International Journal of Natural Science Studies and Development (IJOSS)
Volume 01 Issue 02, 2024, pp 103-113
E-ISSN: 2615-1383
DOI: https://doi.org/10.55299/ijoss.v1i2.10

Degradation of Triadimefon Compounds in Bayleton 250 EC Pesticides by Sonolysis, Ozonolysis and Sonozolysis

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Abstract

The increase in the agricultural sector is supported by the use of pesticides to kill pests and diseases in plants. However, the continuous use of pesticides in excessive amounts can have a detrimental impact on the environment, especially the waters around agricultural land. Therefore, efforts are needed to degrade pesticide residues containing triadimefon compounds that pollute the environment such as sonolysis, ozonolysis, and sonozoisis methods. These three methods are organic compound degradation methods that utilize OH radicals to degrade organic compounds. Based on the data obtained, the sonolysis method with the addition of TiO2-anatase catalyst produced a degradation percentage of 76.25% after degradation for 150 minutes. While in a much shorter time, namely 15 minutes, using the ozonolysis method without the use of a catalyst, triadimefon can be degraded by 69.10%, and 71.87% with the sonolysis method. Since only a small amount of triadimefon is degraded by the sonozolysis method, it is more effective to use the ozonolysis method to degrade this triadimefon compound.

Keywords: *sonolysis, ozonolysis, sonozoisis*

1. INTRODUCTION

Pesticides, "Pest Killing Agents" are drugs or chemical compounds that are generally toxic, used to eradicate plant pests, both pests, diseases and weeds caused by fungi, bacteria, viruses, nematodes (root-destroying worms), snails, rats, and other animals that are considered harmful. (Sudarmo, 1991)

Triadimefon is a chemical compound that is present in the triazole group (Dukkanci, 2005). The effects triadimefon contamination include weight loss, decreased production of red blood cells, cancer, and a decrease in high doses (Dukkanci, 2005).

Sonolysis is a method used to degrade organic compounds in water media using ultrasonic waves operating at a frequency of 20-500 kHz (Destaillets, 2001). Ozonolysis is a method for degrading organic compounds using ozone (O3). Sonozoisis is a combination of sonolysis and ozonolysis degradation methods, in which OH radicals are produced by the decomposition of ozone (O3) and sonolysis (Wang J., 2005).

TiO2 was used as the catalyst in the degradation process has been developed. Previous research results showed that Rhodamine B was degraded by 90% using a TiO2-anatase catalyst in sonolysis for 6 h, and by 68.48% after the addition of TiO_2-rutile (Arif, 2007). The use of the TiO_2-anatase catalyst has also been carried out for the degradation of Sudan I, which resulted in a degradation percentage of 100% after irradiation for 180

History:
Received : November 19, 2024
Revised : December 20, 2024
Accepted : December 27, 2024
Published : December 30, 2025

Publisher: Inovasi Pratama Int. Press

min (Safni, 2008a), naphthol blue black was degraded by 100% after irradiation for 60 min (Safni, 2007) and alizarin was degraded by 100% after irradiation for 30 min (Safni, 2008b).

Formulation of the problem

Based on the description above, the problem underlying this research can be formulated, namely whether sonolysis, ozonolysis, and sonozolysis methods can degrade the triadimefon compound, and how large the percentage of degradation of the triadimefon compound is using these methods.

Research purposes

The objectives of this study were to determine the percentage of degradation of triadimefon compounds using sonolysis, ozonolysis, and sonozolysis methods and to compare the degradation results of these three methods.

Benefits of research

This research should be useful for environmental observer institutions, Environmental Impact Analysis (AMDAL), environmental study centers (PSLH), and other related institutions. For the industry, especially the pesticide producing industry and other industries that use hazardous organic compounds, to be able to utilize this method in processing industrial waste.

LITERATURE REVIEW

Triadimefon

Triadimefon is an organic chemical compound belongings to the triazole group (Sieke, 2005). This triadimefon compound is an active ingredient in one type of fungicide with the trademark Bayleton 250 EC, which is used by farmers to control fungi on cashew, cocoa, rubber, soybean, coffee, tea, and tobacco plants. It is very stable in water and does not immediately undergo hydrolysis (Sieke, 2005).

