



# REMOVAL OF PARACETAMOL DRUGS FROM PHARMACEUTICAL WASTE WATER USING TiO<sub>2</sub>/SOLAR AND ZnO/SOLAR

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## ABSTRACT

Pharmaceutical waste is one of the major complex and toxic industrial wastes. Pharmaceutical industry produces a wide variety of products. This industry uses both inorganic and organic as raw materials the latter being either of synthetic or of vegetable and animal origin. Antibiotics and vitamins are produced by fermentation of fairly complex nutrient solutions of organic matter and inorganic salts by fungi or bacteria. Photocatalyst system is selected as an attractive choice in organic effluent treatment due to its properties. This process has been widely investigated as a promising technology for the efficient wastewater treatment since the photocatalyst is an environmentally friendly process and has considerable advantages such as the ability to destroy pollutants without the exertion of potentially hazardous oxidants. The effect of concentration on % reduction of paracetamol using ZnO/Solar and TiO<sub>2</sub>/Solar process. For concentrations 5, 10, 15, 20, 25 and 30 ppm solutions % reduction of paracetamol using ZnO/Solar process are 78, 70, 65, 61, 54, 51 and using TiO<sub>2</sub>/solar process are 80, 73, 68, 64, 57, 54 resp. for 120-150 contact time. As per observation as the concentration of paracetamol increase, rate of reduction decrease. As per observations both the processes are economical and having same efficiency for paracetamol reduction. For TiO<sub>2</sub>/Solar Process gives more reduction of drugs than the ZnO/Solar process. There is little bit difference in reduction of paracetamol between two processes. Both the processes are effective for paracetamol reduction. As the concentration of pollutant increase the rate of reduction decreases. As per observation the maximum reduction for paracetamol drugs from synthetic wastewater is 72.5 and 69.44% at 150 min time with 10 ppm concentration of dugs for TiO<sub>2</sub>/Solar and ZnO/Solar process resp.

**Keywords –TiO<sub>2</sub>/Solar Process, ZnO/Solar Process, Reduction of Paracetamol, Concentration, pH, Contact Time.**

## Introduction

Due to rapid growth of population and industrialization the requirement of water increases but the natural source of water which is useful for the domestic and industrial uses is very limited. From the industrial process the large amount of waste water is coming out treatment of this waste is necessary to protection of environment and human being from harmful effect. Availability of water is becoming an increasing concern in the globalized world, both in developed and in developing countries. A sustainable use of water sources could result in the search of additional water sources or even in recycling wastewater treatment plant effluents. Goal of biological wastewater treatment is stepwise oxidation of organic pollutants.

## Application of AOPs in Wastewater Treatment

1. Chemical Industry
2. Pharmaceutical Industry
3. Pulp and Paper Industry
4. Textile Industry
5. Food Industry

6. Landfill Leachates
7. Dye-Process Industrial Waste
8. Pre-treatment to wastewater
9. Organic pollutant destruction
10. Toxicity reduction
11. Biodegradability improvement

### Materials

#### 1. Hydrogen Peroxide (H<sub>2</sub>O<sub>2</sub>)

This is the strong oxidant and its application in the treatment of various inorganic and organic pollutants is well established. H<sub>2</sub>O<sub>2</sub> consist of two hydrogen molecules and two oxygen molecules.

#### 2. Titanium Oxide (TiO<sub>2</sub>)

This is the strong oxidant and its application in the treatment of various inorganic and organic pollutants is well established.

#### 3. Acid or Alkali

H<sub>2</sub>SO<sub>4</sub> acid or NaOH alkali to be used for Ph maintain of waste water. The optimum Value of pH necessary for the Fenton process.

#### 4. Solar Light

Solar light is the oxidizing agent used for the process

#### 5. Zinc Oxide (ZnO)

This is the strong oxidant and its application in the treatment of various inorganic and organic pollutants is well established.

