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Photocatalytic Degradation of Textile Dye from Wastewater by using ZnS/TiO_2 Nanocomposites Material

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Abstract

Global efforts are being made to find a solution to the problem of industrial waste accumulation. When dyes are introduced into natural water reservoirs, they have a detrimental influence on the ecology. Dye removal from wastewater streams is crucial. Semiconductors are typically used to convert organic contaminants in wastewater into nontoxic inorganic compounds. This contamination attempt is currently a severe concern of public health. Decolorizing and photoctlytic degradation of textile dye in the industry effluent also helps the environment from this wastes, since textile industries are characterized chemically and water-demanding, leading in greater pollution levels and water consumption. In this work the azo Acid blue AB- 113 dye was removed using irradiation as an illumination source, and UV was utilized to show the photocatalysis process. Photocatalyst activity was evaluated to remove AB-113 dye in an aqueous solution. Three experimental factors, including pH photodegradation, dye concentration, and catalyst dose, were examined for their effects on AB - 113 removals. 97% of the dye was removed by employing a catalyst dose of 0.5 gm, an initial dye concentration of 25 mg/L, and a pH of 6.2 The photodegradation of AB – 113 adsorptions on nanocomposite ZnS over the TiO_2 surface may be explained kinetically using pseudo-second-order and modified Freundlich models.

Keywords: Nanocomposites ZnS/TiO_2; Dye removal; Azo Acid Blue 113; Photocatalysis, Wastewater, Textile Dye.

1. Introduction

Drainage of the dye of wastewater into the river, lake and natural stream from paper, carpet, textile, printing industries, leather and distillery causes serious threats because dyes damage the environmental nature and transfer poisoning to aquatic life. Biodegradation, oxidizing agents, and photodegradation are all resistant to most textile dyes [1]

When both liquid waste combustion and volume drainage are taken into account, the textile industry of wastewater is ranked as the most polluting of all major industries.

The values of tinctorial dyes used in the textile industry are incredibly high; less than one part per million (ppm) of dye concentration gives a visible coloring.

In recent years, rigorous, and new restrictions have been imposed coupled with strict enforcement related to wastewater discharge. So, there is a certain need to have technology that can function adequately under the above conditions and it must be an effective cost [2-20].

Several traditional methods of textile wastewater treatment are presently categorized as physical and chemical and biological treatments [3]. Many wastewater treatment technologies such as adsorption, membrane, separation and coagulation have been implemented [4 -5]. In addition of the using of

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adsorption methods utilizing natural low-cost materials such as rice husk, lemon peel, walnut, olive stone, broad beans, orange peels and other materials [21-30]. Others used different physical methods such as using porous media and filtration [31-40].

Recently, many authors used low-cost natural materials in wastewater treatment [6-8]. However, these technologies attempt to convert the waste from one to another phase [liquid phase to the solid phase. In addition, many researchers have proposed enhanced oxidation processes to remove a contaminant, with photocatalysis being one of the [9].

Miljana et al., studied the UV/H_2O_2, UV/persulfate, Fenton, photo-Fenton, and UV/TiO_2 degradation of eight typical pharmaceuticals and pesticides in simulated urban wastewater [10-20].

One of the most efficient methods for destroying organic contaminants in aqueous media is heterogeneous photocatalysis Numerous research have shown that TiO_2 nanoparticles could oxidize several organic wastes and convert them to inorganic materials throughout the last few decades.

The development of a TiO_2 photocatalyst to visible light by loading it with metals [6,7] or nonmetallic components such as nitrogen, sulfur, carbon, boron, and fluoride is a notable achievement in this field [1,8,9]. Furthermore, metal and nonmetallic TiO_2 materials are difficult to prepare because of the need for mesh exchange in multiple steps of the experimental procedure and a high temperature [5]., it is generally difficult to prepare metal and nonmetallic TiO_2 materials with the need for mesh exchange at multi-step of the experimental process and a high temperature [11].

TiO_2 is a widely used semiconductor photocatalyst according to its activity, non-toxic nature, strong oxidizing potential, photoresponse, high photostability, photo efficiency, and low solubility in water under various conditions.