Triadimefon has a general formula of $C_{14}H_{16}N_3O_2Cl$. The chemical name of triadimefon based on IUPAC is 1-(4-chlorophenoxy)-3,3-dimethyl-1-(1H-1,2,4-triazol-1-yl) butanone (2). The molecular weight of triadimefon is: 291.73 g/mol, its solubility in water is 260 mg/L at 20 °C , it is generally soluble in organic solvents and has a melting point of 82.3 °C (Sieke, 2005). The structure of triadimefon is shown in Figure 1.

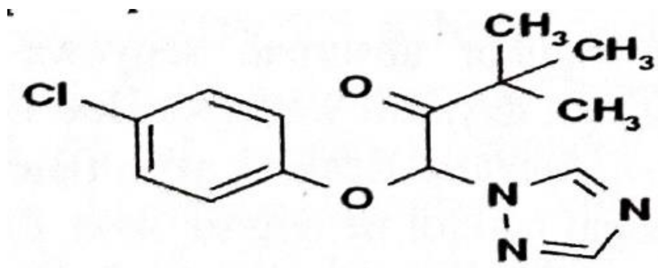


Figure 1. Structure of Triadimefon

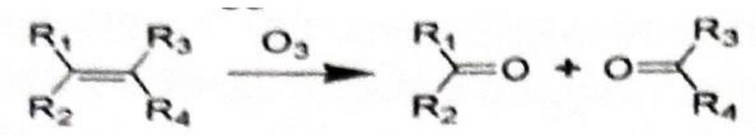
Sonolysis

Sonolysis is method used to degrade organic compounds in water media using ultrasonic waves operating at a frequency of 20-500 kHz (Destailauts, 2001). Ultrasonic waves in wastewater have the ability to degrade compounds that are difficult to decompose because in the process it will produce hydroxyl radicals and cavity effects (Stock, 2000). The effect of sonolysis on water solutions is to break down water into H and OH radicals, which can damage the organic compounds in solution.

2.3 Ozonolysis

Ozonolysis is a method of organic compound degradation using ozone (O_3), in which the C=C bond is broken down to produce a double bond of C=O (Tietze, 1998). The results of this degradation depend on the type of double bond oxidized and working conditions.

The degradation reaction using ozone is as follows:



Combined Sonolysis and Ozonolysis (Sonozolysis)

The combination of sonolysis and ozonolysis (sonozolysis) is a degradation method that involves flowing ozone (O_3) or ozonation into a solution irradiated by ultrasonic waves. During ozonolysis, ozone is decomposed by hydroxyl ions (OH) into OH and H_2O_2 radicals in the water phase. With ultrasonic irradiation during the sonolysis process, water is also decomposed into OH radicals in the presence of cavity bubbles.

Titanium Dioxide and Its Characteristics as a Semiconductor Catalyst

Some of the characteristics of TiO_2 are that it is a semiconductor, where by providing low energy light, the surface of TiO can produce hydrogen gas from water and produce electric current directly (Gunlazaurdi, 2001).

The availability of positive holes on the surface of TiO_2 produces hydroxyl radicals (•OH), which are known oxidizing species.

Applications of TiO_2 include purification, disinfection, wastewater treatment, and hazardous waste control. TiO_2 is often used because it is stable and relatively inexpensive.

2. METHOD

. Place and Time of Research

This research was conducted at the Applied Analytical Chemistry Laboratory, Department of Chemistry, Faculty of Mathematics and Natural Sciences, Andalas University, Padang, from November 2022 to March 2023.

Tools and materials

- Tool

The tools used in this study were a UV/Vis Spectrophotometer (S.1000 Secomam, Sarcelles, France) to measure the absorbance of triadimefon compounds, Ultrasonic VC-1 with a frequency of 45 kHz and a power of 60 watts (As One Comp, Japan), used to degrade triadimefon compounds, ozone reactor (Bio-ozone space age sterilizer, Japan), which was used as an ozone-producing reactor, membrane filter (Advantac membrane filter brand, polymer: mixed cellulose ester (0.45 µm, 25 mm)), which was used to filter TiO_2-anatase from the solution. An analytical balance was used to weigh TiO_2-anatase, a pH meter was used to adjust the pH, a thermometer was used to measure the temperature, and glassware was used to make reagents.

