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RESEARCH PAPER

Synthesis, Characterization of NiO/ZnO Nanocomposite as Photocatalyst for Treatment of Industrial Dye Wastewater

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ABSTRACT

In this article preparation of NiO/ZnO nanocomposite using co-precipitation method, the characteristic of nanocomposite was studied using various techniques such as X-ray diffraction (XRD), Fourier transform infrared (FTIR), Energy dispersive X-ray spectroscopy (EDX), Scanning electron microscopy (SEM), and Thermal gravimetric analysis (TGA), The particle size of synthesized NiO/ZnO nanocomposite was calculated using the Scherer equation (23.769nm). The second part includes the study of the photo catalytic activity of prepared nanocomposite, using an Fast green dye. The rate of the photocatalytic degradation process was affected by different parameters such as the initial concentration of dye, the effect of the mass of catalyst, the initial pH of the solution, and the effect of temperature. The experiments were performed under optimum conditions from (0.15 g / 100 mL) of NiO/ZnO nanocomposite, and (10 mL/min) air flow rate, The best removal percentage of Fast green dye was (94.33%), The effect of dye concentration has been used to study the optimum value of Fast green dye (10mg/L) dye with a pH=8. The activation energy for the photo degradation process calculated using the Arrhenius equation was equal to (31.67 KJ/mol),

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INTRODUCTION

Numerous investigations into photocatalysis have been carried out in recent years. An electron-hole pair is created via oxidation or reduction on the catalyst's surface at light energies below its intensity. An organic pollutant can be oxidized in the presence of a photocatalyst either directly by a photo-generated hole or indirectly through a reaction with various reactive groups (ROS), such as the hydroxyl radical, created in solution [1].

Textile dyes are one of the most common organic compounds that causes water contamination. Azole dyes make up the majority of the dyes now on the market, followed by anthraquinone dyes. UV and visible light irradiation do not affect them. They are also resistant to aerobic degradation and can be transformed into carcinogenic aromatic amines in anaerobic or in vivo settings [2]. Azo dyes are the most common synthetic colourants used in everyday life, particularly in textile processing. Azo dyes are distinguished by the presence of one or more azo bonds (–N=N–) in combination with aromatic systems and auxochromes (–OH, –SO_3, and so on). Poisonous, mutagenic, carcinogenic, and non-biodegradable textile effluents have been discovered, posing a public

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health danger. Traditional methods for eliminating azo dyes from the aquatic environment include physical, chemical, and biological processes such as adsorption, coagulation, photo-catalysis, ozonation, and biosorption. However, these tried-and-true procedures have proven to be costly, time-consuming, and ineffective [3]. Azo dye compounds have a wide range of applications in the industry, including photodynamic therapy, photosensitivity, and biological activity due to their use in anti-inflammatory, antibacterial, and antifungal treatments [4].

Heterogeneous reactions have risen in prominence among the various types of chemical reactions in recent years. Heterogeneous reactions, particularly in industrial fields, necessitate solid catalysts. Heterogeneous environments are used in 80 percent of commercial catalytic processes

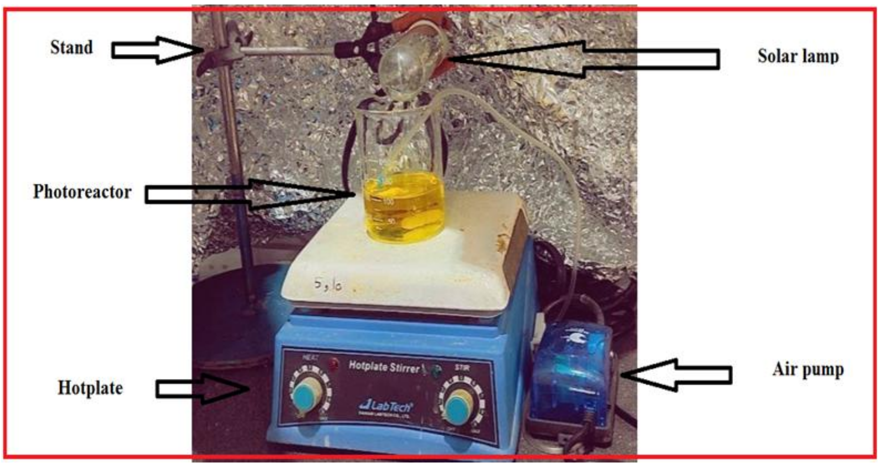


Fig. 1. Optical photo for main parts of the Photocatalytic degradation system.