TiO_2 drawbacks are unsuitable for practical application in environmental processes. TiO_2 can only absorb light in about 4% of the visible light spectrum because it has a high gap band about 3.2 eV wide. Due to the high recombination rate of photogenerated electron-hole pairs, which are simpler to recompose and reduce photocatalytic performance, the catalyst must be changed to absorb light in the visible region [11-13].

A suitable dye degradation process is necessary to prevent severe water contamination caused by wastewater dyes. In 2021 Badvi, and Javanbakht found that o n the surface of ZSM-5 zeolite, TiO2 is distributed, calcined at different temperatures, and subsequently precipitated nickel nanoparticles. In addition to photocatalytic degradation, additional photocatalytic processes explored included hydrogen peroxide oxidation and dye removal adsorption. The effects of beginning pH, initial dye concentration, photocatalyst dose, hydrogen peroxide concentration, nanophotocatalyst nickel content, calcination temperature, and kinetic studies on dye removal were also explored. Nickel nanoparticles improved photocatalytic dye degradation efficiency. They found that t he best results were obtained in photocatalytic hydrogen peroxide oxidation, photocatalytic hydrogen peroxide degradation, and adsorption processes. 100 percent UV photocatalytic degradation employing 0.5 percent nickel nanoparticles at 600°C [40].

The photo-sensitization of TiO_2 by metal sulfides or metal oxides has been documented in various techniques, like (CdS, ZnS, WS_2, ZnO, Fe_2O, WO_3, …etc.) [14]. Because of their nontoxicity, high stability, and photoactivity, semiconductor oxides, such as TiO2 and ZnS, have been widely used in the degradation of organic pollution particles. Furthermore, due to the effects of quantum confinement, the photocatalytic effectiveness of composite catalysts might be considerably improved when compared to pure TiO2 or ZnS [15].

Several researchers investigated the impact of employing composite TiO_2-ZnS catalyst in the degradation of various dyes by photo-catalysis process [14], [16 - 17]. In contrast, other studies investigated the removal of various types of organic contaminants using other composite photocatalysts such as MS/TiO_2 (M = Zn, Cd and Pb) studied by [11], poly-o-phenylenediamine (PoPD)/TiO_2 [18] and ZnO/ TiO_2 was studied by many authors [19].

The photocatalytic degradation of Acid Blue-113 dye [AB-113] was examined in this work using a composite ZnS-TiO2 nanoparticle catalyst.

Dyes derived from the diazO compound AB-113 are commonly used to dye various fabrics, such as wool, nylon, silk, leather, and polyester. It has also been employed in degradation and adsorption experiments to mineralize and decolorize hazardous intermediates, as well as in other applications. The creation of highly reactive atoms of oxygen [RS] such as hydroxyl and superoxide radicals to react with dyes and degrade dyes in the presence of ultraviolet or visible [UV] radiation and semiconductors is referred to as photo-catalysis in dyes.[16].

This study's objective was to prepare photocatalyst nanocomposite ZnS–TiO_2 and evaluate the

photocatalyst's activity to remove Acid Blue AB-113 dye under UV-irradiation in an aqueous solution. In addition, study the effect of experimental parameters such as pH on photodegradation, dye concentration, and catalyst dosage on the photocatalytic degradation performance of dyes using dye photocatalytic degradation.

2. Experimental

2.1 Synthesis of Photocatalyst

A specific amount of titanium isopropoxide $Ti(OCH(CH_3)_2)_4$ dissolved in anhydrous ethanol though being stirred rapidly. Next, a solution of [ethanol-water-acetic acid] is added to the titanium solution drop by drop and constantly stirred till a clear solution is made. Over time (about 60 minutes) milky hydrated gelatin is formed at 60°C. Gelatin was aged for 24 hours at room temperature after that TiO_2 nanocatalysts were made by mixing and roasting.

ZnS nanoparticles were also prepared by the precipitating method of zinc nitrate [$Zn(NO_3)_2$] and sodium sulfide [Na_2S] precursors. ZnS nanoparticles were obtained, washed, and dried at 60 °C.

Nanoparticles of ZnS/ TiO_2 were produced by droplet addition of ethylenediamine to acetone solution of the ZnS precursor and TiO_2; a solution was refluxed while the mixture was stirred and solids were gathered and evaporated at 60 °C.

