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Research paper

Solar Photocatalytic Degradation of Food Dye (Tartrazine) using Zinc Oxide Catalyst

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Abstract

Food dyes are used in food processing industries as coloring and additive so that the food can be stored for a long time. The effluent discharged from the industries has become a serious issue as it can cause water pollution. Solar photocatalytic degradation was widely used to degrade most of the dyes in wastewater due to its high effectiveness. The main objective of this study was to investigate the effectiveness of degradation of Tartrazine using the photocatalytic process by using ZnO as photocatalyst and sunlight as the irradiation source. This study also was conducted to compare the percentage degradation of Tartrazine at different conditions which are amount of catalyst, pH of dye solution, exposure time and presence and absence of sunlight and photocatalyst. The degradation of Tartrazine was analyzed by using UV-Vis spectrophotometer. A laboratory set-up was designed to evaluate the optimum weight of ZnO, optimum pH of Tartrazine solutions, optimum time exposure and also was operated in the presence of ZnO as photocatalyst and sunlight as the irradiation source. The result showed that, the photocatalytic degradation of Tartrazine proved to be effective under solar radiation by using ZnO as photocatalyst. The optimum photodegradation occurred at optimum weight of ZnO, pH of Tartrazine and time of radiation. The optimum weight of ZnO was 20 mg, and the optimum pH of Tartrazine solution was pH 6. The most effective condition for photodegradation of Tartrazine was degradation with ZnO catalyst and with solar radiation. The radiation time was 1 hour with (93.91±0.57) % degradation. The formation of OH· required both the presence of catalyst and solar radiation.

Keywords: *Solar Photocatalytic, Tartrazine, Food Dyes, Wastewater treatment, catalyst Zinc Oxide*

1. Introduction

Nowadays, environmental pollution has become a very serious issue in the world. As the concentration of pollutants in the water streams such as rivers and lakes is high, it will cause water pollution. Industry is a huge source of water pollution, it produces harmful pollutants that will dangerously harm people and affect the environment. Industrial water pollution is caused by the released of dangerous chemicals and compounds into the water which makes it not suitable for drinking and other purposes. Food industry uses huge amounts of water, for example as the sterile water to produce food, cleaning process, for transportation and refrigeration [1]. Dye and other organic toxic compounds from food industry are very hazardous to aquatic life [2]. The presence of dye in the water even at low concentration can change its physical properties.

Dyes are widely used as additives or colorant in most of the industrial process such as food and textile. As in food industry, food dyes are used to improve the taste, appearance, and texture of the food. Besides that, food dyes are also used as additive to make the food can be stored in a longer period of time. Food dyes are usually used in production of drinks, ice cream and food concentrates. Synthetic dyes are used for restoration and providing color during foods production because natural dyes are unstable and easily undergo degradation during manufacturing process [3]. But, synthetic dyes are very dangerous since it can cause allergic reaction and also can lead to various diseases. Some of the most common used food dyes are Tartrazine, Allura Red, Erythrosine, and Brilliant Blue. Tartrazine is an azobenzene synthetic yellow dye that is reduced to aromatic amine which can cause allergic reactions [4].

Dyes contain various kinds of chemicals that can cause carcinogenic and mutagenic effects to human beings and also nature [5]. Other than that, dyes also can cause water pollution as it is released into the environment. The chemical reactions occurring in the waste water phase produces products that will adversely affects the animals and humans [6]. Therefore, it is important to treat the dyes with appropriate and efficient method before being released into the environment.

Since waste water from the industries can bring harmful effects to the environment, human being and aquatic life, therefore it must be treated properly. There are three methods that can be used to treat waste water which are physical, biological and chemical treatment. Physical treatment such as membrane filtration, adsorption and precipitation is not so effective because it only separate dye from waste water. Therefore, it is considered as non-destructive process. Other than that, biological process also has been used in the removal of dye. But, it is not favorable as most dyes are designed to be highly stable towards light and oxidative degradation. Among all of the treatment processes available, advanced oxidation process is the best as it has high efficiency of degradation of dyes. Photocatalytic degradation can destroy the toxic substances in organic constituents which are hazard to human health and also environment [7]. At ambient temperature and pressure, it can destruct wide range of recalcitrant pollutant into harmless products [5]. Photocatalysis can be employed to decompose wide range of recalcitrant pollutants to harmless compounds. Photocatalysis is a light induced catalytic process that oxidizes organic pollutants through redox reactions that occurs on the surface of metal oxides which act as photocatalyst [8].

