherer formula

$$D_{hkl} = k\lambda/\beta\cos\theta \qquad (1)$$

Where: λ is the wavelength (λ=1.542Å) (Cu Kα), β is the full width at half maximum (FWHM) of the line θ is the diffraction angle. The crystallite size was found to be 19.8 nm which confirms the nanoscale nature of the formed product.

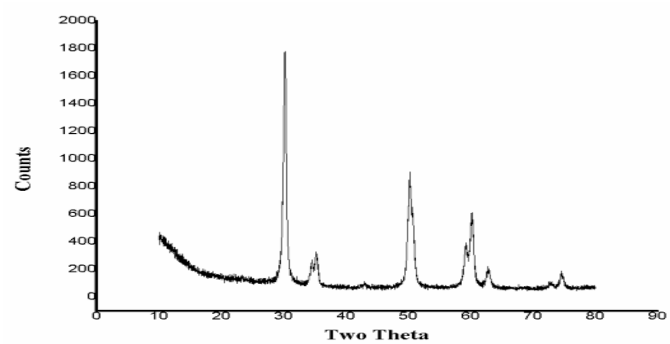


Figure-2: XRD pattern of NiO NPs.

The diffraction lines of the XRD pattern of NiO nanoparticle displayed in Figure-2 is relatively strong which indicates the high crystallinity of the nanoparticles. The peaks appeared at 2θ values ranging from 30.24°, 50.23° and 50.70°. The XRD of NiO NPs confirms the formation of face-centered cubic (FCC) structure of NiO phase which was perfectly matched with JCPDS No. 47-1049. The intense and sharp peaks show the crystalline nature of the sample and absence of other peaks indicate the phase purity. The peaks are in good agreement with the literature report[27].

FT-IR analysis shows the stretching vibrations at 3413.93 cm^{-1}, 2902.53 cm^{-1} 1625.99 cm^{-1}, 679.44 cm^{-1} within the region of 400-40000 cm^{-1}. These peaks confirm the formation of NiO-NPs. The peak at 3413.93 cm^{-1} corresponds to the O-H bond stretching. This shows the aqueous phase as well as the reduction of $NiSO_4$.The peak at 1625.99 cm^{-1} represent the C-O precursor. The strong peak at 679.44 cm^{-1} denotes the Ni-O-H stretching bond. 2902.53cm^{-1} shows the C-H stretching mode as given in Figure-3.

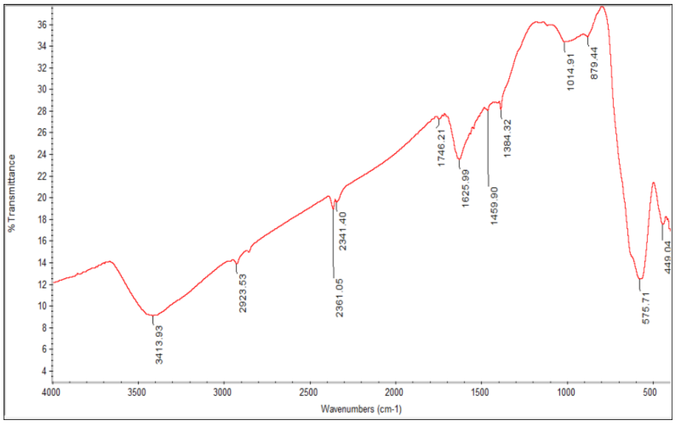


Figure-3: FT-IR Spectrum of NiO nanoparticle.

Absorption spectral characteristics of nanoparticles: The synthesized nanoparticles were characterized by taking the absorption spectra of the solutions. UV-visible spectroscopy measures the extinction of light passing through a sample. Nanoparticles have unique optical properties that are sensitive to the size, shape, concentration, agglomeration state, and refractive index near the nanoparticle surface, which makes UV-Vis a valuable tool for identifying, characterizing, and studying nanomaterials. Spectral measurements were done using Systronics - 2203 Double Beam Spectrophotometer. The following figures show the absorption spectra of the nanoparticles.

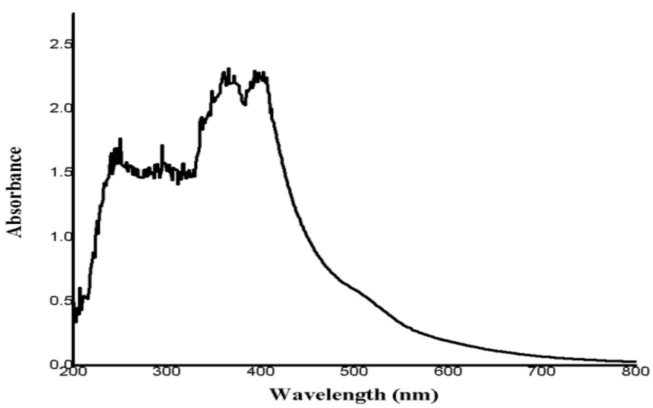


Figure-4: Absorption spectra of clove extract.