### Chemicals and Reagents

1. ZnO
2. TiO<sub>2</sub>
3. 0.1 N NaOH or H<sub>2</sub>SO<sub>4</sub>
4. Paracetamol
5. Distilled Water

### Experimental Process

#### Experimental Process for various Contact Time

1. Take 0.5 L of 10 ppm pharmaceutical wastewater in beaker and stirred for mixing.
2. Measure the absorbance of known 10 ppm solution or wastewater.
3. Add 1 gm/ L of ZnO/TiO<sub>2</sub> catalyst in beaker.
4. Stirred the solution for 10 min.
5. Keep the beakers of various concentrations solutions in sunlight.
6. For various time of interval 30, 60, 90, 120 and 150-min withdrawal of sample.
7. Check absorbance unit for each sample wastewater help of colorimeter.
8. Calculate % reduction of various for paracetamol by comparing initial and final absorbance unit taken using colorimeter.
11. Follow this procedure for various pH for 10 ppm solution and calculate the % reduction for the paracetamol.

#### Experimental Process for various Concentration

1. Take 0.5 L of 5 ppm pharmaceutical wastewater in reactor and stirred for mixing.
2. Measure the absorbance of known 5 ppm solution or wastewater.
3. Add 1 gm/ L of ZnO/TiO<sub>2</sub> catalyst in beaker.
4. Stirred the solution for 10 min.
5. Follow the same procedure for 10, 15, 20 and 25 ppm synthetic wastewater solution.
6. Measure the absorbance for above various ppm solution.
7. Keep the beakers of various concentrations solutions in sunlight.
8. For contact time of interval 90-min withdrawal of sample.
9. Check absorbance unit for each sample wastewater help of colorimeter.
10. Calculate % reduction of various for paracetamol by comparing initial and final absorbance unit taken using colorimeter.

### Experimental Process for various pH

1. Take 0.5 L of 10 ppm pharmaceutical wastewater in beaker
2. Stirred for mixing.
3. Stirred the solution for 10 min.
4. Measure pH for solution.
5. If pH less than 7 we can increase by adding 0.1 N NaOH solution and pH more than 7 we can reduce by adding 0.1 N H<sub>2</sub>SO<sub>4</sub> solution.
6. Make the various pH solutions 2, 4, 6, 8 and 10.
7. Add 1 gm/ L of ZnO/TiO<sub>2</sub> catalyst in beaker.
8. Keep the beakers of various concentrations solutions in sunlight.
9. For 90 min contact time withdrawal of sample.
10. Check absorbance unit for each sample wastewater help of colorimeter.

$$\% \text{ Reduction} = \frac{(\text{Initial Absorbance} - \text{Final Absorbance})}{\text{Initial Absorbance}} * 100$$

### Effect of Contact Time on Paracetamol Reduction for TiO<sub>2</sub>/Solar

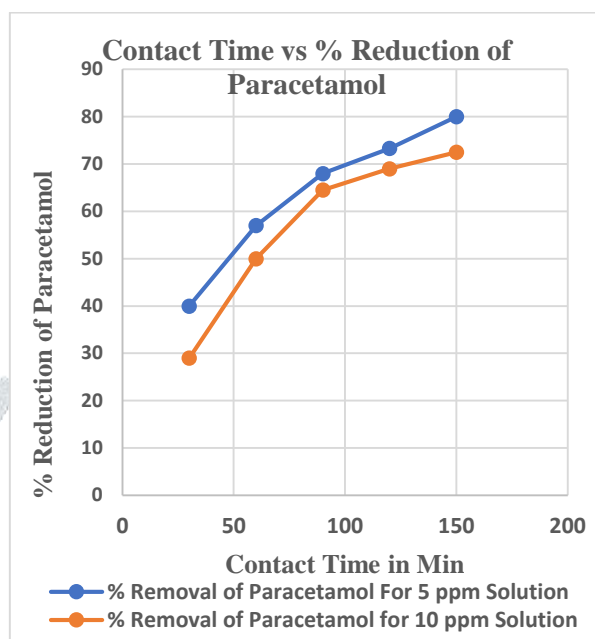
Table shows the effect of contact time on % reduction of paracetamol using TiO<sub>2</sub>/Solar process. The experimental analysis carried out for 5 ppm and 10 ppm solutions. For 5 ppm solution at contact time 30, 60, 90, 120 and 150 mins. The % reduction for paracetamol drugs is 40, 57, 68, 73 and 80 resp. using TiO<sub>2</sub>/Solar process. For 10 ppm solution at contact time 30, 60, 90, 120 and 150 mins. The % reduction for paracetamol drugs is 29, 50, 64, 69 and 72.5 resp. using TiO<sub>2</sub>/Solar process. As per analysis it's clear that the % reduction increase with increase in contact time. The max. reduction shown at 120-150 mins. For 5 and 10 ppm max. reduction at 150 mins. can use as optimum time.