2.2 Experiments of Photocatalysis

The experimental reactor used for the photocatalytic of AB-113 consisted of a beaker holding a photocatalyst suspension with AB- 113 with illumination provided by a UV light. To achieve an equilibrium of adsorption-desorption between the catalyst surface and dye molecules during illumination, the suspension was magnetically agitated for 5 minutes before illumination began [20].

2.3 The Activity of Photo-catalytic

The activity of photo-catalytic ZnS/ TiO_2 was determined by studying the AB – 113 photodegradation in an aqueous solution. By diluting AB -113 in distilled water, a dye mixture with concentrations of 25, 50, 75, and 100 mg/L was formed. Adjusted pH solution to between 3.0 and 8.0 using diluted NaOH or HCl.

Added one hundred millilitres of the solution to a 500 mL flask with different catalyst doses (0.5-1, 1.5 and 2) g. After irradiation for a specific time of (15–90) min, the photo-catalyst nanoparticles were separated by centrifugation. Then, samples were evaluated photometrically at a wavelength of 600 nm. After determination of the concentration of AB – 113 dye vs. time, the % of the amount of dye that was removed was determined using Equation (1):

$$\textbf{Dye Removal}\ \% = \frac{C_0 - C_t}{C_0} * 100 \quad \ldots . 1$$

[C_t and C_0] are the concentration at irradiation time t and the initial concentration, respectively

2.4 kinetic models of Photodegradation

The kinetic results of the Acid Blue AB-113 Dye degradation were examined using 6 kinetic models in this study on prepared composite photocatalytic ZnS/TiO_2 [15].

First-order, second-order, pseudo-first-order, pseudo-second-order, and modified Freundlich models were employed, as shown in the equation, (2, 3, 4, 5), respectively [16].

$$\ln \frac{C}{C_0} = -k\,t \quad \ldots . 2$$

$$\frac{1}{C} - \frac{1}{C_0} = k\,t \quad \ldots . 3$$

$$ln(C - C_0) = ln\,C - k\,t \quad \ldots . 4$$

$$\frac{t}{C} - \frac{t}{C_0} = \frac{1}{k\,C^2} \quad \ldots . 5$$

$$\frac{(C_0 - C_t)}{C_0} = k\,t^b \quad \ldots \ldots 6$$

The dye molecule concentration in the solution is represented in the equations above by C_0 and C, respectively, at time t. a and b are undefined chemical constants, and k is the constant rate.

3. Results and Discussion

3.1 Decolorization efficiency of UV Process

The work has conducted three experiments; the first one examined the pH effect on the photodegradation, whereas the other set considered the initial dye concentration and dosage of catalyst. Contact time was examined implicitly in sets.

3.2 The pH Effect

In the heterogeneous photocatalytic process, pH has a massive effect on the degradation mechanism of dyes.

The pH influence on dye concentration removal has been examined as the first set of tests shown in figure 1. initial dye concentration of [25 mg/L], while the dosage of the catalyst of [0.5 gm] and the pH of 4, 6.2, 8, and 10.

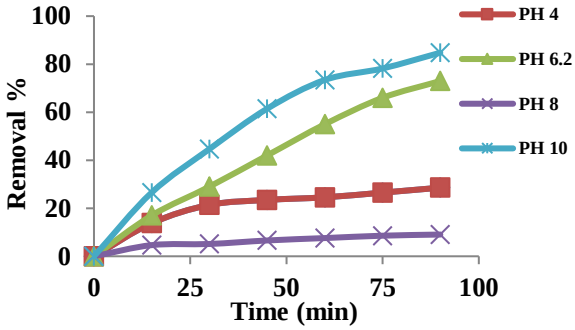

Figure 1: The pH effect on removal efficiency

The initial pH solution was determined for being (6.2), and the pH was adjusted by adding acid or base [NaOH and H_2SO_4] to reach the desired level of pH. As a result, the highest dye removal occurred at pH 10 while the smallest dye removal (9.057%) occurred at pH 8 as shown in figure 1.

The production of hydroxyl radicals would be inhibited at low pH values because of a deficient concentration of hydroxide ions and an excessive concentration of H+ this is agreed with Xue et al., 2015. A progressive decrease in AB -113 degradation occurred as a result of resistance between the photocatalyst and the negatively charged surface of OH. Furthermore, the oxidized radical is rapidly rescued at higher pH values and is unable to react with dye molecules [16][40].