Photocatalytic degradation by using sunlight is considered as environmentally friendly process to degrade organic compounds [9]. The most commonly used photocatalyst are Titanium Dioxide ($TiO2$) and Zinc Oxide (ZnO) because they have higher efficiency of removing wide range of organic chemicals and synthetic dyes [10]. When photocatalyst absorb sufficient amount of energy from sunlight, electron on its surface will excite to conduction band and positive hole is produced in the valence band [11].

In response to this problem, this study proposed the waste water treatment by photocatalytic degradation by using zinc oxide as the photocatalyst to degrade food dye which is Tartrazine. In this study, the dye that is used is Tartrazine. It is a synthetic dye that is used in food production to give yellow color appearance. Tartrazine is one of a group of dyes known as azo dye. An azo dye is a chemical compound where two hydrocarbon groups are joined by two nitrogen atoms. Solar irradiation has been used in this study instead of UV radiation because it is safer and cheaper [12]. Metal oxide is usually used in the treatment of waste water by photocatalysis process. ZnO is used in this study because it has high photosensitivity and it is a non-toxic photocatalyst. Besides, ZnO has high efficiency in absorption of large area of solar spectrum than TiO2 [13]. Furthermore, ZnO can convert toxic organic pollutants exist in water into non-toxic compounds by using solar radiation or UV radiation.

2. Method and Material

An easy way to comply with the paper formatting requirements is All materials in this experiment were prepared in the laboratory and of analytical grade. The model dye, Tartrazine was purchased from Bendosen Laboratory Chemicals manufacturer and a stock solution of the dye (1 M) was prepared in distilled water. The instrument UV-Vis Spectrophometer – UV 1800 Shimadzu and Solar Power Meter TES 1333 was used in this experiment. All materials were prepared in the laboratory.

The photodegradation of Tartrazine dye process was operated in different conditions which were the presence and absence of solar radiation, and the presence and absence of catalyst. Then, the percentage of degradation of Tartrazine was analyzed by using UV-Vis Spectrophotometer by tabulating the absorbance reading of the dye solution. The rate of degradation for the condition was also studied.

2.1. To Study The Optimun Weight of Catalyst, ZnO

Six 100 mL beakers were prepared. The beakers were first wrapped with black plastics. Then, 50 mL of 5 ppm Tartrazine solution were added into each beaker. Different weights of ZnO which were 0 mg, 10 mg, 20 mg, 30 mg, 40 mg and 50 mg were added into each beaker. After that, the beakers were exposed under sunlight for five hours and the black plastics were removed. During the radiation, the solutions were stirred by using magnetic stirrer. The intensity of sunlight radiation was measured by using Solar Power Meter TES 1333 for every hour of radiation. Then, its average was calculated. After five hours, the suspension from each beaker was centrifuged at 4000 rpm for 10 minutes. Then, the supernatant from each beaker was collected and its absorbance was measured under UV-Vis spectrophotometer at the maximum wavelength of Tartrazine solution.

2.2. To Study the Optimum pH of Tartrazine Solution

Six 100 mL beakers were prepared. The beakers were first wrapped with black plastics. Then, 50 mL of 5 ppm Tartrazine solution were added into each beaker. The optimum weight of ZnO was added into each beaker. The pH of the solutions was adjusted into six different pH which were pH 6, pH 7, pH 8, pH 9, pH 10 and pH 11 respectively. The pH was adjusted by using 0.01M of HCl and 0.01M of NaOH. The beakers were exposed under sunlight for five hours and the black plastics were removed. During the radiation, the solutions were stirred by using magnetic stirrer. The intensity of sunlight was obtained for every hour. Then, its average was measured. After five hours, the solutions were centrifuged at 4000 rpm for 10 minutes. Then, the supernatant was collected to be measured by UV-Vis spectrophotometer.

2.3. To Study The Optimum Time of Photocatalytic Degradation

The optimum time for all four different conditions was determined. From the absorbance obtained, the concentration of each solution was calculated by using the equation of the graph. By using the concentration calculated, the %D was calculated.

2.4. To Study The Effect of Tartrazine Degradation In The Absence Of Sunlight (without Catalyst, ZnO)

Six beakers wrapped with black plastics were added with 50 mL of 5 ppm Tartrazine solution. The pH of the solutions was adjusted to the optimum pH of the solution. After that, the solutions were stirred in a box for 0 hour (0 minute), 1 hour, 2 hours, 3 hours, 4 hours and 5 hours respectively.