**Table Effect of Contact Time on Paracetamol Reduction Using TiO<sub>2</sub>/Solar**

Contact Time Min	% Removal of Paracetamol For 5 ppm Solution	% Removal of Paracetamol for 10 ppm Solution
30	40	29
60	57	50
90	68	64.5
120	73.3	69
150	80	72.5

### Graphical Representation % Reduction of Paracetamol Using TiO<sub>2</sub>/Solar

Graph shows the effect of contact time on % reduction of paracetamol using TiO<sub>2</sub>/Solar process. For 5 ppm solution at contact time 30, 60, 90, 120 and 150 mins. The % reduction for paracetamol drugs is 40, 57, 68, 73 and 80 resp. using TiO<sub>2</sub>/Solar process. For 10 ppm solution at contact time 30, 60, 90, 120 and 150 mins. The % reduction for paracetamol drugs is 29, 50, 64, 69 and 72.5 resp. using TiO<sub>2</sub>/Solar process. % Reduction increases with increase in contact time. The max. reduction shown at 120-150 mins.



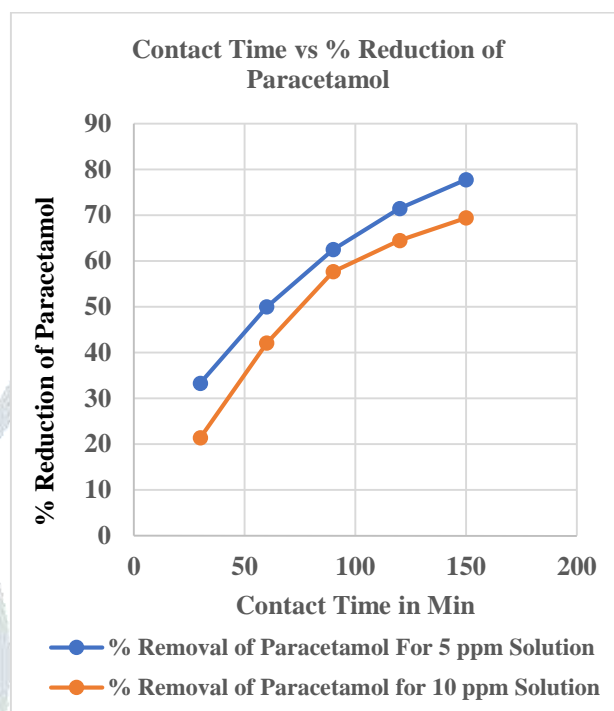
**Fig. Graphical Representation % Reduction of Paracetamol for TiO<sub>2</sub>/Solar**

#### Effect of Contact Time on Paracetamol Reduction Using ZnO/Solar

Table shows the effect of contact time on % reduction of paracetamol using ZnO/Solar process. The experimental analysis carried out for 5 ppm and 10 ppm solutions. For 5 ppm solution at contact time 30, 60, 90, 120 and 150 mins. % Reduction for paracetamol drugs is 33.3, 50, 62.5, 71.5 and 77.7 resp. using ZnO/Solar process. For 10 ppm solution at contact time 30, 60, 90, 120 and 150 mins. The % reduction for paracetamol drugs is 21.42, 42.10, 57.69 and 69.44 resp. using ZnO/Solar process. As per analysis it's clear that the % reduction increase with increase in contact time. The max. reduction shown at 120-150 mins. For 5 and 10 ppm max. reduction at 150 mins. can use as optimum time.

**Table Effect of Contact Time on Paracetamol Reduction Using ZnO/Solar**

Contact Time Min	% Removal of Paracetamol For 5 ppm Solution	% Removal of Paracetamol for 10 ppm Solution
30	33	21
60	50	42
90	61.5	58
120	71.5	64.5
150	78.77	70