The Figure-4 shows the absorption spectrum of clove extract. It has maximum absorption in the visible region of the spectrum.

Results and Discussion

Photodegradation Study of Congo red Dye with NiO NPs: Photodegradation of CR dye carried out in presence of artificial light of 200W. The reaction was carried out using 0.01g of the nanophotocatalyst and 100 mL of the CR dye was taken. The solution was stirred using magnetic stirrer. Absorption maximum of the dye solution was taken out every half an hour interval.

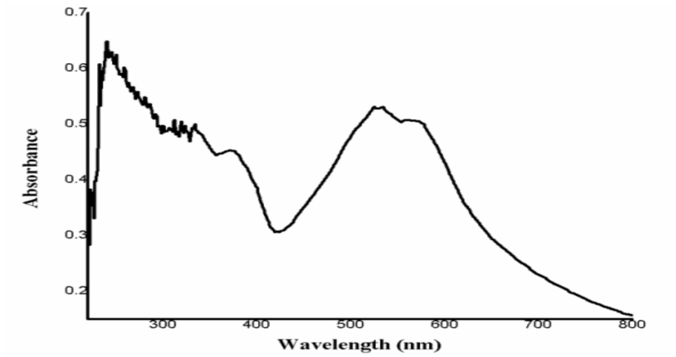
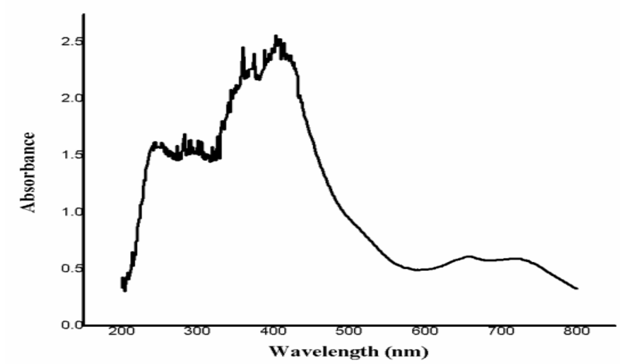


Figure-5(a): Absorption spectrum of Congo Red Dye.



(b)

Figure-5(b): Absorption spectrum of NiO nanoparticle.

Absorption maximum of the dye solution was taken out every half an hour interval. Figure-5 shows the absorption spectrum of Congo Red Dye and the synthesized NiO nanoparticle. They have an absorption maximum of 532 nm and 403 nm respectively.

Degradation of Congo red Dye in presence of NiO nanoparticle: Figure-6 displays the degradation of CR dye in presence of NiO nanocatalyst in induced light. Degradation studies were carried out for about three hours. Absorption spectrum was carried out to the dye solution for every thirty minutes and the readings were recorded. The decrease in absorbance with increase in time shows that the dye degrades with time in presence of induced light.

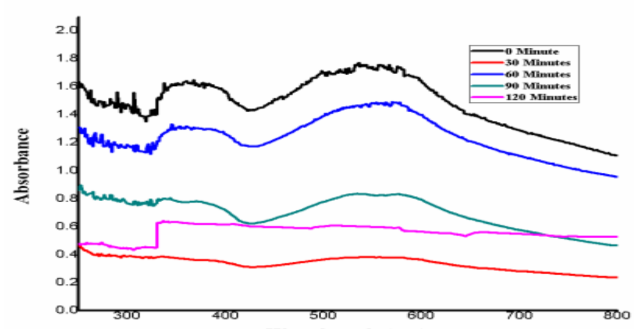


Figure-6: Degradation of Congo red dye in presence of NiO in presence of induced light.

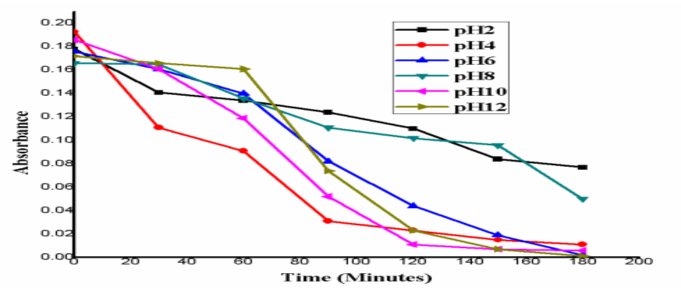


Figure-7: Plot of Time *vs* absorbance for the degradation of Congo Red Dye in presence of NiO nanoparticle.