- Material

The materials used in this study were a solution of triadimefon in Bayleton 250 EC pesticide (PT. Bayer Indonesia), which was purchased from an agricultural supply store, TiO_2-anatase (Ishihara Sangyo, Ltd., Japan), ethanol, and distilled water.

Work procedures

- Preparation of Triadimefon Solution 200 mg/L

To prepare a 200 mg/L triadimefon solution, 40 µL of Bayleton 250 EC solution was pipetted and placed into a 50 mL measuring flask. The solution was diluted using ethanol and the aqueous treatment at a ratio of 3: 7, according to the optimum solvent conditions in previous studies.

- Triadimefon Spectrum Measurement

The stock solution of triadimefon 200 mg/L was diluted to (10 mg/L, 20 mg/L, 30 mg/L, 40 mg/L, and 50) mg/L. The absorption spectra of each of the five variations in solution concentration were measured using a UV-Vis spectrophotometer. Then, the absorbance data were recorded at a wavelength that gave the maximum absorption.

- Degradation of Triadimefon by Sonolysis Without Addition of TiO_2-Anatase

Determination of Optimum Degradation Time

A total of 20 mL of 40 mg/L triadimefon solution was placed in a sonolysis bottle. The samples were sonicated for various times (30 min, 60 min, 90 min, 120 min, and 150) minutes. The absorbance of each sonolyzed solution was measured using a UV/Vis spectrophotometer. Furthermore, the percentage degradation was calculated.

- Degradation of Triadimefon by Sonolysis with the addition of TiO_2-anatase

a. Determination of Optimum Degradation Temperature

Triadimefon solution (20 mL, 40 mg/L) was added to each of five sonolysis bottles. Then, 16.0 mg of TiO_2-anatase was added to each solution. Furthermore, Degradation was carried out at temperatures of 25°C, 30°C, 35°C, 40°C ± 1°C for 30 min. The degradation products were filtered using a membrane filter to separate the TiO_2-anatase from the solution. The absorbance of each sonolyzed solution was determined using a UV/Vis spectrophotometer. Furthermore, the percentage degradation was calculated.

b. Determination of Optimum Degradation Time

A total of 20 mL of 40 mg/L triadimefon solution was placed into six sonolysis bottles. Then each solution was added with 16.0 mg of TiO_2-anatase was added to each solution. Furthermore, degradation was carried out with variations in time (30, 60, 90, 120, and 150 min) at the optimum temperature (procedure 3.3.4 a). The degradation products were filtered using a membrane filter to separate the TiO_2-anatase from the solution. The absorbance of each sonolyzed solution was determined using a UV/Vis spectrophotometer. Furthermore, the percentage degradation was calculated.

- Degradation of Triadimefon by Ozonolysis

A total of 20 mL of 40 mg/L triadimefon solution was placed in an Erlenmeyer flask. Then, degradation was carried out by flowing ozone into the solution at various times (5, 10, 15, and 20 min). The absorbance of each ozonolyzed solution was determined using a UV/Vis spectrophotometer. Furthermore, the percentage degradation was calculated.

- Degradation of Triadimefon by Combined Sonolysis and Ozonolysis (Sonozolysis)

A total of 20 mL of 40 mg/L Triadimefon solution was placed in an Erlenmeyer flask and placed in a sonolysis apparatus. Then, degradation was carried out by flowing ozone while sonolyzing at various times (5, 10, 15, and 20 min).

The absorbance of each solution was determined using a UV/Vis spectrophotometer. Furthermore, the percentage degradation was calculated.