**Fig. Graphical Representation of % Paracetamol Reduction for ZnO/Solar**

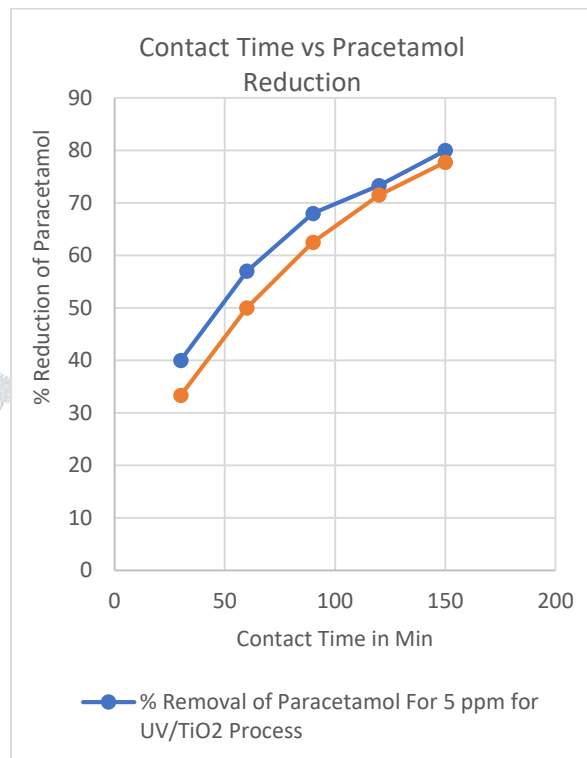
Fig. shows the effect of contact time on % reduction of paracetamol using ZnO/Solar process. The experimental analysis carried out for 5 ppm and 10 ppm solutions. For 5 ppm solution at contact time 30, 60, 90, 120 and 150 mins. The % reduction for paracetamol drugs is 33.3, 50, 62.5, 71.5 and 77.7 resp. using ZnO/Solar process. Graph shows the for 10 ppm solution at contact time 30, 60, 90, 120 and 150 mins. The % reduction for paracetamol drugs is 21, 42, 58, 64.5 and 70 resp. using ZnO/Solar process. As per analysis it's clear that the % reduction increase with increase in contact time. The max. reduction shown at 120-150 mins. For 5 and 10 ppm max. reduction at 150 mins. can use as optimum time.

#### Reduction of Paracetamol for TiO<sub>2</sub>/Solar & ZnO/Solar

**Table Reduction of Paracetamol for TiO<sub>2</sub>/Solar and ZnO/Solar for 5 ppm**

Contact Time Min	% Removal of Paracetamol For 5 ppm for TiO <sub>2</sub> /Solar Process	% Removal of Paracetamol For 5 ppm for ZnO/Solar Process
30	40	33.33
60	57	50
90	68	62.5
120	73.3	71.5
150	80	77.77

Table shows the comparison of two AOP for reduction of paracetamol drugs from synthetic wastewater for 5 ppm at various contact time. For TiO<sub>2</sub>/Solar Process gives more reduction of drugs than the ZnO/Solar process. There is little bit difference in reduction of paracetamol between two processes. Both the processes are effective for paracetamol reduction. As per observation the maximum reduction for paracetamol drugs from synthetic wastewater is 80 and 77.77% at 150 min time with 10 ppm concentration drugs for TiO<sub>2</sub>/Solar and ZnO/Solar process resp



**Fig. Graphical Representation of % Reduction of Paracetamol 5 ppm**

Graph shows the comparison of two AOP for reduction of paracetamol drugs from synthetic wastewater for 5 ppm at various contact time. For  $\text{TiO}_2/\text{Solar}$  Process gives more reduction of drugs than the  $\text{ZnO}/\text{Solar}$  process. There is little bit difference in reduction of paracetamol between two processes. Both the processes are effective for paracetamol reduction.

#### Reduction of Paracetamol for $\text{TiO}_2/\text{Solar}$ & $\text{ZnO}/\text{Solar}$ for 10 ppm

Table shows the comparison of two AOP for reduction of paracetamol drugs from synthetic wastewater for 10 ppm at various contact time. For  $\text{TiO}_2/\text{Solar}$  Process gives more reduction of drugs than the  $\text{ZnO}/\text{Solar}$  process. There is little bit difference in reduction of paracetamol between two processes. Both the processes are effective for paracetamol reduction. As the concentration of pollutant increase the rate of reduction decreases. As per observation the maximum reduction for paracetamol drugs from synthetic wastewater is 72.5 and 69.44% at 150 min time with 10 ppm concentration of drugs for  $\text{TiO}_2/\text{Solar}$  and  $\text{ZnO}/\text{Solar}$  process resp.