3.3 Catalyst Dosage Effect

The best catalyst dosages are by oxidizing the same dye concentration with various doses of catalysts. the catalyst doses in the range of (0.5 – 2) g were examined with each other conditions held the dye concentration and pH were both kept 25 mg/L and 6.2. The maximum dye removal was obtained at a dose of 2 gm (92.5%) and the minimum dye removal was obtained at a dose of 0.5 gm (73.2%) as shown in figure 2.

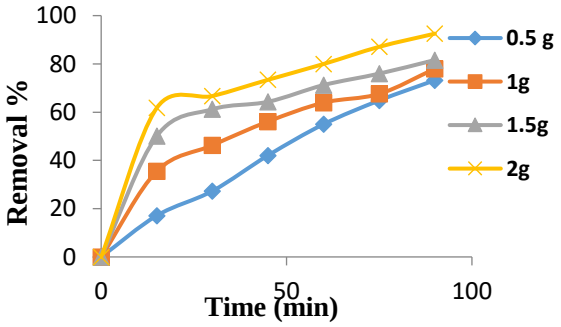

Figure 2: The Catalyst dosage effect on removal efficiency

It was noticed that the % removal of dye raised with more doses of catalyst. An increase in catalyst amount led to an increase in increased ROS generation and, as a result, dye degradation when there are more active surface sites on the ZnS-TiO2 surface [16].

3.4 The Effect Dye of Concentration

The influence of dye concentration on the efficiency of dye removal is illustrated in figure 3. At a dye concentration of 25 mg / l, the highest dye removal was obtained 73.2 %, and at a dye concentration of 100 mg / l, the least dye removal was obtained 25 % . Dye concentrations in the range of (25-100) mg/l were tested with all other conditions held at constant catalyst doses of 0.5 g and pH of 6.2, as shown in figure 3.

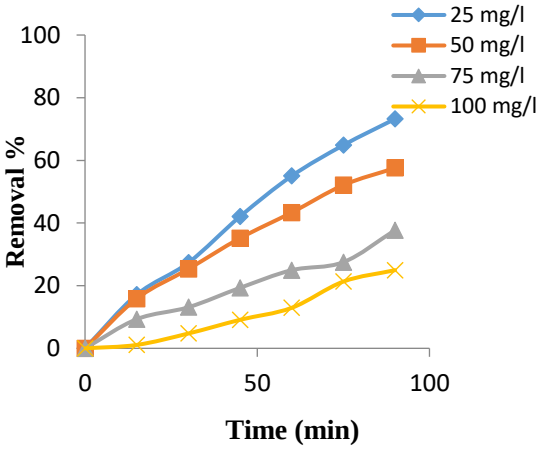

Figure 3: dye concentration effect on removal efficiency

3.5 The Kinetic Modeling of Photodegradation Process

The kinetic model of the photodegradation process can be used as an optical design tool and scale-up, The fitting of different models and their related coefficients of determination R^2 suggest that the modified Freundlich and pseudo-second-order models may more effectively describe the kinetics of acid-base 113 dye degradation on ZnS/TiO_2 surfaces. Additionally, R^2 is near to unity for these models as shown in figure (4).