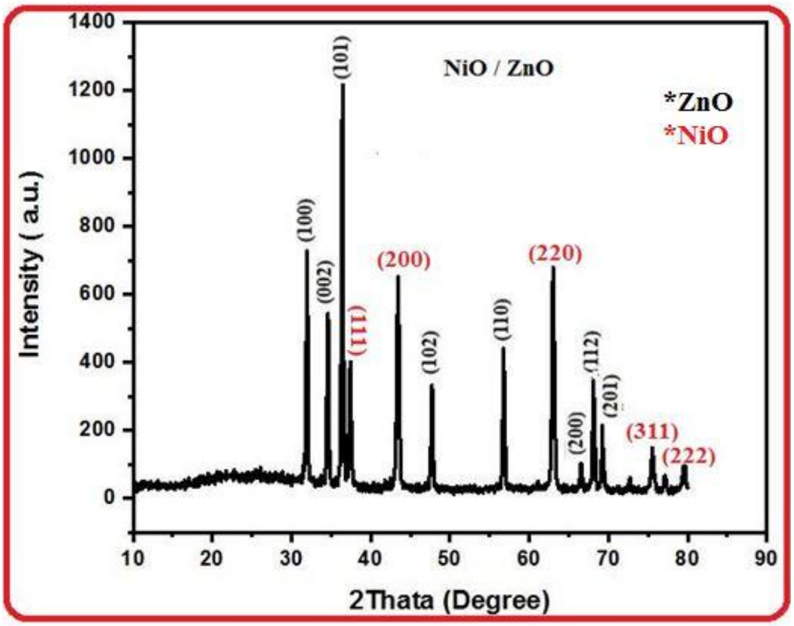


Fig. 2. XRD patterns of NiO/ZnO nanocomposite, where zinc oxide in black color and nickel oxide in red color .

today. Among numerous forms of heterogeneous reactions, the mechanism of surface reactions on a solid substance has been extensively studied. Heterogeneous reactions in other systems, on the other hand, have not been investigated in depth from a macroscopic perspective due to technical constraints. In liquid–liquid heterogeneous materials, for example, phase separation is

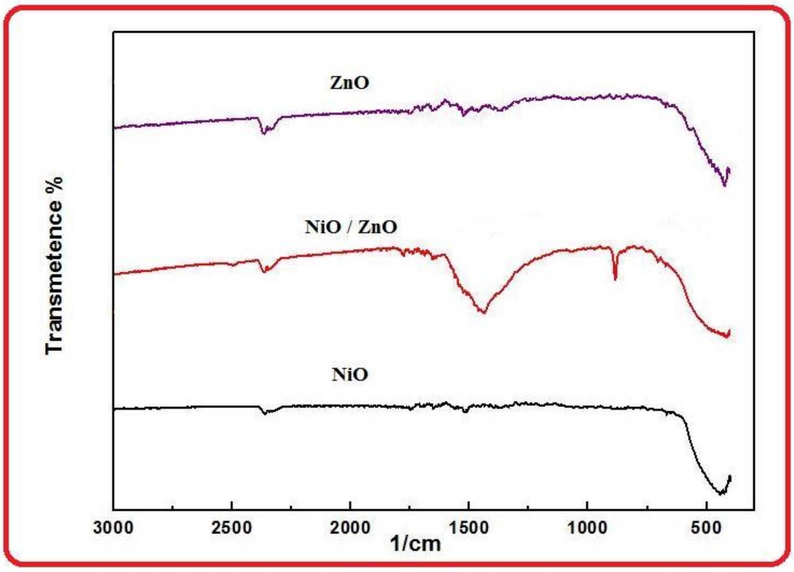


Fig. 3. FTIR NiO/ZnO nanocomposite.