2.5. To Study The Effect of Tartrazine Degradation In The Presence Of Sunlight (without Catalyst, ZnO)

Six beakers were added with 50 mL of 5 ppm Tartrazine solution. The pH of the solutions was adjusted to the optimum pH of the solution. After that, the solutions were stirred under sunlight for 0 hour (0 minute), 1 hour, 2 hours, 3 hours, 4 hours and 5 hours respectively.

2.6. To Study The Effect of Tartrazine Degradation In The Presence Of Catalyst, ZnO (without Sunlight)

A 50 mL of 5 ppm Tartrazine solution were added into six beakers wrapped with black plastics. The optimum weight of ZnO was added into each beaker. The pH of the solutions was adjusted at the optimum pH. Then, the solutions were stirred in a box for 0 hour (0 minute), 1 hour, 2 hours, 3 hours, 4 hours and 5 hours respectively. After each hour, the solution was centrifuged at 4000 rpm for 10 minutes to separate the supernatant and precipitate. The supernatant was used to analyze using UV-Vis spectrophotometer.

2.7. To Study The Effect of Tartrazine Degradation In The Presence Of Catalyst, ZnO (with Sunlight)

A 50 mL of 5 ppm Tartrazine solution were added into six beaker. The optimum weight of ZnO was added into each beaker. The pH of the solutions was adjusted at the optimum pH. Then, the solutions were stirred under sunlight for 0 hour (0 minute), 1 hour, 2 hours, 3 hours, 4 hours and 5 hours respectively. After each hour, the solution was centrifuged at 4000 rpm for 10 minutes to separate the supernatant and precipitate. The supernatant was used to analyze using UV-Vis spectrophotometer.

3. Results and Discussions

Based on the experiment, Tartrazine dye can be degraded by using ZnO photocatalyst under sunlight radiation. The percentage of degradation was calculated by using the formula:

Degradation efficiency (%) = Co – Ct x 100

Co

Where Co are the initial concentration of Tartrazine and Ct are concentration at time.

The maximum wavelength of Tartrazine was obtained by measuring the absorbance of 5 ppm Tartrazine solution in the wavelength range of 350 nm – 470 nm. Fig. 1 showed the wavelength and the absorbance obtained from this experiment. The maximum wavelength was the highest peak in the graph of absorbance versus wavelength. Based on Figure 1, the highest peak was at wavelength 258 nm. But it was not considered as maximum wavelength of Tartrazine since in the range of 200 – 300 nm, there was interference from the plastic cuvettes. Therefore, 427 nm was the actual maximum wavelength of Tartrazine.

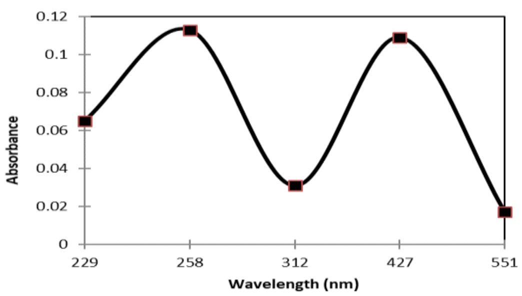

Fig. 1: Maximum wavelength of Tartrazine

3.1. The Optimum Weight of Catalyst, ZnO

Table 1: Optimum Weight of ZnO

Weight of Zinc Oxide (mg)	Concentration, ct (mol/L)	Percentage of Degradation (%)
0	5.4139	-8.96
10	1.6732	66.32
20	1.1610	76.63
30	3.0537	38.54
40	1.4282	71.25
50	1.4505	70.81

Different weights of ZnO which were 0 mg, 10 mg, 20 mg, 30 mg, 40 mg and 50 mg was added into six beakers. After exposed to sunlight radiation for five hours, the absorbance of all solutions was measured using UV-Vis spectrophotometer. Before the solutions were analyzed, the standard solutions of 1 ppm, 2 ppm, 3 ppm, 4 ppm and 5 ppm of Tartrazine were analyzed. Table 1 showed the concentration obtained and the percentage of degradation calculated.

From Fig. 2, the equation was y=0.0449112x + 0.00785675 with correlation coefficient (r2) of 0.99883. The closer the value of r2 to 1, the more excellent the linearity obtained.