To calculate the percentage degradation value, the following equation is used:

$$\text{Persentase degradasi} = \frac{A_{awal} - A_{akhir}}{A_{awal}} \times 100\ \%$$

Keterangan :

A_{awal} = absorban awal

A_{akhir} = absorban akhir

3. RESULTS AND DISCUSSION

. Triadimefon Spectrum Measurement

Triadimefon solution was prepared with various concentrations (10, 20, 30, 40, and 50 mg/L) in ethanol solvent: aquadest (3: 7) was measured its absorption spectrum using a UV/Vis spectrophotometer. The absorption spectra are shown in Figure 2.

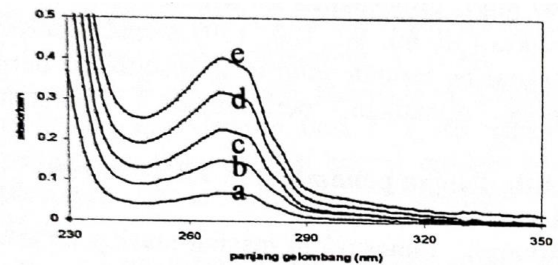


Figure 2. Absorption spectrum of triadimefon, with varying concentrations

a = 10 mg/L, b = 0 mg/L, c = 30 mg/L, d = 40 mg/L, e = 50 mg/L

From Figure 5, it can be seen that there is a linear relationship between absorbance and solution concentration. With an increase in the solution concentration, the absorbance increased linearly. This indicates that UV/Vis spectrophotometry can be used for the quantitative analysis of triadimefon compounds.

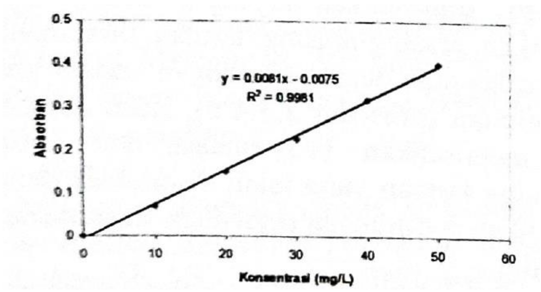


Figure 3. Calibration curve of triadimefon standard

Degradation of Triadimefon by Sonolysis without Addition of TiO_2-anatase

As shown in Figure 4, the percentage of degradation produced without the addition of the catalyst reached the optimum at 150 min, that is, 19.16%.

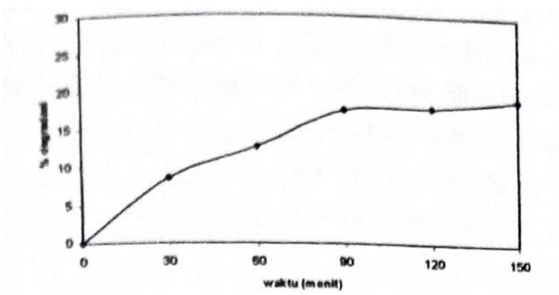


Figure 4. Effect of sonolysis time on the percentage of triadimefon degradation.

With increasing degradation time, the number of degraded compounds also increaseds. With the addition of this cavity effect, more water is broken down into OH and H radicals, which play an important role in the degradation of toxic organic compounds (Wang, 2005).

Degradation of Triadimefon with Addition of Catalyst

a. Effect of Temperature on Degradation Percentage

From Figure 5, it can be seen that the temperature variation from (25 to– 40) °C does not have a significant effect on the degradation percentage, which is approximately 25%. However, at higher temperatures (> 40 °C), the degradation percentage began to decrease. This is because at high temperatures the absorption capacity of TiO_2-anatase decreases, which causes a decrease in the degradation efficiency (Wang, 2005).