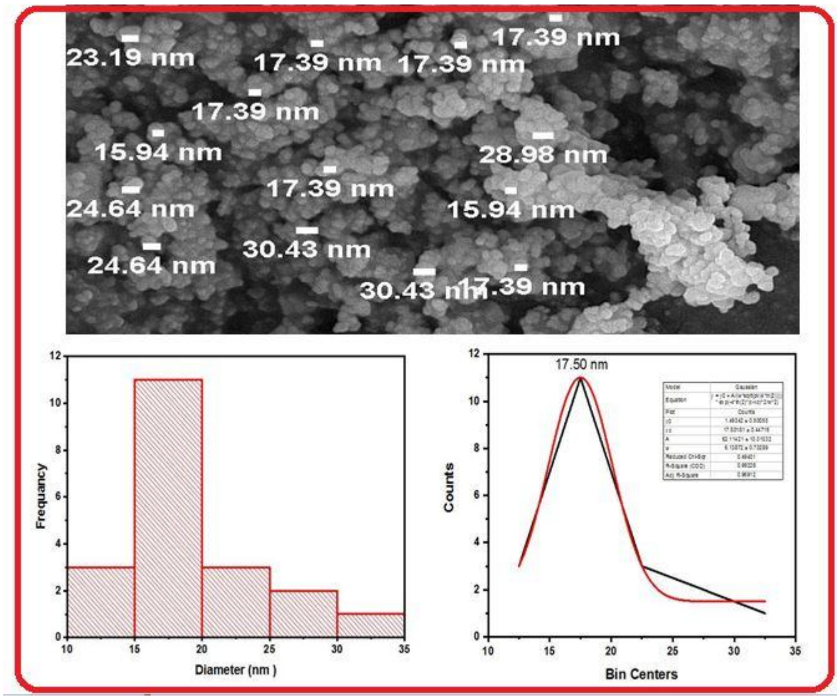


Fig. 4. SEM pattern of NiO/ZnO nanocomposite.

difficult, whereas the optical spectroscopic investigation is difficult due to the substantial light scattering generated by heterogeneity [5]. Using semiconductor oxides, metal oxides, and solar lamps, heterogeneous photo catalytic methods have been employed to remove organic and inorganic pollutants from carbon dioxide and water in the last decade. Zinc oxide is one of the most important semiconductors because it is a highly stable, inexpensive metal oxide that is both nontoxic and environmentally benign [6].

MATERIALS AND METHODS

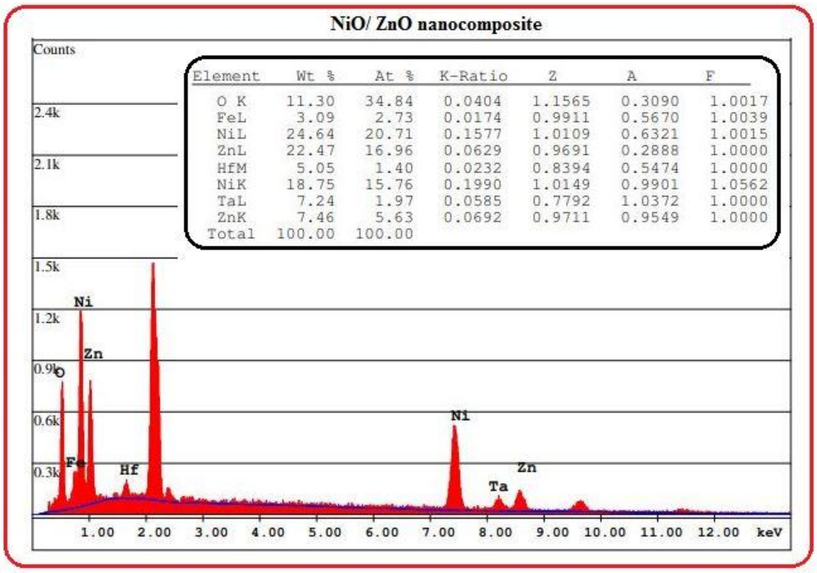
Chemicals

a- Zinc nitrate hexahydrate [$Zn(NO_3)_2·6H_2O$, has been supplied by Fluka.