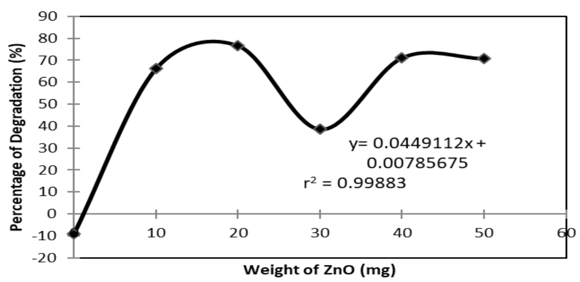

Fig. 2: Optimum Weight of ZnO

Based on the result obtained, 20 mg of ZnO had the highest percentage of degradation which was 76.63%. The amount of catalyst added increased the amount of hydroxyl radical produced. This is because the active surface sites increased, more light was absorbed caused the formation of more hydroxyl radical [14]. Therefore, the more Tartrazine dye degraded. When excess amount of ZnO was added which was 30 mg, the percentage of degradation decreased which was 38.54%. This happened because less sunlight radiation was penetrated through the solution due to the suspended catalyst in the solution [15].

3.2. The Optimum pH of Tartrazine Solution

Before the solutions were analyzed, the standard solutions of 1 ppm, 2 ppm, 3 ppm, 4 ppm and 5 ppm of Tartrazine were analyzed. Table 2 showed the concentration and percentage of degradation calculated.

Table 2: Concentration and Percentage Of Degradation

pH of Tartrazine	Concentration, c_t (mol/L)	Percentage of Degradation (%)
pH 6	1.8097	63.68
pH 7	1.8319	63.23
pH 8	2.3866	52.10
pH 9	3.4517	30.73
pH 10	4.3392	12.91
pH 11	4.1173	17.37

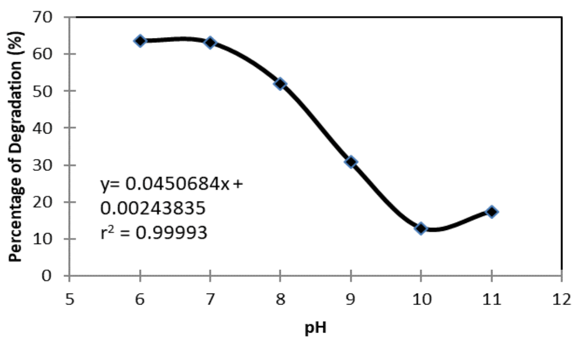

Fig. 3: Optimum pH of Tartrazine Solution

From Fig. 3, the equation was y=0.0450684x + 0.00243835 with correlation coefficient (r2) of 0.99993.

Commonly, the industries released wastewater with a wide range of pH [16]. The initial pH of dye solution is an important factor in photocatalytic degradation process. From the data obtained, the optimum pH for Tartrazine solution was pH 6. The percentage of degradation was 63.68%. The Tartrazine was dissolved in aqueous solution, and the sulphonate groups at the Tartrazine (D—SO3Na) was dissociated and converted into anionic dye ions (D—SO3Na ⇋ D—SO3- + Na+) (Gautam et al., 2015). Therefore, in acidic solution, the presence of hydronium ion (H3O+) in the solution attracted Tartrazine dye. This contributed to the higher removal efficiency of the dye.

However, in alkaline solution, the presence of hydroxyl ion (OH-) competed with the anionic sulphonic groups of Tartrazine molecules. Thus, decreasing the attraction ability of Tartrazine in the solution. Therefore, the highest pH in this experiment which was pH 11 had the lowest percentage of degradation which was 17.37%.

3.3. The Optimum Time of Photocatalytic Degradation

Four conditions were set up to determine the optimum time for photocatalytic degradation process. For the first condition which was degradation with catalyst and without sunlight, the optimum time obtained was 3 hour. For the second condition which was degradation without catalyst without sunlight, the optimum time was 5 hour. The third condition, degradation with catalyst with sunlight, the optimum time obtained was 1 hour. For the forth condition which was degradation without catalyst with sunlight, the optimum time obtained was 2 hour. The concentration and percentage of degradation calculated was tabulated in Table 3.