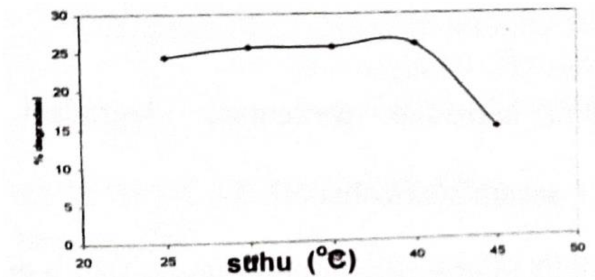


Figure 5. Effect of temperature on the percentage of triadimefon degradation

b. Effect of Time on Degradation Percentage

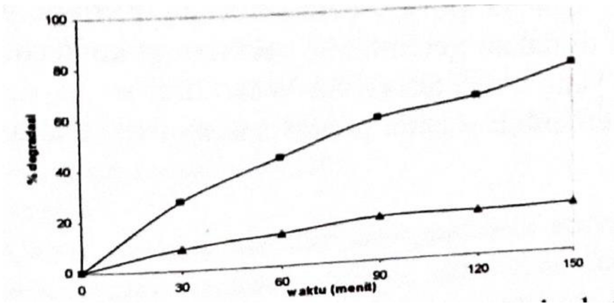


Figure 6. Effect of time variation on the percentage of triadimefon degradation

▲ = without addition of TiO_2, ■ = with addition of TiO_2

The percentage of triadimefon degradation increases with increasing time (Safni, 2008d), because the longer the sonication time, the more OH radicals are formed during the degradation process (Wang, 2005).

Degradation of Triadimefon by Ozonolysis

Figure 7 shows that, by flowing ozone (ozonation), the degradation process takes place in a short time. With increasing degradation time, ozone (O_3) can produce more OH radicals, and within 15 min, degradation reaches 69.10%. OH radicals can attack pesticide compounds that are not only on the surface of the solution, but can also attack compounds in the solution (Ruan, 2004).

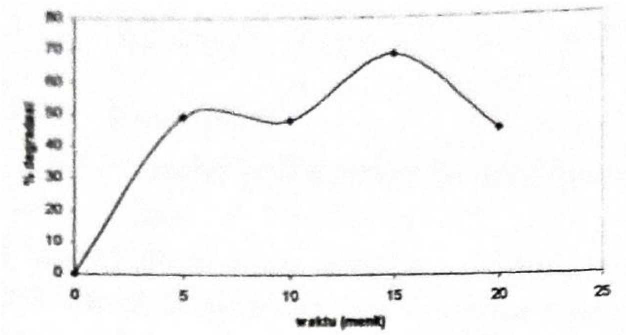


Figure 7. Effect of time variation on the percentage of triadimefon degradation by ozonolysis

Degradation of Triadimefon by Combined Sonolysis and Ozonolysis (Sonozolysis)

Figure 8 shows the combined sonolysis and ozonolysis (sonozolysis) methods in degrading triadimefon.

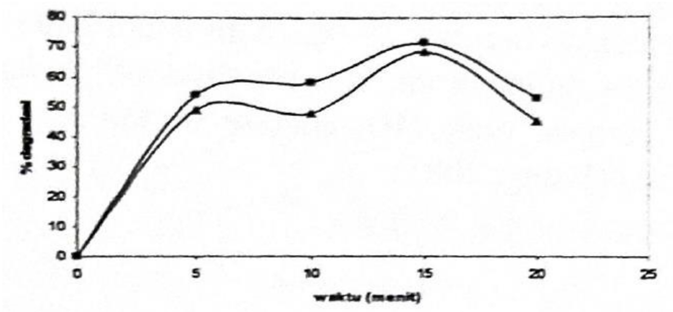


Figure 8. Effect of time variation on the percentage of triadimefon degradation

▲ = sonolysis ■ = sonozolysis

During ozonolysis, ozone is decomposed by hydroxyl ions (OH) into OH radicals and H_2O_2 in the aqueous phase. Meanwhile, with ultrasonic irradiation of the sonolysis process, water also decomposes into OH radicals in the presence of cavity bubbles. Thus, with the ozonation process accompanied by the sonolysis process, ozone decomposition occurs in the cavity bubbles, which increases the number of OH radicals formed (Xian-Wen, 2005).

The spectrum obtained during sonolysis is shown in Figure 9.