b- Nickel nitrate hexahydrate [$Ni(NO_3)_2·6H_2O$, supplied by Fluka AG.

c- Fast green dye, supplied by sigma – Aldrich.

d- Sodium hydroxide, supplied by Fluka



Element	Wt %	At %	K-Ratio	Z	A	F
O K	11.30	34.84	0.0404	1.1565	0.3090	1.0017
FeL	3.09	2.73	0.0174	0.9911	0.5670	1.0039
NiL	24.64	20.71	0.1577	1.0109	0.6321	1.0015
ZnL	22.47	16.96	0.0629	0.9691	0.2888	1.0000
HfM	5.05	1.40	0.0232	0.8394	0.5474	1.0000
NiK	18.75	15.76	0.1990	1.0149	0.9901	1.0562
TaL	7.24	1.97	0.0585	0.7792	1.0372	1.0000
ZnK	7.46	5.63	0.0692	0.9711	0.9549	1.0000
Total	100.00	100.00				

Fig. 5. EDX spectrum of NiO/ZnO nanocomposites.

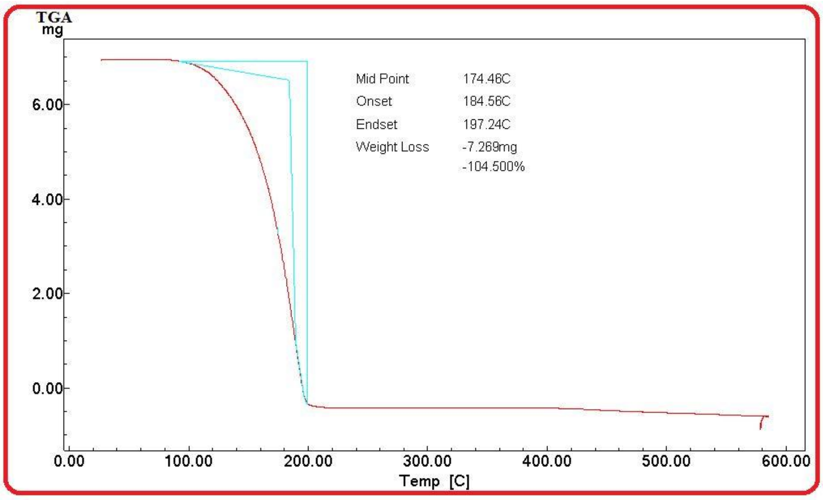


Fig. 6. Thermogravimetric analysis (TGA) of NiO/ZnO nanocomposite

e- Hydrochloric acid, supplied by Fluka.

All chemicals were employed without any further purification.

Photocatalytic Reactor Set up

Fig. 1 shows the photocatalytic degradation system that was used to carry out all experiments.

Photocatalytic degradation experiment

The photocatalytic degradation experiment was carried out with the use of a photocatalytic reactor system, which included all of the major components illustrated in Fig. 1. The photocatalytic activity of the produced NiO/ZnO nanocomposite was studied using dye. Under solar light, photocatalytic degradation of dye in an aqueous solution was completed using NiO/ZnO composites as a photo catalyst. There are two pieces to a photo reactor. The first was employed to cool the suspension solution with the help of the cooling water that passed through it. The second component contains a suspension solution with a capacity of (100 mL) for degrading the dye. Using distilled water, a (100mg/L) stock solution of dye solutions was created. Stirring produced a suspension solution combination for concentrating dye. By adding (0.15 g) of NiO/ZnO nanocomposite to (100 mL) of dye and stirring, a suspension mixture was created. A benchtop solar light source was used to irradiate the respective suspension solution mixture. Every 10 minutes, about 2-3 mL of sample was taken with a syringe and centrifuged at 3000 rpm for 10 minutes, after which the dye absorbance was measured with a UV–Vis spectrophotometer.