Table 3: Optimum Time for Various Conditions

Time (hour)	Percentage of Degradation (%)			
	With Catalyst Without Sunlight	Without Catalyst Without Sunlight	With Catalyst With Sunlight	Without Catalyst With Sunlight
0	24.10	-0.90	12.02	-0.91
1	24.58	30.37	92.03	-12.15
2	10.82	21.87	91.11	3.60
3	45.23	14.74	61.05	0.44
4	20.65	-0.46	83.24	0.89
5	0.50	30.81	78.61	2.70

Fig. 4 showed the differences in percentage of degradation for all four conditions.

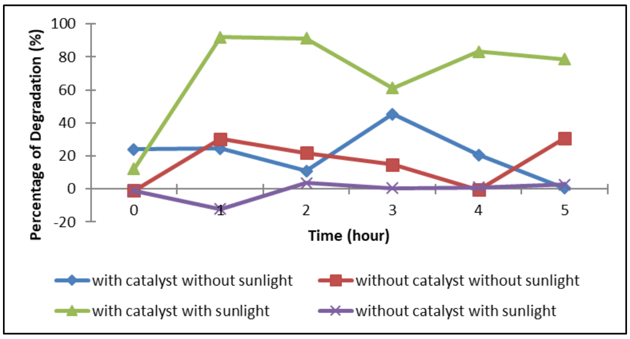

Fig. 4: Optimum Time for Different Conditions

Based on the graph plotted, among these four conditions, the highest percentage of degradation was degradation with catalyst with sunlight. The optimum time was 1 hour with the percentage of degradation 92.03%. Among all four conditions, this condition had the shortest optimum time with highest percentage of degradation. This is because, both sunlight radiation and catalyst caused the photodegradation process to occur effectively. The energy from sunlight caused the formation of free electron in the conduction band and also hole in the valence band. The electron produced reacted with oxygen gas (O2) in the solution while the hole reacted with hydroxyl ion (OH-) to form hydroxyl radical (OH·) which then attacked the dye molecule to degrade it [17].

$$hv + ZnO \rightarrow h^{+} + e^{-} \qquad (1)$$

$$e - + O_2 \rightarrow O^{2-} \qquad (2)$$

$$h^{+} + OH- \rightarrow OH\bullet \qquad (3)$$

While the condition with lowest percentage of degradation was degradation without catalyst with sunlight. The optimum time was 2 hour with percentage of degradation 3.60%. Supposedly, degradation without catalyst and without sunlight was the condition that had the lowest percentage of degradation among these four conditions. This is because there was no energy from the sunlight and no catalyst to form hydroxyl radical. Some error might occur during the experiment and affected the result obtained. The error was the beakers were not perfectly covered with black plastics and this enable the sunlight to penetrate through the solutions.

3.4. The Effectiveness of Photocatalytic Process

The determination of effectiveness of the process was done by doing three repetitions for every condition. Then, the average concentration was used to calculate the percentage of degradation. The Tartrazine solutions were added with the optimum weight of ZnO which was 20 mg, set at optimum pH which was pH 6, radiated under sunlight for optimum time for each condition. After that, the percentage of degradation for all conditions was compared.
From Table 4, the degradation of Tartrazine dye with catalyst ZnO and with sunlight radiation was the most effective condition to degrade Tartrazine dye. The percentage of degradation was (93.91±0.57)%. The least effective condition was degradation of Tartrazine dye without catalyst ZnO and without sunlight radiation. Its percentage of degradation was (0.29±0.67)%.

Table 4: Percentage of Degradation for All Conditions

Condition	Percentage of Degradation (%)
With catalyst without sunlight	7.74±2.20
Without catalyst without sunlight	0.29±0.67
With catalyst with sunlight	93.91±0.57
Without catalyst with sunlight	1.17±0.51

4. Conclusion

As a conclusion, the photocatalytic degradation of Tartrazine proved to be effective under solar radiation by using ZnO as photocatalyst. From the result obtained, the amount of photocatalyst ZnO must be added at the optimum weight which was 20 mg so that more hydroxyl radical produced. The optimum pH for the Tartrazine solution was pH 6. The percentage of degradation of Tartrazine decreased as the pH of the solution was increased. For the degradation with ZnO catalyst and with solar radiation, the optimum time obtained was lowest which was 1 hour. This condition also was the most effective condition compared to other condition because the percentage of degradation was the highest (93.91±0.57)%. The other conditions used were presence of catalyst absence of sunlight, absence of catalyst presence of sunlight and absence of both catalyst and sunlight. Both catalyst and solar radiation were needed to achieve highest efficiency of photocatalytic process because both helped in the formation of hydroxyl radical.

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