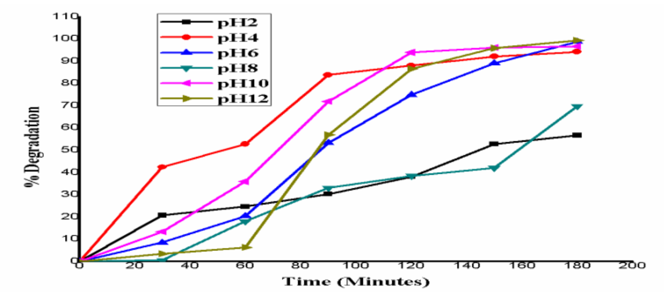


Figure-8: Plot of pH *vs* % Degradation for the degradation of Congo Red Dye in presence of NiO nanoparticle.

pH plays an important role in the degradation of dyes. The effect of pH is a pivotal parameter in the efficiency of the decolorization process because it can affect the photocatalyst surface, dye characteristics, and the rate of the degradation process. pH affects the charge on the catalyst particles, size of catalyst aggregates and the positions of conductance and valence bands. Due to the nature of catalyst used, any variation in the operating pH is known to affect the isoelectric point or the surface charge of the photocatalyst used. Since dyes can to degrade at different pHs in coloured effluents, comparative experiments were performed at different pH values. Figures-7 and 8 displays the variation of degradation at different pH scales from acidic to basic medium. It is seen that the degradation efficiency is maximum at pH12. It has 99.4 % degradation at this pH.

Kinetics of the photocatalytic degradation of Congo Red Dye in presence of NiO photocatalyst: The kinetics plot for CR dye degradation in presence of NiO nanocatalyst is shown in Figure-9. The regression coefficient R^2 of the experimental values was found to be 0.8888 degradation was carried out at various time intervals. This confirms that the degradation of the dye molecules obeys the pseudo first order linear kinetics.

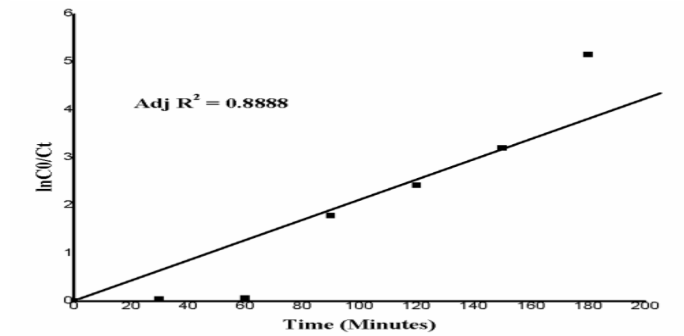


Figure-9: Plot of Time *vs* Ln Co/Ct for the degradation of Congo Red Dye in presence of NiO nanoparticle.

Antibacterial Activity: The antibacterial activity was determined by the well diffusion method. 25mL of nutrient agar was poured into a sterile petri plate. The plates were allowed to solidify 100µL of pathogenic bacteria and were transferred into the dish and made culture lawn by using a sterile L rod spreader. After five minutes setting of the pathogenic microbes, a sterile cork borer was used to make a 5 mm well on the agar. The test samples were loaded into wells. The solvent saline loaded well served as the negative control and ampicillin (30 µg/mL) served as the positive control. The plates were incubated at 37^{o}C for 24 hours. The antibacterial activity was determined by measuring the diameter of the zone of inhibition around the well.

Table-2: % Degradation efficiency of CR dye in presence of NiO nanoparticle with various pH.

Time (Min)	pH 2	pH 4	pH 6	pH 8	pH 10	pH 12
0	0	0	0	0	0	0
30	20.78652	42.48705	8.522727	0.60241	13.44086	3.488372
60	24.7191	52.84974	20.45455	18.07229	36.02151	6.395349
90	30.33708	83.93782	53.40909	33.13253	72.04301	56.97674
120	38.20225	88.0829	75	38.55422	94.08602	86.62791
150	52.80899	92.22798	89.20455	42.16867	96.23656	95.93023
180	56.74157	94.30052	98.86364	69.87952	96.77419	99.4186