Preparation of NiO/ZnO using co-precipitation method

The synthesis of NiO/ZnO nanocomposite were prepared using the homogeneous co-precipitation method, starting with a solution with a Zn:Ni mole ratio of 1:1 in 100 mL deionized water with the amount of nickel salt 2.9079 g and zinc salt 2.9748 g. Finally, dried NiO/ZnO nanocomposite were grinded to obtain fine powders and then annealed at 600 ◦C for 2 h.

RESULTS AND DISCUSSION

Characterization NiO/ZnO nanocomposite

X-Ray Diffraction analysis for NiO/ZnO nanocomposite

X-ray diffraction technique was used to study the crystallinity of the NiO/ZnO nanocomposite as a catalyst, and measure the particles size of the synthesized catalyst. All the measurements has been performed using XRD6000, Shimadzu, Japan. The measuring parameters were set with 45 Cu Kα radiation (λ = 1.54056 Å) at 40kV, 30mA with a rate of 5deg / min and ran at the 2θ range (3-90˚). As shown in Fig. 2 the peaks positions of NiO/ZnO nanocomposite appearing at 2θ (31.9616˚,34.5979 ˚,36.4426˚, 37.6856˚, 43.4265˚,

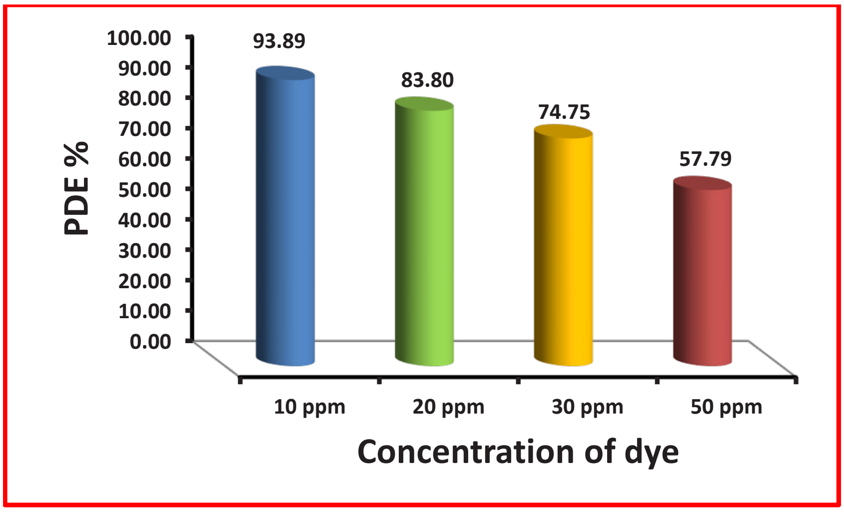


Fig. 7. Photocatalytic degradation efficiency using (10mg/L) of Fast green dye against mass of NiO/ZnO nanocomposite.

47.7306˚, 56.7618˚, 63.0019˚, 66.5386˚, 68.1023˚, 69.2379˚, 75.5144˚, and 79.4791˚) can be readily indexed as (100), (002), (101), (111), (200), (102), (110), (220), (200), (9112), (201), (311), and (222) crystal planes of the ZnO and NiO, which agrees well with the standard data of zinc oxide (JCPDS card no 36-1451)and standard data of Nickel oxide (JCPDS card no. 47-1049).

Fourier- Transform Infrared Spectroscopy synthesized NiO/ZnO nanocomposite

For the NiO/ZnO nanocomposite consists of the peaks at (432.39,470.75,952.45,1489.16, and 2349.50). The band located at 2922 cm⁻¹ represents (CO_2) groups. The band located at 1482 cm⁻¹ represents (C=O) symmetric groups.The peak at 952 cm⁻¹ can be attributed to the (Ni-O) bond and 470 cm⁻¹ (Ni-O). The band located 470 cm⁻¹ of Zn-O [7]. All results are shown in Fig. 3.