**Table % Reduction of Paracetamol for  $\text{TiO}_2/\text{Solar}$  &  $\text{ZnO}/\text{Solar}$  for 10 ppm**

Contact Time Min	% Removal of Paracetamol for 10 ppm for $\text{TiO}_2/\text{Solar}$ Process	% Removal of Paracetamol for 10 ppm $\text{ZnO}/\text{Solar}$ Process
30	29	21.42
60	50	42.10
90	64.5	57.69
120	69	64.5
150	72.5	69.44

### Graphical Representation of % Reduction of Paracetamol 10 ppm

Fig. shows the comparison of two AOP for reduction of paracetamol drugs from synthetic wastewater for 10 ppm at various contact time. For TiO<sub>2</sub>/Solar Process gives more reduction of drugs than the ZnO/Solar process. There is little bit difference in reduction of paracetamol between two processes. Both the processes are effective for paracetamol reduction.

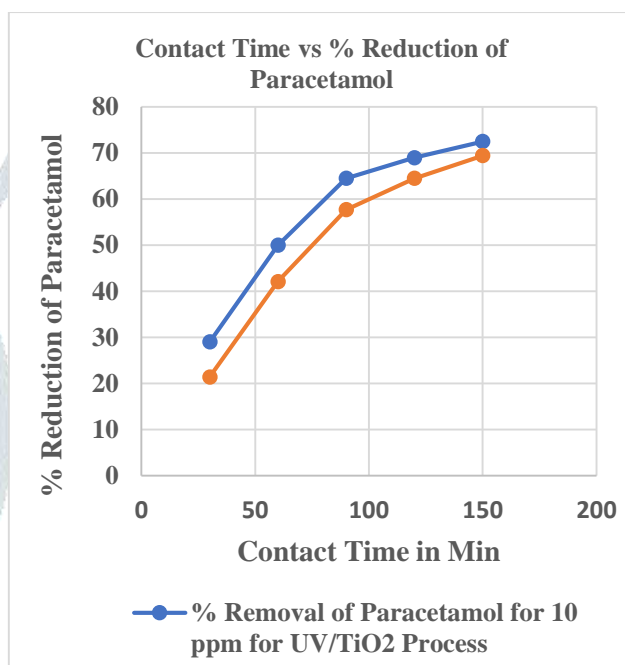


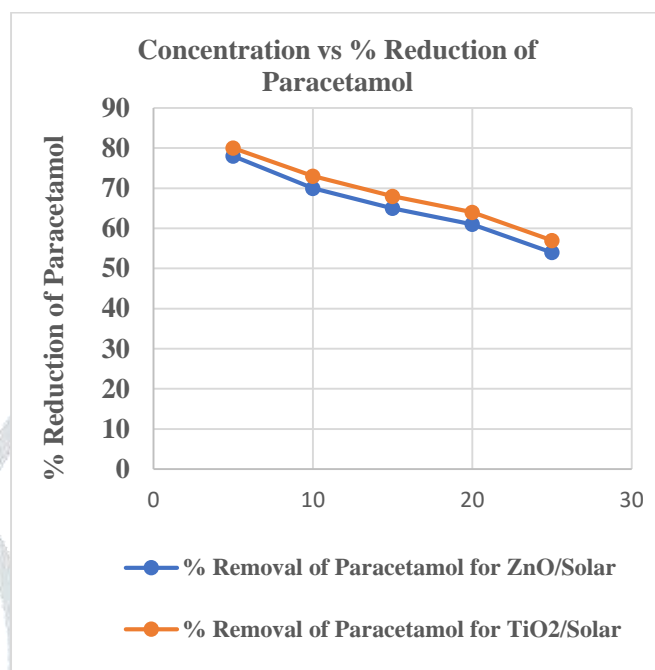
Fig. Graphical Representation of % Reduction of Paracetamol 10 ppm

Effect of Time on Paracetamol Reduction for ZnO/Solar and TiO<sub>2</sub>/Solar at various concentrations

Table Effect of Contact Time on Paracetamol Reduction Using TiO<sub>2</sub>/Solar

Concentration In ppm	% Removal for ZnO/Solar	% Removal for TiO <sub>2</sub> /Solar
5	78	80
10	70	73
15	65	68
20	61	64
25	54	57
30	51	54

Table shows the effect of concentration on % reduction of paracetamol using ZnO/Solar and TiO<sub>2</sub>/Solar process. For concentrations 5, 10, 15, 20, 25 and 30 ppm solutions % reduction of paracetamol using ZnO/Solar process are 78, 70, 65, 61, 54, 51 and using TiO<sub>2</sub>/solar process are 80, 73, 68, 64, 57, 54 resp. for 120-150 contact time. As per observation as the concentration of paracetamol increase, rate of reduction decrease. As per observations both the processes are economical and having same efficiency for paracetamol reduction.