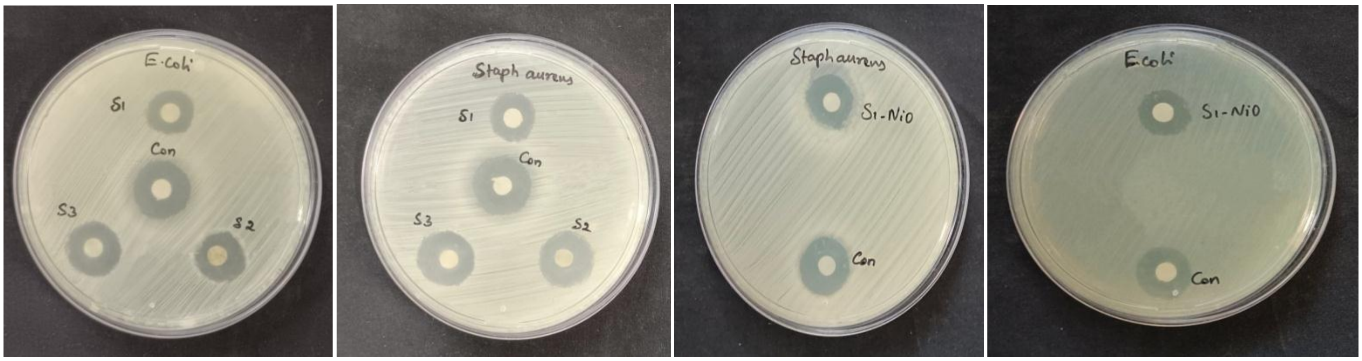


Figure-10: Antibacterial activity of the metal oxide nanoparticles against *Staphelococcus* aureus (Gram positive bacteria).

Table-3: Antibacterial activity of the metal oxide nanoparticles.

Bacteria name	NiO	Clove extract	Control (Amikacin)
Escherichia. coli	15 mm	16.2mm	17mm
Staphilococcus aureus	15.2mm	17.5mm	18mm

Antibacterial activity of the synthesized nanoparticles and the clove extract were examined using *Staphelococcus* aureus (Gram positive bacteria) and *Escherichia coli* (Gram- negative bacteria) (Figure-10). Amicacin tablet was used as a control. The results are tabulated in Table-3. The results show that NiO nanoparticle shows a good antibacterial activity against both *Staphelococcus* aureus *and E.coli.* The clove extract also shows good inhibitory action against the bacterial stains.

Antifungal Activity: The antifungal activity was tested by the well diffusion assay. The potato dextrose agar medium was poured into a sterile petri dish. The plates were allowed to dry. Then 100µL of fungi were transferred into the plate. After, five minutes, a sterile cork borer was used to make a 5mm well on the agar. The samples were loaded into the well and Nystatin (30 µg/mL) served as the positive control. The plates were incubated at 37°C for 24 to 48 hours. The antifungal activity was determined by measuring the diameter of the zone of inhibition around the well (Figure-11).

Figure-11: Antifungal activity of the Nickel oxide nanoparticles and clove extract.

Antifungal activity was examined using well diffusion method. Nystatin was used as a control. *Candida albicans* was the fungi used for examination. NiO nanoparticle shows good antifungal activity. The precursor clove extract shows good antifungal activity (Table-4). They have a larger zone of inhibition against the fungal stain. The bioactivities of the synthesized nanoparticles show that they can be used effectively in pharmaceutical fields.

Table-4: Antifungal activity of the NiO NPs and clove extract.

S. No.	Sample	Zone of inhibition
1	NiO NPs	13 mm
2	Clove extract	15 mm
3	Nystatin (Control)	14 mm

Conclusion

Today, nanotechnology has played an important role in green chemistry and the use of nanoparticles in the removal of environmental pollutants is one of the newest methods of removing pollutants in the world. Nanoparticles exhibit distinct features than their bulk materials and they find many applications in various fields of science. Green synthesized nanoparticles have attracted a lot of attention. In the present study metal oxide nanoparticle namely NiO NPs, were green synthesized using clove extract. The present study deals with the photocatalytic degradation of organic dyes namely Congo Red Dye using green synthesized Nickel oxide nanoparticles. The synthesized nanoparticles were characterized using FT-IR, UV-Visible spectroscopy, EDAX, SEM and XRD techniques. The crystallite size of the nanoparticles were calculated using Debye Scherer formula. All the measurements confirm that the synthesized particles are nano sized. XRD analysis confirmed the elements present in the nanoparticles.

Degradation studies were carried out using the synthesised Nickel oxide nanoparticle. Studies were done under various conditions like induced light, sunlight and sonicator.

Antibacterial and antifungal studies were carried out for the clove extract and the nanoparticles. In presence of induced light CR dye degrades efficiently in presence of NiO nanoparticle. It shows maximum efficiency.

Degradation studies were carried out by changing the pH of the dye solution. Next, the sample collected was applied for the photodegradation of CR dye under natural sunlight irradiation and the natural pH of the reaction mixture without adding any external oxidant. The influence of varying dye initial concentration as well as NiO catalyst dosage on the elimination performance of the dye was also investigated.

So these nanoparticles are ecofriendly as well as they can be used to reduce pollution by degradation of many textile dyes. The results show that NiO nanoparticle shows a good antibacterial activity against both *Staphelococcus aureus and E.coli.* The clove extract also shows good inhibitory action against the bacterial stains.

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