Scanning Electron Microscopy (SEM) for synthesized NiO/ZnO nanocomposites

The Scanning Electron Microscopy techniques was used to study the morphology of NiO/ZnO nanocomposites to obtained useful information about the structure of the prepared nanocomposite by using SEM, Zeiss, Germany. SEM images, Gaussian and histogram of nanocomposites are illustrated in Fig. 4 SEM micrograph analysis appeared as irregular distributed aggregated along with aspherical shape. The grain size was equal to (17.50 nm) from Gaussian [8,9].

Energy Dispersive X-ray spectroscopy (EDX) of NiO/ZnO nanocomposites

Energy dispersive x-ray spectroscopy (EDX) analysis was carried out to determine the composition of the prepared NiO/ZnO nanocomposite. The EDX spectrum in Fig. 5 indicates that the particles in the synthesized NiO/ZnO nanocomposite consisting of (O,Ni and Zn) elements confirmed the success in preparing the nanocomposite. The atomic percentage of O k is (8.53%), Ni L is (12.16 %) and Zn L is (40.48 %), Nik (11.01%) and Zn k (16.35%), respectively [10,11].

Thermo Gravimetric Analysis (TGA) for NiO/ZnO nanocomposite

TGA is an analytical technique which was used to determine the temperature decomposition of metal precursors using the NETZSCH STA 2500 TGADTA setup, operating's at room temperatures of up to (800 ˚C). The thermal properties of NiO/ZnO nanocomposite were investigated using Thermogravimetric analysis (TGA), the sample weighed approximately (10 mg) and was placed inside a platinum crucible with air or nitrogen flowing at a rate of (20 cm³ min⁻¹). As shown in Fig. 6, a major weight loss (7.269 mg) was observed in the range (0 – 200 ˚C), this weigh loss was about (104.500 %) for NiO/ZnO nanocomposite was contributed by the destruction of the water and

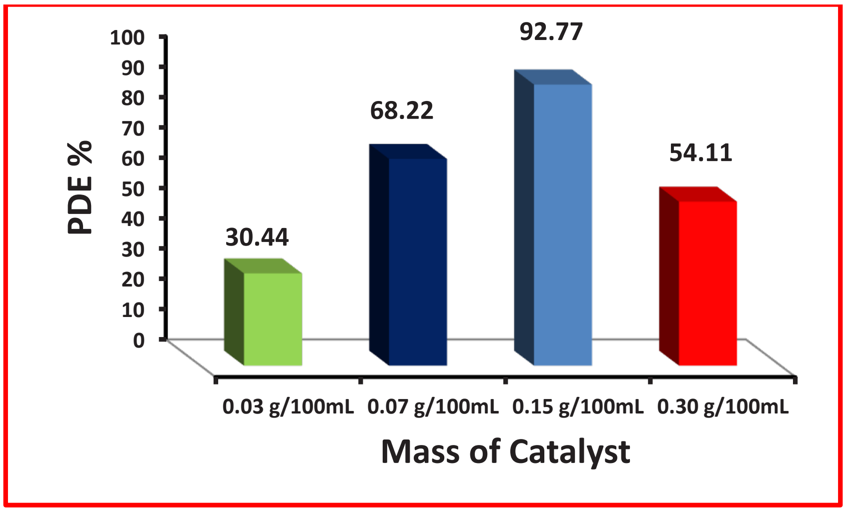


Fig. 8. Photocatalytic degradation efficiency using 0.15 g / 100 mL NiO/ZnO nanocomposite against different concentration of Fast green dye.

carbon dioxide, or the removal of impurities, and unreacted monomers of composite [12].