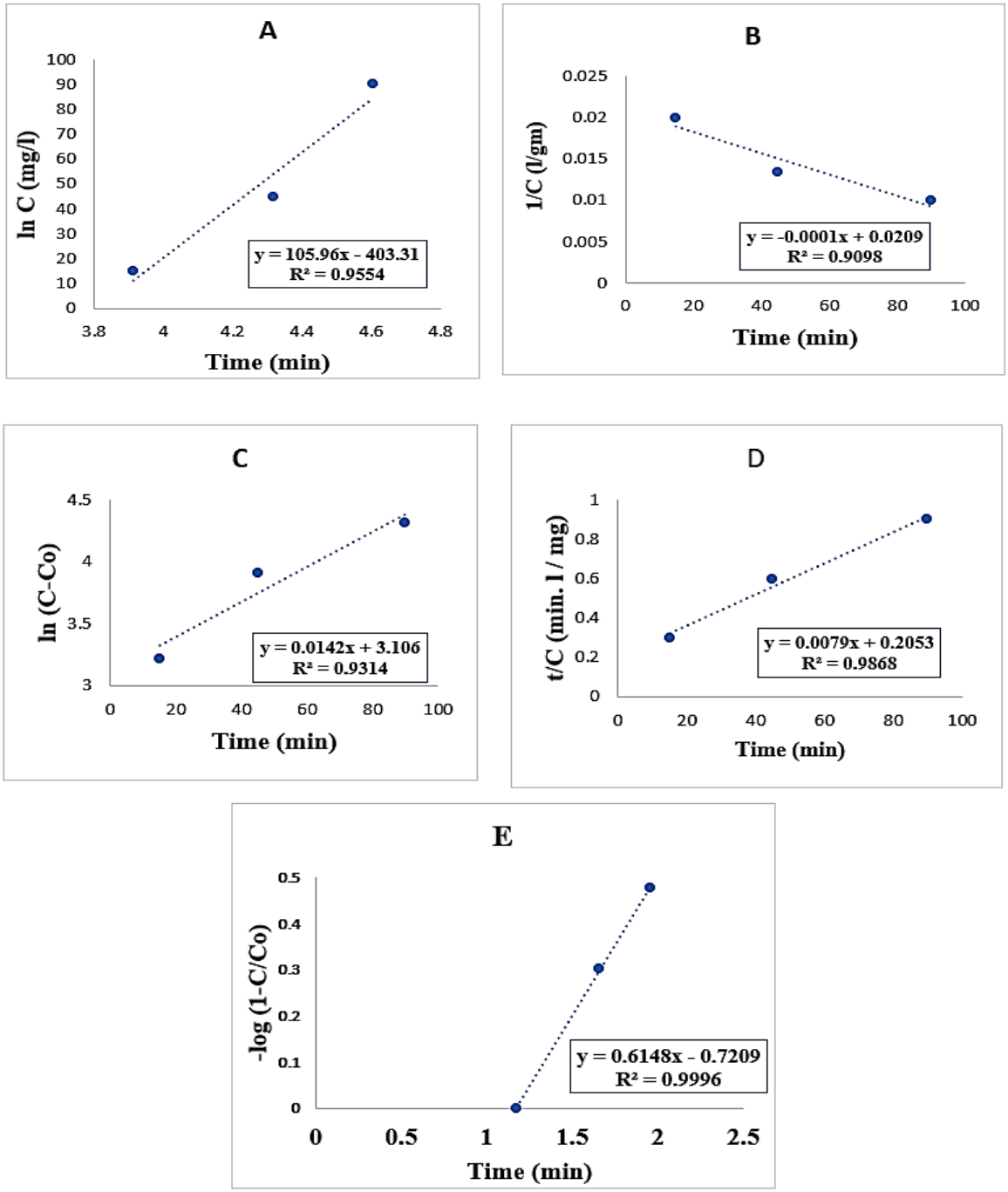

Figure 4: Kinetic rate relation of the: A- First-Order, B- Second-Order, C- Pseudo-First-Order, D- Pseudo-Second Order, E- Modified-Freundlich model.

It has been determined that the single layer chemical sorption optical electron transmission between ZnS/TiO2 particles and AB - 113 molecules is the rate-limiting phase in the pseudo-second-order mechanism [16]. Molecular ion exchange is used to explain heterogeneous diffusion from a smooth surface in the modified Freundlich model of aqueous solution [15]. It was observed that the adsorption photo-catalytic degradation of ZnS / TiO2 particles removed the dye from the solution.

4. Conclusions

Nanoparticles ZnS / TiO_2 were effectively produced and utilized to photodegrade the azo Acid Blue 113 dye in an aqueous solution in the presence of UV irradiation. The experimental parameters effects that have been studied involved pH on the photodegradation, concentration of dye and catalyst dosage were investigated.

Optimal conditions are found to be so: pH of 6.2,

catalyst dosage of 0.5gm, initial dye concentration of 25 mg / L The photodegradation of AB-113 increased as the contact time with ZnS / TiO_2increased.

The photodegradation of AB-113 on ZnS/TiO2 nanoparticles match pseudo-second-order and modified Freundlich kinetic models, respectively, for photodegradation.

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