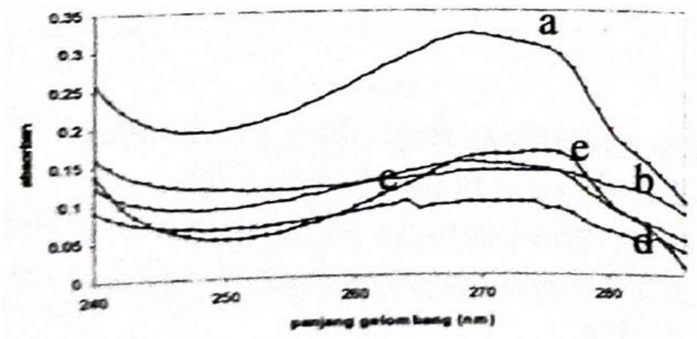


Figure 9. Absorption spectrum of triadimefon during the sonolysis process.

a = before degradation, b = 5 min, c = 10 min, d = 15 min, e = 20 min

For the spectra at 10 and 15 min, a peak shift was observed, namely in the 274 nm region. This absorption shift to a larger wavelength is called the bathochromic effect (red shift). This bathochromic effect occurred when the length of the conjugated system increased. Consequent, a larger wavelength (Creswell, 1982).

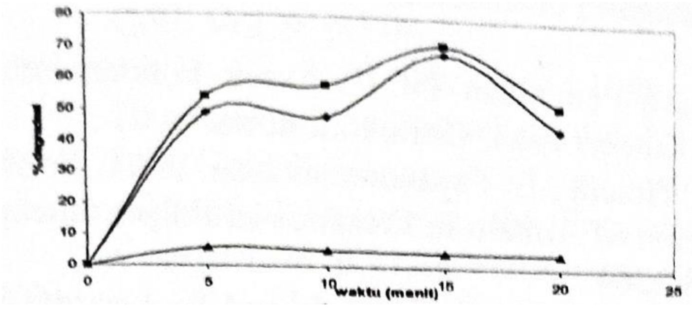


Figure 10.

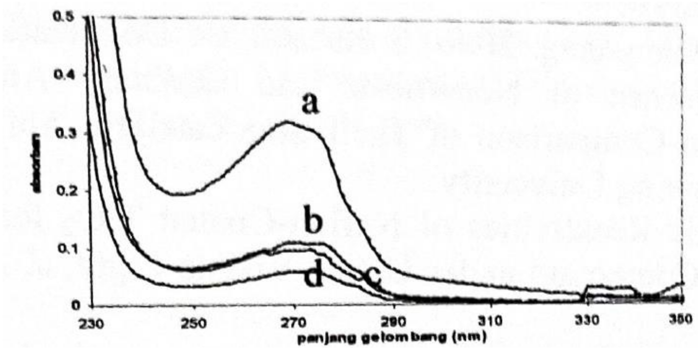


Figure 11. Absorption spectrum of 40 mg/L triadimefon solution

a, = before degradation b, = after ozonolysis; c, =after sonolysis; d, = after sonolysis

4. CONCLUSIONS AND SUGGESTIONS

Conclusion

Based on the obtained data, the sonolysis method with the addition of TiO_2-anatase catalyst resulted in a degradation percentage of 76.25% after degradation for 150 min. In a much shorter time (15 min), using the ozonolysis method without the use of a catalyst, triadimefon was degraded by 69.10%, and 71.87% with the sonolysis method. Therefore, there is only a slight increase in the percentage of triadimefon degraded by the sonolysis method, and it is more effective to use ozonolysis to degrade this triadimefon compound.

Suggestion

To identify and analyze the compounds produced from the triadimefon degradation process by sonolysis, ozonolysis, and sonozolysis, it is recommended to identify the degradation results using chromatographic methods such as HPLC and GC-MS.

5. ACKNOWLEDGMENTS

I would like to express my sincere gratitude to all those who supported me throughout this research. Special thanks to the Applied Analytical Chemistry Laboratory, Faculty of Mathematics and Natural Sciences, Andalas University, for providing the necessary facilities and equipment. I am also grateful to my colleagues for their invaluable assistance and guidance. Lastly, I extend my appreciation to my family and friends for their unwavering encouragement and support during this journey.

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