Effect of the mass of NiO/ZnO nanocomposite on photo catalytic degradation of the Fast green dye

The effect NiO/ZnO nanocomposite mass on the Photocatalytic degradation of Fast green dye, was studied using (10 mg/L) of the dye, air flow rate of (10mL/min), temperature of (25°C) and (pH=8). When the masses of NiO/ZnO nanocomposite increase reaching (0.15g/100mL) photo catalytic degradation of Fast green dye, gradually increases and the Photocatalytic degradation efficiency (93.89 %), then gradually diminishing. When the mass of NiO/ZnO nanocomposite (0.15g/100mL) is greatest, the semiconductor provides the greatest light absorption. The decline in photodegradation effectiveness at NiO/ZnO nanocomposite concentrations more than (0.15 g/100 mL) due to light absorption is restricted to the initial layers of Fast green dye; The photons are not transmitted to the other layers of the solution. In addition, light scattering at high NiO/ZnO nanocomposite loading causes a reduction in photon intensity. As illustrated in Fig. 7, the significant absorption of light by the initial consecutive layers of the solution prevents light from penetrating all other layers in the reaction vessel. Many researchers have explored this impact, At the loading mass of NiO/ZnO nanocomposite below the optimal value (0.15 g/100 mL), the rate of photo degradation of Fast green dye was also decreased due to a decrease in the surface area of NiO/ZnO nanocomposite, which resulted in a decrease in the light absorption of light by NiO/ZnO nanocomposite, which in turn caused a decrease in the photo degradation rate of Fast green dye [13,14].

Effect of Fast green dye initial concentration on the photocatalytic degradation process

The effect of “Fast green dye” concentration solution in the photocatalytic degradation processes in the range of (10-50 mg/L) was tested by keeping all other experimental conditions constant at a temperature of (25°C), a flow rate of air of (10mL/min), (pH=8) and mass of NiO/ZnO (0.15g/100mL).The results are plotted in Fig. 8.These results indicate the Photocatalytic rate degradation was reduced with increasing “initial dye concentration” and the Photocatalytic degradation efficiency(92.77%) . As the initial Fast green dye concentration decreases, the path length of the photon entering the solution increases. Therefore, the number of photons reaching to catalyst surface increases hence the rate of formation of hydroxyl radicals and super oxide ions also increased thereby increasing the rate of degradation [15].

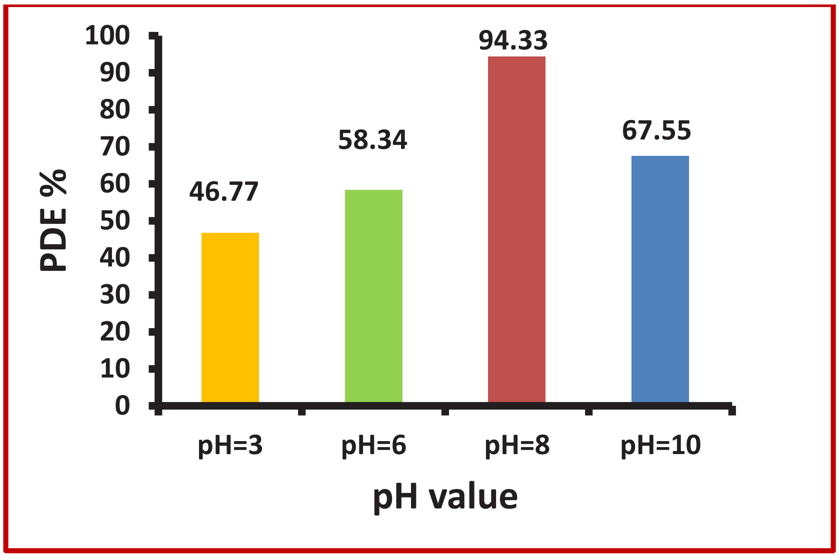


Fig. 9. Photocatalytic degradation efficiency using (0.15g / 100 mL) NiO/ZnO nanocomposite and (10 mg/L) of Fast green dye for different initial pH.