**Fig. Graphical Representation % Reduction of Paracetamol for TiO<sub>2</sub>/Solar**

Graph shows the effect of concentration on % reduction of paracetamol using ZnO/Solar and TiO<sub>2</sub>/Solar process. As per graphical representation, as the concentration of paracetamol increase, rate of reduction decrease. As per observations both the processes are economical and having same efficiency for paracetamol reduction.

### Effect Parameters on Rate of Degradation

#### 1. Effect of Catalyst Weight

The effect of TiO<sub>2</sub> and ZnO catalyst amount on the degradation of paracetamol and procaine solutions at natural pH. Typical weight of TiO<sub>2</sub> and ZnO 1-5 gm/l for 5-50 ppm paracetamol solution. Stability the rate of photocatalytic degradation when amount of catalyst reached to 05 gm/l 25-30 ppm solution.

#### 2. Effect of Initial Concentration Paracetamol Solutions

The effect of initial concentration for paracetamol solutions is important in terms of % reduction. Concentration is studied in the range (5-30 ppm) for both processes TiO<sub>2</sub>/solar and ZnO/solar paracetamol solutions.

#### 3. Effect of pH

pH is an important factor in evaluation of photocatalytic degradation rate because the pH change affects the adsorption quantity of organic pollutants. Photocatalytic degradation rate optimal at pH is 8.0 for paracetamol and 4.0 for procaine in presence of certain conditions. pH effect range (4, 6, 8, 10 and 12) for both paracetamols.

#### 4. Effect of Temperature

The study of temperature effect on photocatalytic degradation rate optimal temperature 55-60 °C for paracetamol with initial concentration 10 mg/l for both processes TiO<sub>2</sub>/solar and ZnO/solar & pH solution 6-8 for paracetamol.

#### 4. Effect of Contact Time

As per analysis it's clear that the % reduction increase with increase in contact time. The max. reduction shown at 120-150 mins. For 5 and 10 ppm max. reduction at 150 mins. can use as optimum time.

### CONCLUSIONS

Both solar light and catalyst are important factors in paracetamol degradation. The initial concentration increases for both drugs lead to reduce degradation rate with foundation of the mentioned weights of ZnO/TiO<sub>2</sub>. Parameters that effect on photocatalytic degradation of pharmaceutical wastewater are initial effluent concentration, catalyst dosage, irradiation time, intensity of solar light and effect of pH. When the amount of Photocatalyst load increased in the wastewater photocatalytic degradation efficiency significantly increase. As concentration of paracetamol in wastewater increase the rate of reduction decrease. The concentration of paracetamol in wastewater be the important parameter that impact on the rate of reduction of paracetamol from the wastewater. The amount of catalyst (ZnO and TiO<sub>2</sub>) use for treatment is fixed which is 1-6 gm/l using solar radiation as light source. As pH of paracetamol in



wastewater increase the rate of reduction also increase. The pH of paracetamol in wastewater be the important parameter that impact on the rate of reduction of paracetamol from the wastewater. For alkaline pH solution the rate of paracetamol reduction using ZnO/solar process will be maximum. The degradation of pharmaceuticals is highly dependent on the operational parameters of the system. Operating parameters such as initial pH, initial concentration of the pollutant, catalyst loading and irradiation time can influence the removal rate of pollutants. The photocatalytic degradation able to removal of pharmaceutical compounds such as ketoprofen, ibuprofen, tetracycline, amoxicillin and naproxen. The three catalysts (Titanium dioxide, zinc oxide and  $\text{TiO}_2/\text{H}_2\text{O}_2$ ) used are effective catalysts in photocatalytic degradation of real pharmaceutical wastewater. Antibiotics and vitamins are produced by fermentation of fairly complex nutrient solutions of organic matter and inorganic salts by fungi or bacteria. Photocatalytic degradation might be facilitated by using semiconductor  $\text{TiO}_2$  and ZnO which is act to increase the surface area of paracetamol solution to exposed solar light which resulted increase photocatalytic degradation efficiency. Paracetamol and tetracycline are well known with tremendous annual worldwide production and high global consumption rate.

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