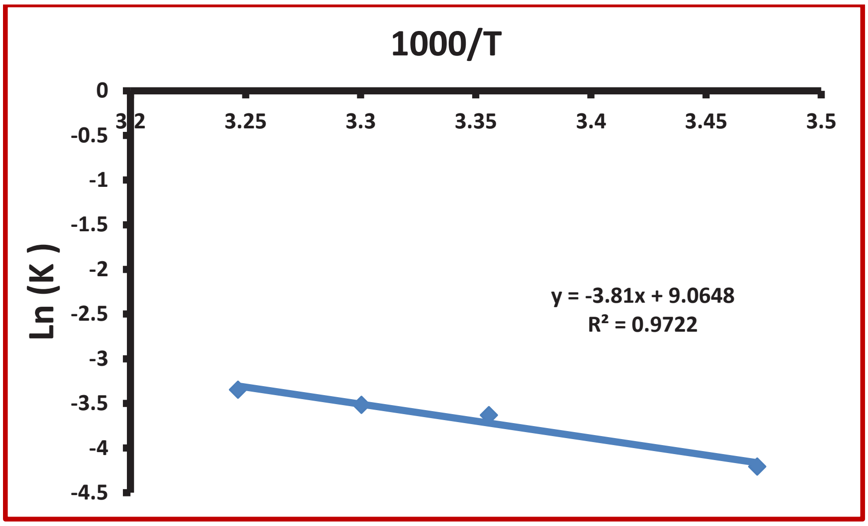


Fig. 10. Arrhenius plot of Fast green dye.

Effect of pH at photocatalytic degradation of Fast green dye

A set of works whereas performed to investigate the effect of the initial pH of suspension solution on ability of photo catalytic digradation of Fast green dye over which showed (0.15 g /100mL) of NiO/ZnO nanocomposite, (10mg/L) of "Fast green dye concentration", (10 mL/min) "flow rate" of an air bubble at temp. (25°C). The practical experiments were carried out under different pH solutions (3, 6, 8 and 10). As Shown in Fig. 9 the optimum value of degradation of "Fast green dye", was determined at (pH=8).At pH in the acidic medium the surface of the catalyst becomes acidic, which lead to an increase in the attractive force of dye molecules with the catalyst surface, hence increasing photocatalytic degradation efficiency (94.33 %) [16].Meanwhile a higher pH function in the basic medium increased the repulsion force between the molecular dye and catalyst surface hence, decreasing the photocatalytic degradation efficiency[17].

Temperature effect of on photo-catalytic-degradation for Fast green dye

Experiments were conducted to determine the temperature effect on the "photocatalytic degradation" of "Fast green dye" in range (285 – 311) kelvin. Other experimental conditions apply the constant at the initial Fast green dye concentration of (10 mg/L), prepared NiO/ZnO catalyst dosage was (0.15 g /100mL), (10 mL/min) flow rate of an air bubble and (pH of =8).The showed that the degradation process of dye progressively increased with the increase in temperature. This may be due to the increased reactive hydroxyl radical [18].The activation energy associated with the photo degradation of the dye was calculated according to the Arrhenius equation. By plotting In k versus 1/T.Shown in Fig. 10. the activation energy found was 31.67 ±1 kJ.mol^{-1}.

CONCLUSION

NiO/ZnO nanocomposite was prepared using the sonocatalysis method. The mean crystallite size for the prepared NiO/ZnO nanocomposite was calculated using the Scherer equation, and the results showed that the average crystallite size and average particle size of all photocatalysts were 35.81 nm according to the XRD. The photocatalytic degradation processes of Fast green dye depended on the amount of catalyst dosage and the optimum value was equal to (0.15 g / 100 mL) of NiO/ZnO nanocomposite. The best removal percentage of Fast green dye was (94.33%) when pH=8. The effect of dye concentration has been used to study the optimum value of Fast green dye (10mg/L) dye. The photocatalytic degradation of Fast green dye decreased with an increase in the concentration of the dye due to the decrease of the concentration OH$^-$ adsorbed on the catalyst surface. The activation energy calculated when using the Fast green dye was = 31.67 kJ.mol^{-1}.

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CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this manuscript.

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